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The fabrication and testing of a self-sensing MWCNT nanocomposite sensor for oil leak detection

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Abstract

Oil spillage, due to either direct or indirect accidents, can cause major environmental and economic issues if not detected and remedied immediately. In this study, the unique properties of carbon nanotubes have shown a substantial sensing capability for such a purpose when incorporated into a nanostructured composite material. A high-efficiency self-sensing nanocomposite sensor was fabricated by inserting highly conductive multi-walled carbon nanotubes (MWCNTs) into an elastomeric polymer substrate. The microstructure of the nanocomposite sensor was studied using scanning electronic microscopy and Raman spectroscopy. The response rate of the sensor was evaluated against different MWCNT concentrations, geometrical thickness and applied strains (causing by stretching). The results indicated that the response rate of the sensor (β) decreased with increasing MWCNT concentration and showed the strongest response when the sensor contained a 1.0 wt % concentration of MWCNTs. Additionally, it was found that the response time of the self-sensing nanocomposite sensors decreased in keeping with decreases in the sensor thickness. Moreover, when the sensor was subjected to strain, while immersed in an oil bath, it was found that the response rate (β) of the unstretched self-sensing nanocomposite sensor was significantly lower than that of the stretched one. The sensors given a 3% applied strain presented a response rate (β) $\approx$ 7.91 times higher than of the unstretched one. The self-sensing nanocomposite sensor described here shows good potential to be employed for oil leakage detection purposes due to its effective self-damage sensing capability and high sensing efficiency and low power consumption.

Keywords: piezoresistive properties; MWCNTs; nanocomposite; expanded elastomer; oil leak; self-sensing

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1 INTRODUCTION

The world today relies heavily on oil as a major source of energy. In particular, the automotive, aerospace and maritime sectors requires various types of oil products to operate [1]. However, accidents involving oil pipelines, tankers and cargo ships can result in the release of large volumes of crude oil. These spillages constitute a potential major source of environmental contamination and damage and so pose a significant hazard to human life. Substantial efforts have been devoted to developing reliable, highly sensitive, low-cost and effective assessment tools that can be used directly in this field [2, 3]. There are numerous commercially available devices that are capable of monitor oil contamination. These devices mainly operate by measuring electromagnetic absorption [4], capacitance [5], image spectroscopic [6] and other properties [7]. However, these expensive devices are usually relatively large in size, and their operation involves considerable power consumption. Recently, polymer-based sensors that use piezoelectric transduction property have been considered as promising analytical tools for detection applications [8, 9]. These innovative detection tools use electrically conductive polymer composites or self-sensing nanocomposites materials

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as potential gas and liquid sensor materials. Researchers studying self-sensing materials are currently focusing on developing new materials based on polymeric matrices that have been modified by incorporating a highly conductive filler material, such as carbon nanotubes (CNTs) or graphene [10]. CNTs are considered to be one of the most promising materials that can be used in these applications [11] due to their superior electrical [12], thermal [13] and mechanical [14] properties. The addition of CNTs as a filler material can also enhance the mechanical properties of the matrix polymer [15] as well as making it electrically conductive [16]. An effective level of electrical conductivity of the nanocomposite material can be achieved even at low filler concentrations [17]. The main driver for this research, viz. using self-sensing nanomaterials for these applications, is their relatively low cost and the ease with which they can be manufactured into a variety of appropriate shapes [18]. Many different sensor designs containing CNTs have been proposed [19]. For instance, the proposed designs include CNTs that are fully embedded in a polymer matrix [20] and while others have CNTs dispersed in a dense network on the surface of the fibres [16]. Many of these studies have incorporated CNTs into different matrix resins. Such combinations include placing CNTs in epoxy [21], MWCNTs in polydimethylsiloxane [22], MWCNTs in phenolic [23], and CNTs in thermoplastic polyurethane [24]. These studies successfully integrated CNTs within the polymer (matrix) in order to fabricate thin films used for heating, coating and for strain sensing applications. In addition, the solution casting of polymer–CNT dispersions (i.e. direct mixing) is considered one of the most promising methods for sensor preparation of thin films [25]. In the context of the large-scale processing of sensory materials, fully embedded CNTs have the most potential (in terms of cost) when compared to other methods such as compression or injection moulding [26]. The use swelling elastomers is also becoming more prevalent in this context [27, 28]. These modern swelling elastomers include natural rubber, butyl rubber, styrene-butadiene rubber, polybutadiene rubbers, polyisoprene rubbers and ethylene propylene diene monomer (EPDM). These materials are highly elastic and swell naturally when exposed to appropriate swelling solvents (media) such as oil or water [29, 30]. Therefore, these elastomers are a very attractive choice as a swelling element to be incorporated into sensors for use in the oil and gas sectors. When CNTs are integrated within such a swelling elastomer, it possible to develop a smart nanocomposite sensor that can monitor various parameters, such as oil spillage and seepage, since volume expansion (swelling) can be directly correlated with sensor electrical conductivity. If the contact solvent is able to diffuse into the matrix of the nanocomposite (i.e. the sensor), the volume expansion of the sensor induces an increase in distance between adjacent filler particles [31]. This increase will reduce the tunnelling resistance, and, therefore, the electrical conductivity of the sensor decreases as swelling proceeds. This occurs since the separation distance between individual CNTs particle increases in the developed matrix network [32]. This can occur when the critical tunnelling distance [17] exceeds 1.8 nm [32]. In this paper, we introduce a self-sensing nanocomposite based on a swelling elastomer matrix material, namely EPDM filled with CNTs. The capability of this material to sense an oil spillage will be examined. EPDM is an elastomer that experiences a good degree of swelling easily when exposed to oil. This is due to its high isoprene content. Moreover, EPDM rubber is also low cost, easy to use and is widely used in many industrial applications [33]. The principle on which this sensor works is its ability to alter the electrical resistance (R) of the nanocomposite when it is exposed to a solvent, which in this study is oil. A simple method was developed to investigate the response rate of the sensor when it was stretched and so exposed to an applied strain. The results are reported in the context of a simulated real application where the sensor response rate is likely to be influenced by the filler content (concentration) and sensor material thickness. In addition, the changes in the electrical conductivity of the sensor under an applied strain (in tension) and the effect of the sensor response rate to this were also studied in detail.

