

Development of pressure sensors based on printed electronics for applications in robotics

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Abstract

We report on the preparation and characterization of polydimethylsiloxane/ multi-walled carbon nanotubes (PDMS / MWCNT) nanocomposites for application in pressure sensors. Samples with various concentrations of nanotubes and cross-link density were prepared and characterized in terms of their mechanical, thermal and electrical properties. The MWCNT content varied between 4 and 10%, by weight. In general, we observed an increase of the E-modulus, as assessed by Dynamic Mechanical Analysis, with the increase of the carbon nanotubes contents in DMA tests. The electrical conductivity showed a steep increase with the MWCNT content. In addition, we found a conductivity decrease upon increase of temperature (in the range 20°C-160°C) which we attribute to an expansion of the PDMS matrix and/or an agglomeration of the nanotubes. We found it very difficult to correlate the mechanical and thermal properties with the samples' composition, which we attribute to their heterogeneity. However, a change of the electrical resistance upon a strain deformation was found, indicating that this is a system that could well serve the purpose of obtaining a pressure sensor.

1 Introduction

Flexible electronics is an emerging field with a huge range of applications. It features electronic components and devices that can be wrapped around while retaining their functionality and are of great interest. Automotive, mobile and smart devices, health care and energy are examples of technological areas targeted by flexible electronics. Flexible electronics has advantages such as light-weight, easy fabrication of prototypes, less waste of materials, production of a large area systems by using cheaper techniques (such as screen printing, roll to roll printing, knife coating), when comparing with the lithographic techniques used in the traditional semiconducting industry. In addition, material changes are easier to perform. There is a large number of materials, both small molecules and polymers, with a wide range of properties (such as electrical and optical properties, solubility). Nanocomposites are materials with nanoscale structures aimed to improve the macroscopic properties of the polymers. Due to its high surface area, there is a larger number of possible interactions between the nanofiller and polymeric matrix leading to a greater improvement of the final properties. This being said, polymer nanocomposites are an emerging area with diverse applications in very different fields. Carbon Nanotubes are among the strongest and most resilient materials in nature. Due to its strong sp^2 based sigma bonds between carbon atoms, high tensile strength modules were expected. Indeed MWCNT were reported to withstand 150 GPa while SWCNT go up to 53 GPa. Carbon Nanotubes are very good conductors with electrical conductivities reaching 2×10^7 S/m [1]. However, SWCNT can show either semiconductor or conductor-like behaviour, depending on their chirality and diameter. MWCNT's electrical properties depend on the number of shells and, in bulk samples, on the aspect ratio [2]. MWCNT are preferred for application in electrically conducting systems because there is a large possibility that some of the shells (each being a SWCNT) will be conductor-like. The variation of MWCNT electrical conductivity with temperature was also studied [3] [1]. It was shown that its response to temperature depends on the metallic/semiconducting content. Resistance increases for carbon nanotubes with metallic characteristics while for semi-conducting ones the opposite behaviour is observed, as typical of each class of conductors [1].

Polydimethylsiloxane (PDMS) is a silicon-based organic polymer and one of the most widely used in microfluidic devices and microelectromechanical systems (MEMS). PDMS has also been applied in sensors, electronic components

and biomedical devices. PDMS is easy to process and synthesise, low cost, transparent and biocompatible [4] [5]. Due to its biocompatibility, PDMS has also been used as substrate for cell culture [6] [7]. It should be mentioned that the PDMS used in such applications is not a single polymer but a polymer network resulting from the cross-linking of vinyl-terminated PDMS and a cross-linking agent. Mechanical properties of PDMS Sylgard [®] 184 were widely studied and it was demonstrated that curing temperature and cross-link ratio play a crucial role in the final properties [5].