2 EXPERIMENTAL DETAILS

2.1 Sensor materials

Multi-walled carbon nanotubes (MWCNTs) prepared by the chemical vapour depositing process were used as the filler material. These were supplied by US-Research Nanomaterials Inc., USA. The mean diameter, average length and purity of MWCNTs were 40 nm, 35 μm and ∼97%, respectively. The solvent used during samples preparing was high purity of acetone more than 95% purchased from Acros Organics Ltd. (Loughborough, UK). The EPDM rubber sensor matrix material was supplied in liquid form by EPDM Coatings LLC, Shelton, USA. The EPDM pot life was 4–10 h at room temperature, and once constructed, the sensor needed ∼5–7 days to reach a final fully cured state. The response of the prepared sensor was tested after immersion it in a bath containing a light commercial motor oil (Mobil Super High Mileage 5 W-30, USA).

2.2 Sensor fabrication

To start the fabrications process, different concentrations of MWCNTs, ranging from 0 to 3 wt %, were mixed with 50 mL of acetone in a clear beaker. A high frequency horn-type sonicator (BR-21MT-11 L, 1000 Watt) was used to ensure full dispersion of the MWCNTs in the acetone. The sonication process was used to that no agglomeration, caused by electrostatic attraction, of the MWCNTs occurred. Once a good dispersion was achieved, an appropriate amount of EPDM rubber was added to the mixture, which was again sonified for further 10 min to maintain a good dsipersion. To evaporate the acetone from the mixture, the mixture was placed in a vacuum oven at 60°C for 24 h. Once the acetone was fully evaporated, the hardening agent was added with a mix ratio of 21:1. This mixture was then degased in a vacuum chamber for a further 15 min to remove any air bubbles within the mixture. The produced mixture was then cast in a non-stick teflon coated mould to produce the thin-film sensor configuration needed for testing. Finally, the fabricated sensors

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left to dry overnight in a vacuum oven at 60°C and then left to dry at room temperature for a further 6 days to ensure the full completion of the curing process.

2.3 Sensor characterisation

To determine the electrical resistance of the nanocomposite sensors, before and after being placed in the oil bath, a DC digital multimeter (Keithley 2100) was employed. This used a two-point probe technique to measure the resistance of each fabricated nanocomposite sensor with a wide range of voltages. The dimensions of the nanocomposite sensor dimensions were 30 mm (length) × 15 mm (width) × 5 mm (thickness). All the measurements had the same condition and collected a stabilised value from the device. The electrical conductivity (σ) of the nanocomposite sensor was calculated using Equation (1):

$$\sigma = \frac{L}{RA} \tag{1}$$

In Equation (1) L, R and A are the length (m), the electric resistance (Ω) and the cross-section area (m^2) of the nanocomposite sensor, respectively. High purity conductive glue and copper tape were used on both ends of the sensor to ensure that a good electrical contact with the nanocomposite sensor was maintained. For the strain tests, a bespoke laboratory stretching device was built and employed to stretch the sensors. The test sample dimensions were described previously. Throughout each strain test, the normalised electrical resistance (R/R_0) was continuously monitored and recorded. During testing, R is the changing resistance of the sensor, and R_0 is the sensor's initial starting resistance. Scanning electronic microscopy (SEM type JEOL JSM-7001F, Japan) was employed to examine the MWCNT distribution inside the matrix of the nanocomposite sensor. To enhance the SEM image quality, the surface of the nanocomposite sensors was first coated with a thin gold layer. X-ray diffraction (XRD) patterns for the nanocomposite sensor containing different concentrations of MWCNTs (0–3 wt % of MWCNTs) were also obtained. The data collected in this way were recorded on a Bruker D8 Advanced Diffractometer with Cu Kα radiation at a generator voltage of 40 kv and current 40 mA over the 2θ angle range of 5–60°C at ambient temperature.

3 RESULTS AND DISCUSSION

3.1 Electrical conductivity of self-sensing nanocomposite sensors

Figure 1 shows the electrical conductivity of the self-sensing nanocomposite sensor containing MWCNTs in the concentration range 0–3 wt %. The unmodified EPDM (0 wt %) is an insulator with a conductivity of $\approx 10^{-16}$ S/cm. As can be seen in Figure 1, an addition of MWCNTs <0.75 wt % initially has only a very small effect on the electrical conductivity of the self-sensing nanocomposite sensors. However, an increase in the MWCNT content beyond this level leads to the remarkable increase of the electrical conductivity of the self-sensing nanocomposite sensors. For a 1.0 wt % of MWCNTs, the electrical conductivity increased by over 12 orders up to 4.88×10^{-4} S/cm. The addition of more MWCNT will lead to further electrical conductivity increases in the sensor material. The self-sensing nanocomposite sensors filled with 3.0 wt % MWCNTs had electrical conductivities which approached 5.11×10^{-2} S/cm. This was due to a high concentration of MWCNTs embedded in the polymers, allowing for the formation of effective conductive network pathways [19]. The percolation effect has been experimentally observed [34] to play a role in the dependence of electrical conductivity with increasing MWCNTs content. The percolation effect provides a significant change in electrical conductivity of the composite material due to the spatially closer MWCNT filler concentrations. The electrical behaviour, beyond the percolation threshold, of composite materials can be described by the well-known power law developed by Kirkpatrick [35]. Here the percolation threshold was calculated as 0.41 wt % MWCNTs.

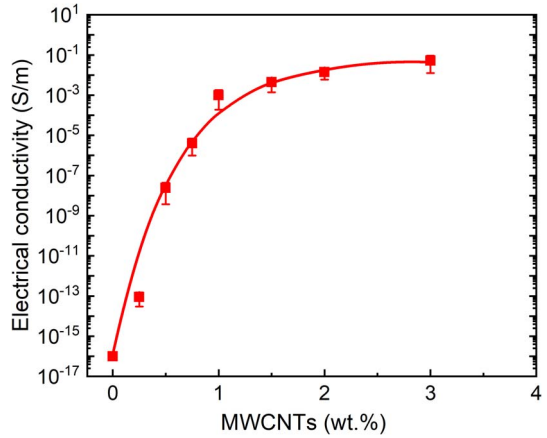

Figure 1. *Electrical conductivity of self-sensing nanocomposite sensor with different MWCNTs concentrations.*

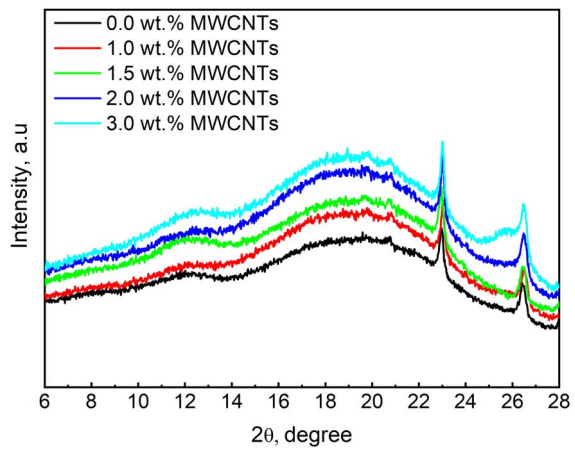

Figure 2. *X-ray diffraction profile for the unmodified EPDM and the EPDM nanocomposite sensors containing different MWCNTs concentrations.*

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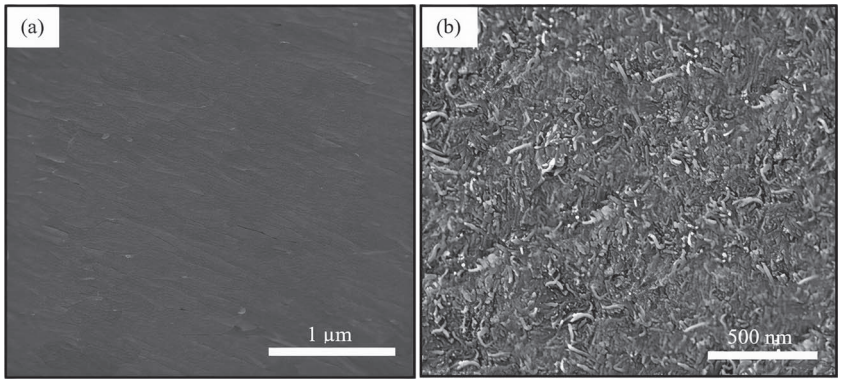

Figure 3. *SEM images of (a) unmodified EPDM (b) the self-sensing nanocomposite sensor containing 3.0 wt.% MWCNTs.*

SEM and XRD techniques were employed to qualitatively assess the dispersion of MWCNTs in the EPDM. Figure 2 shows the XRD profiles for both the unmodified EPDM matrix and that modified with different concentrations of MWCNTs. From Figure 2, it can be clearly seen that the self-sensing nanocomposite sensors, including the pure EPDM sample, show two distinct peaks. The first peak, which has quite a narrow (sharp) peak profile, is located at θ = 23°, whereas the second is located at θ = 26.25° and has a broader peak profile.

It should be noted that the diffraction peak intensities in the self-sensing nanocomposite sensors increase with increasing MWCNT concentration. This observation is due to the ready availability and good dispersion of MWCNTs within the sensor matrix as shown in Figure 3b. Although a percolating network is formed above only~0.41 wt % of MWCNTs, the conductivity value below 1.0 wt % MWCNTs is too low to be effective for sensing purposes. Therefore, self-sensing nanocomposite sensors with MWCNT contents less than 1.0 wt % of MWCNTs were not used during subsequent experiments.

3.2 The effect of MWCNT concentration on sensor response

Figure 4 demonstrates the normalised electric resistance (R/R_0) versus the time (t) when the self-sensing nanocomposite sensor was exposed to oil. Here R is the electric resistance at time t, and R_0 is the initial electric resistance at the start of exposure, i.e. when t = 0. It can be seen that the values of normalised resistance (R/R_0) increased gradually with time and showed the strongest response when the self-sensing nanocomposite sensor contained 1.0 wt % of MWCNTs. This response is possibly due to the sensor swelling, so causing an increase in the gaps (space) between the CNTs. This can explain the observed increase in the normalised resistance (R/R_0) and is in keeping with the tunnelling mechanism theory proposed by Bao *et al.* [36]. Moreover, as shown in Figure 4b, when the filler concentration increases, then there is reduction in the response rate, $\frac{d(R/R_0)}{dt}$,of the self-sensing nanocomposite sensor. In other words, the change in the electric resistance over the selected time interval (Δt) decreases with increasing MWCNT concentration. The data value dependence of the normalised electric resistance (R/R_0) against time can be

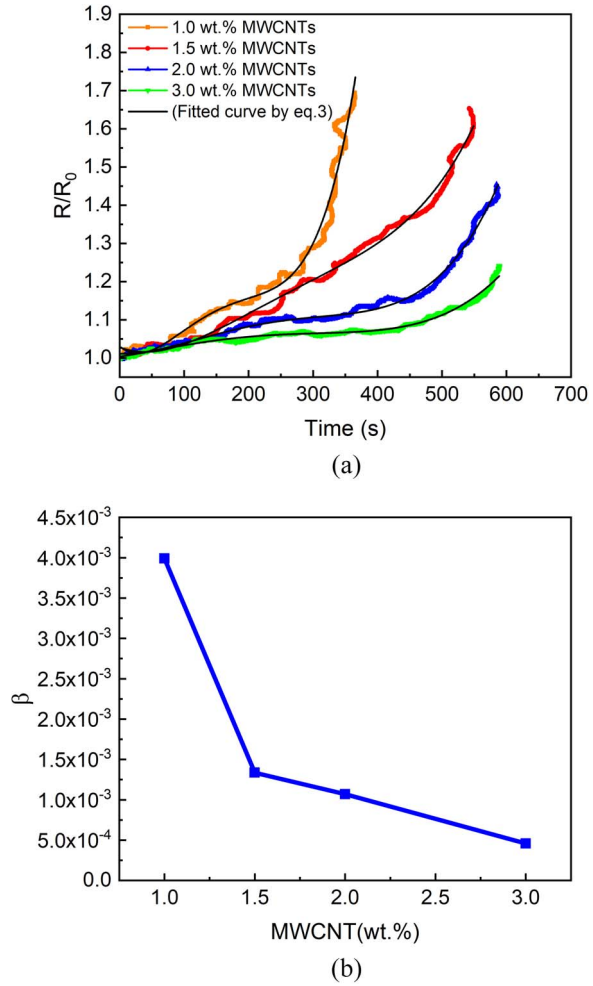

Figure 4. *(a) The normalised electrical resistance (R/R0) versus time during oil absorption at different MWCNTs concentrations and (b) response rate of the sensor.*

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Table 1. *The parameters A_1, A_2, A_3 and A_4 of the investigated self-sensing nanocomposite sensors that contain different MWCNT concentrations.*

Sample	A_1	A_2	A_3	A_4
1.0 wt % MWCNTs	$-1.46 \times 10^{-3} \pm 4.08 \times 10^{-4}$	$3.26 \times 10^{-5} \pm 3.96 \times 10^{-6}$	$-1.67 \times 10^{-7} \pm 1.47 \times 10^{-8}$	$2.83 \times 10^{-10} \pm 1.85 \times 10^{-11}$
1.5 wt % MWCNTs	$-6.51 \times 10^{-4} \pm 6.58 \times 10^{-5}$	$9.67 \times 10^{-6} \pm 4.63 \times 10^{-7}$	$-2.63 \times 10^{-8} \pm 1.21 \times 10^{-9}$	$2.62 \times 10^{-11} \pm 1.05 \times 10^{-12}$
2.0 wt % MWCNTs	$-5.35 \times 10^{-5} \pm 3.59 \times 10^{-5}$	$4.98 \times 10^{-6} \pm 2.44 \times 10^{-7}$	$-1.91 \times 10^{-8} \pm 6.1 \times 10^{-10}$	$2.21 \times 10^{-11} \pm 5.02 \times 10^{-13}$
3.0 wt % MWCNTs	$2.69 \times 10^{-4} \pm 2.49 \times 10^{-5}$	$7.94 \times 10^{-7} \pm 1.68 \times 10^{-7}$	$-5.74 \times 10^{-9} \pm 4.19 \times 10^{-10}$	$7.91 \times 10^{-12} \pm 3.44 \times 10^{-13}$

fitted (modelled) using the following function:

$$\frac{R}{R_0} = 1 + (A_1 \times t) + (A_2 \times t^2) + (A_3 \times t^3) + (A_4 \times t^4) \quad (2)$$

The parameters A_1, A_2, A_3 and A_4 of the investigated self-sensing nanocomposite sensors are summarised in Table 1. This function was used here because it represents the most appropriate fit to the experimental data. In this way, the response rate (indicated by the response parameter β) of the self-sensing nanocomposite sensor can be effectively characterised in a quantitative manner by the derivative of the normalised electric resistance (R/R_0) with respect to time:

$$\beta = d\,(R/R_o)\,/dt \quad (3)$$

It can be seen that the response parameter (β) decreases as the MWCNT concentration increases. This confirms that an increase in the conductive additive filler concentration leads to a reduction in the response rate of the self-sensing nanocomposite sensors when exposed to oil for different time periods.

3.3 The effect of sensor thickness on response rate

The influence of the sensor thickness on the normalised electric resistance (R/R_0) during time (t) when it exposed to oil is shown in Figure 5. The self-sensing nanocomposite sensors filled with 3.0 wt % of the MWCNTs were chosen for further testing since this concentration level showed more stable behaviour (less normalised resistance 'noise') as seen in Figure 4a. Generally, the results obtained show that the response time of the self-sensing nanocomposite sensors decreases with the decrease in the thickness of the sensor. For example, a 30% increase in the resistance for the thickest sensor prepared (5.0 mm) was reached in 600 s, while the same increase was reached in 450 s for a 3.0 mm thick sensor and in 320 s for a 1.0 mm thick sensor. This was due to the actual MWCNT concentration (3.0 wt %) that dominated the behaviour of the sensor. After fitting the experimental data with Equation (2), the response rate (β) was characterised in terms of Equation (3), and the obtained values are demonstrated in Figure 5b.

3.4 The effect of sensor strain on response rate

As demonstrated in this study, and previous studies [10, 37], self-sensing nanocomposite materials containing large concentrations of conductive filler will exhibit very good electrical conductivity.

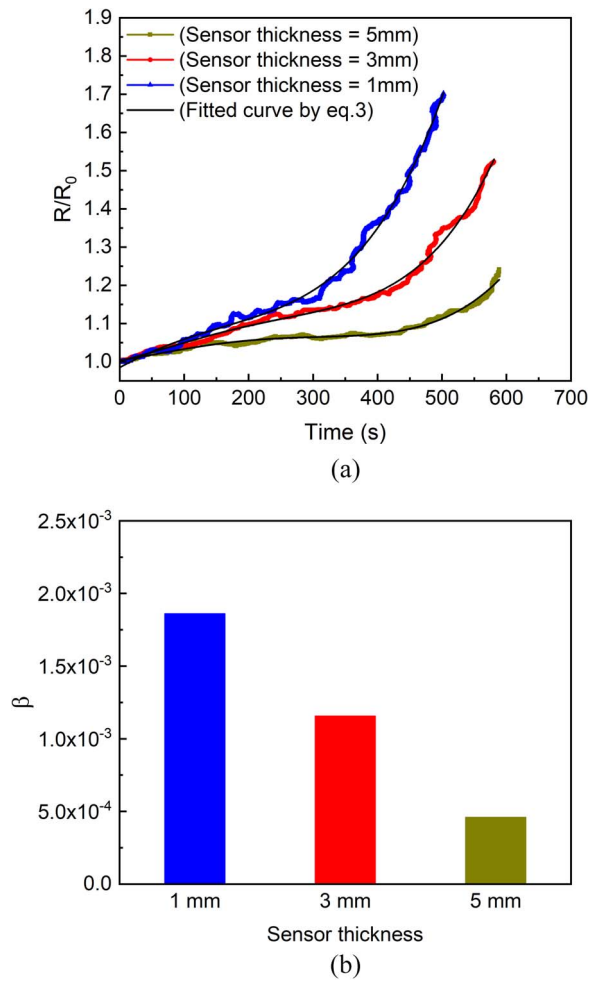

Figure 5. *(a) The normalised electrical resistance (R/R0) versus time during oil absorption for different self-sensing nanocomposite sensor thicknesses and (b) response rate of the sensor.*

This allows for the easy measurement of the electrical current flowing in specimen when it has a potential difference applied to it. However, the permeation of the oil into the swelling sample materials will be difficult so leading to longer response times [30]. To address this issue (i.e. to enhance the sensitivity of the self-sensing nanocomposite sensor without making any additional changes to the sensor), it is proposed to stretch (strain) the self-sensing nanocomposite sensor. The rationale employed here is that stretching the sensor will produce a small deformation that will induce changes in the sensor that can be monitored [38]. For

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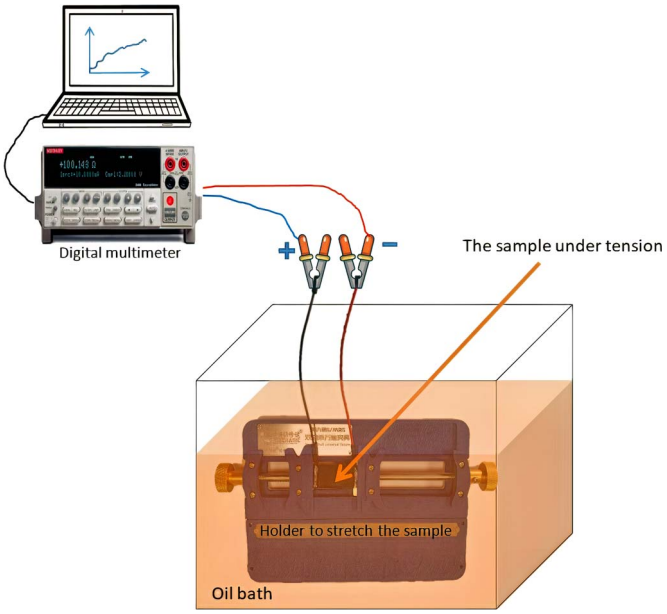

Figure 6. *Photograph of the experimental set-up used to measure the resistance response of the self-sensing nanocomposite sensor when subjected to stretching whilst submerged in oil.*

the case where the adhesion between the filler and the matrix material is low, then voids and micro-cracks will be created (by the stretch) at the interface zone between the matrix and filler [16]. This is unfavourable since the mechanical properties of the nanocomposite material will deteriorate. Much attention has been paid to improving this incompatibility situation between filler and matrix material by treating the surface of fillers [39]. However, in this study, the void and micro-crack formation mechanism can actually be exploited to enhance the sensitivity of a self-sensing nanocomposite sensor when used for oil leakage detection purposes. In this context, micro-crack and void formation beside or around the particles (additive fillers) can increase the penetration of the oil into the sample, thus generating an enhanced, and observable, response rate by the self-sensing nanocomposite sensor. In this test, the dependence of the normalised electric resistance (R/R_0) on time during oil penetration (i.e. absorption) into the sample was recorded for samples in the stretched and un-stretched conditions. During the test, all sensors were subjected to a 3% strain, i.e. was stretched by 3% of its initial length (this was calculated by the ruler that is attached to the device), using the test setup shown in Figure 6. Sandpaper was put on the sensor's ends to prevent it from slipping during the tension stage.

From Figure 7, it can be seen that the stretching of the self-sensing nanocomposite sensor led to an instantaneous increase in the resistance. This was due to the MWCNTs orientation within the sensor matrix material, i.e. a change in the sensor geometry caused by the combined tension and swelling effects. Moreover, it can be seen (following the data fitting technique described earlier) that the response rate (β) of the un-stretched self-sensing nanocomposite sensor is significantly lower than that of the stretched one. It should be noted that the value of the

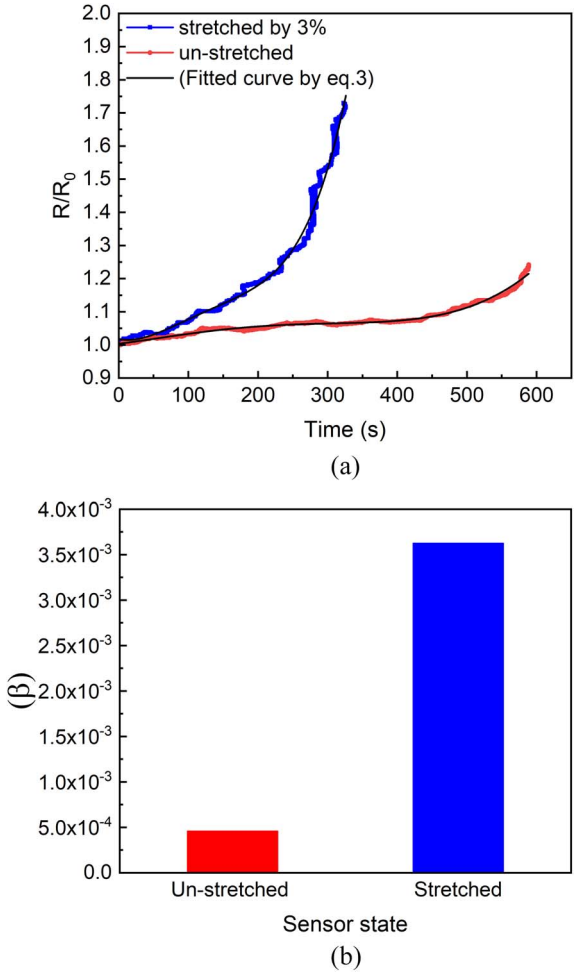

Figure 7. *(a) The normalised electrical resistance (R/R0) against the time during oil absorption for the stretched and un-stretched sensors containing 3.0 wt.% MWCNTs. and (b) response rate of the stretched and un-stretched sensors.*

response rate (β) for stretched sensor was found to be ≈8 times higher than the un-stretched one. This indicates that the change in the electrical resistance of the self-sensing nanocomposite sensor is directly proportional to the oil absorption in the matrix of the sensor.

3.5 Proposed self-sensing nanocomposite sensor behaviour with oil absorption

To understand the self-sensing behaviour of the nanocomposite sensor, when exposed to oil, the schematic shown in Figure 8 is presented. To explain the mechanism operating, the distribution of the MWCNT networks within the matrix material of the sensor must be considered. Since these networks are well integrated within the matrix material, then they will potentially create a large number of conductive pathways to allow easy electron transfer. Subsequently, the matrix of the sensor will change from a completely insulating material (without MWCNTs) to an effective fully conducting nanocomposite. Prior to the self-sensing

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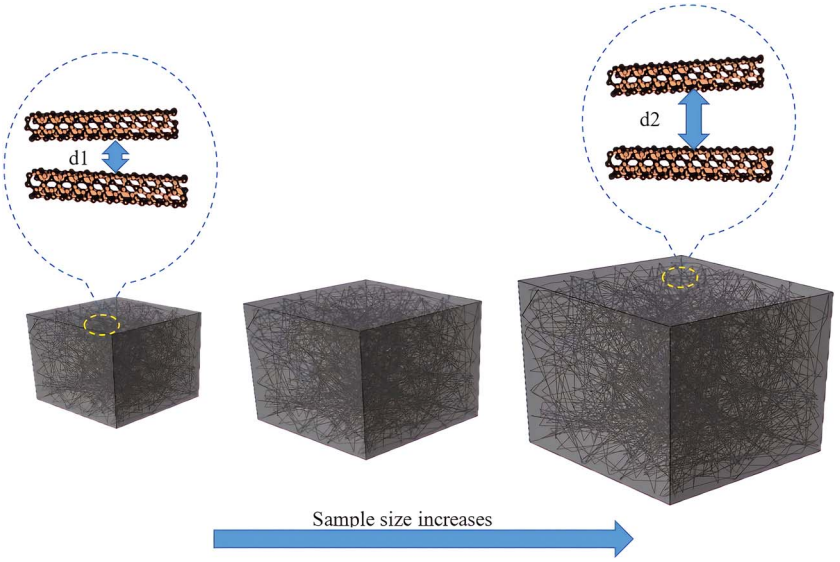

Figure 8. *Diagram showing sample expansion when exposed to oil.*

nanocomposite sensor being exposed to oil, then the positions, and distribution, of the MWCNTs networks were fixed in a relatively stable position with an approximate constant distance (d_1) between neighbouring MWCNTs. After being exposed to the oil, the absorption of the oil will result in the sensor volume increasing (i.e. it will swell). Hence, due to their inherent flexibility, the MWCNT networks will unravel and become stretched out within the matrix of the sensor due to this swelling behaviour. This will lead to a separation of the adjacent MWCNTs conductive pathways, resulting in an increase in the tunnelling distance (d_2) between the MWCNTs. This is in keeping with the theory proposed by Simon [16]. In addition, at the same as this is happening, new MWCNT conductive pathways can be re-established again (i.e. by reconnecting).

Moreover, it should be realised that matrix swelling is a complicated phenomenon and can potentially be affected by polymer swelling and/or relaxation and the extent of oil (liquid) penetration. When the oil diffuses through the polymer mass, it will start to swell gradually, with the potential for a gelatinous viscous (gel) layer to form simultaneously on the surfaces of the MWCNTs. This gel layer and its thickness [40] can significantly affect the sensor conductivity by preventing electrons transferring easily between adjacent MWCNTs. Consequently, the separation in the MWCNTs conductive pathways, and thickness of the gel layer on MWCNTs surfaces, will have the direct effect of causing the gradual increase in the observed normalised resistance (R/R_0) of the self-sensing nanocomposite sensor when it is exposed to oil.

4 CONCLUSIONS

In this study, a MWCNT nanocomposite elastomer sensor was successfully fabricated and its piezoresistive response experimentally studied. This demonstrated its ability, and potential, as a self-sensing nanocomposite sensor for oil leak detection applications. The following conclusions can be drawn from this study.

1. The value of the normalised resistance (R/R_0) increased gradually with increasing time. This behaviour was most noticeable, and effective, when the self-sensing nanocomposite sensor contained 1.0 wt % concentration of MWCNTs.
2. The response time of the self-sensing nanocomposite sensors decreased with the decreases in the thickness of the sensor.
3. The response rate (β) of the stretched self-sensing nanocomposite sensor is significantly higher than that of the unstretched sensor. The value of the response rate (β) was found to be $\approx$8 times higher in the stretched condition.
4. Lower MWCNT concentrations lead to lower levels of electrical conductivity in the nanocomposite material. However, conversely, a low MWCNT concentration increased the response rate (β) of the nanocomposite sensor.

For the future, it is our intention to further develop this preliminary study to include more advanced nanocomposites materials containing CNTs, potentially based on additional types of elastomeric materials and to include the influence of strain rate on the observe phenomena.

CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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SUPPLEMENTARY DATA

Supplementary material is available at *International Journal of Low-Carbon Technologies* online.

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REFERENCES

[1] Devold H. 2013. *Oil and Gas Production Handbook: An Introduction to Oil and Gas Production*. Lulu, Morrisville, NC, USA.
[2] Moroni D, Pieri G, Tampucci M. Environmental decision support systems for monitoring small scale oil spills: existing solutions, best practices and current challenges. *J Mar Sci Eng* 2019;**7**:19.
[3] Baruque B, Corchado E, Mata A, Corchado JM. A forecasting solution to the oil spill problem based on a hybrid intelligent system. *Inf Sci* 2010;**180**:2029–43.
[4] Cheng M, Ju G, Jiang C *et al.* Magnetically directed clean-up of underwater oil spills through a functionally integrated device. *J Mater Chem A* 2013;**1**:13411–6.
[5] Peng S-Y. 2008. *Hasler PE*. Google Patents: System and method for sensing capacitance change of a capacitive sensor.
[6] Clark RN, Swayze GA, Leifer I *et al.* A method for quantitative mapping of thick oil spills using imaging spectroscopy. *US Geological Survey Open-File Report* 2010;**1167**:1–51.
[7] Adegboye MA, Fung W-K, Karnik A. Recent advances in pipeline monitoring and oil leakage detection technologies: principles and approaches. *Sensors* 2019;**19**:2548.
[8] Cichosz S, Masek A, Zaborski M. Polymer-based sensors: a review. *Polym Test* 2018;**67**:342–8.
[9] Al-Bahrani M, Bouaissi A, Cree A. Mechanical and electrical behaviors of self-sensing nanocomposite-based MWCNTs material when subjected to twist shear load. *Mech Adv Mater Struct* 2021;**28**:1488–97.
[10] Al-Bahrani M. 2019. *The Manufacture and Testing of Self-Sensing CNTs Nanocomposites for Damage Detecting Applications*. University of Plymouth, UK.
[11] Akbari E, Buntat Z. Benefits of using carbon nanotubes in fuel cells: a review. *Int J Energy Res* 2017;**41**:92–102.
[12] Huang N, Kirk DW, Thorpe SJ *et al.* Effect of carbon nanotube loadings on supercapacitor characteristics. *Int J Energy Res* 2015;**39**:336–43.
[13] Yang L, Zhang N, Yuan Y *et al.* Thermal performance of stearic acid/carbon nanotube composite phase change materials for energy storage prepared by ball milling. *Int J Energy Res* 2018;**43**:6327–36.
[14] Keru G, Ndungu PG, Nyamori VO. A review on carbon nanotube/polymer composites for organic solar cells. *Int J Energy Res* 2014;**38**:1635–53.
[15] Al-Bahrani M, Cree A. A simple criterion to evaluate the degree of damage in composite materials after sudden impact loads by exploiting the MWCNTs piezoresistive property. *Carbon* 2019;**150**:505–17.
[16] Al-Bahrani M, Gombos ZJ, Cree A. Investigation of the constancy of the MWCNTs on the fibres surface for manufactured self-sensing composites. *Compos Part B* 2019;**173**:106998.
[17] Rahman R, Servati P. Effects of inter-tube distance and alignment on tunnelling resistance and strain sensitivity of nanotube/polymer composite films. *Nanotechnology* 2012;**23**:055703.
[18] Avilés F, Oliva-Avilés AI, Cen-Puc M. Piezoresistivity, strain, and damage self-sensing of polymer composites filled with carbon nanostructures. *Adv Eng Mater* 2018;**20**:1701159.
[19] Al-Bahrani M, Cree A. Micro-scale damage sensing in self-sensing nanocomposite material based CNTs. *Compos Part B* 2021;**205**:108479.
[20] Xiang D, Wang L, Tang Y *et al.* Damage self-sensing behavior of carbon nanofiller reinforced polymer composites with different conductive network structures. *Polymer* 2018;**158**:308–19.
[21] Wang F, Liang W, Wang Z *et al.* Preparation and property investigation of multi-walled carbon nanotube (MWCNT)/epoxy composite films as high-performance electric heating (resistive heating) element. *Express Polym Lett* 2018;**12**:285–95.
[22] Xu W, Allen MG. Deformable strain sensors based on patterned MWCNTs/polydimethylsiloxane composites. *J Polym Sci B Polym Phys* 2013;**51**:1505–12.
[23] Al-Bahrani M, Graham-Jones J, Gombos Z *et al.* High-efficient multifunctional self-heating nanocomposite-based MWCNTs for energy applications. *Int J Energy Res* 2020;**44**:1113–24.
[24] Luo J, Lu H, Zhang Q *et al.* Flexible carbon nanotube/polyurethane electrothermal films. *Carbon* 2016;**110**:343–9.
[25] Obitayo W, Liu T. A review: carbon nanotube-based piezoresistive strain sensors. *J Sens* 2012;**2012**:1–15.
[26] Feng L, Xie N, Zhong J. Carbon nanofibers and their composites: a review of synthesizing, properties and applications. *Materials* 2014;**7**:3919–45.
[27] Wool RP. Self-healing materials: a review. *Soft Matter* 2008;**4**:400–18.
[28] Qamar S, Al-Hiddabi S, Pervez T, Marketz F. Mechanical testing and characterization of a swelling elastomer. *J Elastomers Plast* 2009;**41**:415–31.
[29] Salehi S, Ezeakacha CP, Kwatia G *et al.* Performance verification of elastomer materials in corrosive gas and liquid conditions. *Polym Test* 2019;**75**:48–63.
[30] Qamar SZ, Pervez T, Akhtar M. Performance evaluation of water-swelling and oil-swelling elastomers. *J Elastomers Plast* 2016;**48**:535–45.
[31] Mody R, Gerrard D, Goodson J. Elastomers in the oil field. *Rubber Chem Technol* 2013;**86**:449–69.
[32] Villmow T, Pegel S, John A *et al.* Liquid sensing: smart polymer/CNT composites. *Mater Today* 2011;**14**:340–5.
[33] Dick JS. 2014. *Rubber Technology: Compounding and Testing for Performance*. Carl Hanser, Cincinnati, Ohio, USA (2001).
[34] Bauhofer W, Kovacs JZ. A review and analysis of electrical percolation in carbon nanotube polymer composites. *Compos Sci Technol* 2009;**69**:1486–98.
[35] Deng F, Zheng Q-S. An analytical model of effective electrical conductivity of carbon nanotube composites. *Appl Phys Lett* 2008;**92**:071902.
[36] Bao WS, Meguid SA, Zhu ZH, Weng GJ. Tunneling resistance and its effect on the electrical conductivity of carbon nanotube nanocomposites. *J Appl Phys* 2012;**111**:93726.
[37] Gao L, Chou T-W, Thostenson ET *et al.* Highly conductive polymer composites based on controlled agglomeration of carbon nanotubes. *Carbon* 2010;**48**:2649–51.
[38] Ku-Herrera J, Avilés F, Seidel G. Self-sensing of elastic strain, matrix yielding and plasticity in multiwall carbon nanotube/vinyl ester composites. *Smart Mater Struct* 2013;**22**:085003.
[39] Ma P-C, Siddiqui NA, Marom G, Kim J-K. Dispersion and functionalization of carbon nanotubes for polymer-based nanocomposites: a review. *Compos Part A Appl Sci Manuf* 2010;**41**:1345–67.
[40] Kanoun O, Bouhamed A, Ramalingame R *et al.* Review on conductive polymer/CNTs nanocomposites based flexible and stretchable strain and pressure sensors. *Sensors* 2021;**21**:341.

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