In the present work, a PDMS/MWCNT composite was developed in order to fabricate a flexible conductor, whose electrical resistance changes its value according to mechanical stimulus. This work is motivated not only by the expected properties but also by previous works with this composite that shows very good results. Various polymer/MWCNT composites have been studied 1% wt of MWCNT reinforcement in polystyrene matrix was shown to be effective in increasing elastic modulus and tensile strength by 42% and 25%, respectively [8]. Other polymers such as epoxy resins [9], PVA [10] and polymethylmethacrylate PMMA were also tested [11]. *Engel et al* developed a capacitive sensor dispersing multi walled carbon nanotubes with diameters of 50-100nm and length of 1-5micrometer in PDMS Sylgard 184 and in polyurethane [12]. It was observed that the percolation threshold is 5 times lower for PDMS than polyurethane-based composites (2% and 10% wt respectively). Percolation threshold can be defined as the concentration at which a sharp increase in electric properties is observed as a result of the formation of a continuous conductive network [13]. In addition, for 2% and 10% composites the sheet resistance is approximately 1000 and 150 ohms/square, respectively. Screen printing of PDMS/MWCNT was proven to be effective in producing flexible electronics [14]. *Khan et al* tested PDMS composites with three carbon nanotube concentrations, 3, 4 and 5%. It was reported a decrease in resistance from 40 kOhm to almost 1 kOhm was reported when concentration increases from 3 to 5% [14]. Another recent and interesting article demonstrated the potential of PDMS/MEP(methyl group-terminated PDMS)/MWCNT for wearable electronics [15]. MEP was used to increase the dispersion of the MWCN in the polymeric matrix. *Kim et al* studied the resistance variation of PDMS/MEP/MWCN when 10.000 cyclic strains were applied. MWCNT concentrations from 0% to 20% were studied and conductivity values of 2.281 ± 0.137 S/cm and 0.003 ± 0.001 S/cm for 20% and 1% respectively were reported. Cyclic strains with resistance measurements showed no significant changes after 10000 cycles for 4%, 8% and 12% samples. These previous studies support our selection of the PDMS/MWCNT composites to fabricate the pressure sensor. We investigate the effect of carbon nanotube loading and cross-linking density on the dynamic mechanical properties of the composites, the effect of carbon nanotube loading on the electrical properties, the variation of the electrical resistance with temperature; the effect of cross-linking density and carbon nanotube loading on the electrical properties, the effect of cyclic stress and temperature on the electrical conductivity of PDMS/MWCNT composites and, finally, a preliminary study of the variation of samples electrical resistance upon deformation. We found that the set of studies was still not enough to assess the role of MWCNT loading and cross-link density on the properties of the composites, being the homogeneity of the samples a critical issue. Further studies will be needed to obtain a more complete description of the system properties.

2 Materials and Methods

Sylgard[®] 184 was supplied by Sigma-Aldrich as a two-part kit consisting of a pre-polymer (Part A) and cross-linking agent (Part B). The recommended polymer/cross-linker mass ratio is 10:1 and curing time varies according to the curing temperature. Vinyl groups in part A react with Si-H groups in part B being catalysed by a Pt compound. The MWCNT used in this project had >90% carbon basis, diameter \times length, 110-170 nanometer \times 5-9 micrometer and were purchased from Sigma-Aldrich.

The chosen procedure for PDMS/MWCNT nanocomposites preparation is the following:

- Dispersion of MWCNT in isopropanol(IPA)(S1) in ultrasonds for 30 minutes. MWCNT/IPA ratio of 1:50 %wt. ;
- Dispersion of PDMS Part A in isopropanol(S2) with magnetic stirrer for 1 hour (PDMS/IPA ratio of 1:2 wt);
- Mixture of S1 with S2, increase temperature to 60°C and agitate with a magnetic stirrer;
- Wait until isopropanol evaporates;
- Cool down the resulting solution, add Part B and mix the solution;
- Pour the "solution" into a mold or spread with a knife on a flat surface;
- Put the samples in a vacuum chamber for 1 hour at room temperature;
- Set furnace temperature to 80°C, insert samples and wait 2 hours(under ambient pressure);

In order to assess the effect of cross-link density and carbon nanotubes content on the mechanical and electrical properties, 12 samples were prepared and characterised, as shown in Table 1.

Table 1: Samples identification according to their cross-linker and MWCNT content

Part A: Part B \ CN weight %	4%	6%	8%	10%
5:1	AB05C4	AB05C6	AB05C8	AB05C10
10:1	AB10C4	AB10C6	AB10C8	AB10C10
15:1	AB15C4	AB15C6	AB15C8	AB15C10

The electrical properties of the PDMS/MWCNT nanocomposites were assessed by a two-contacts and four-contacts probe method. The evaluation of the electrical resistivity of the samples using the four contacts method, involved the thermal deposition of gold stripes (ca. 40-50 nm thick) through a shadow mask in an Edwards Coating System E3. Current was applied with a Keithley 2400 Source Measure Unit and the voltage drop was measured with an Agilent 34401A Multimeter. For the two-contacts method, an Agilent 34401A Multimeter was used. The nanocomposites were also characterised by Dynamic Mechanical Analyses (DMA). DMA is a common technique to evaluate the viscoelastic behaviour of materials. A sinusoidal stress is applied and the sinusoidal strain is measured along with the phase angle. DMA technique allows the analysis of the mechanical response as function of stress frequency, temperature and time. Viscoelastic materials exhibit a combination of elastic and viscous properties. The Dynamic Mechanical Analysis of PDMS/multiwalled carbon nanotubes composites was performed on a Perkin Elmer Dynamic Mechanical Analysis 8000 in tension mode with sinusoidal strain input. The studies were carried out in the temperature range from ca. -120°C to 250°C. Two frequencies, 1Hz and 10Hz, were tested. Heating rate was 3°C/min. DMA allowed the determination of the storage modulus(E'), loss modulus(E'') and $\tan \delta$ of each sample.

3 Results

3.1 Dynamic Mechanical Analyses of PDMS/MWCNT nanocomposites

Dynamic Mechanical Analysis (DMA) was performed on all twelve PDMS/MWCNT samples (identified in Table 1). Figure 1 shows the typical results obtained for the sample AB05C8 at 1Hz and Figure 2 compares this response with that obtained upon stimulation at 10Hz.

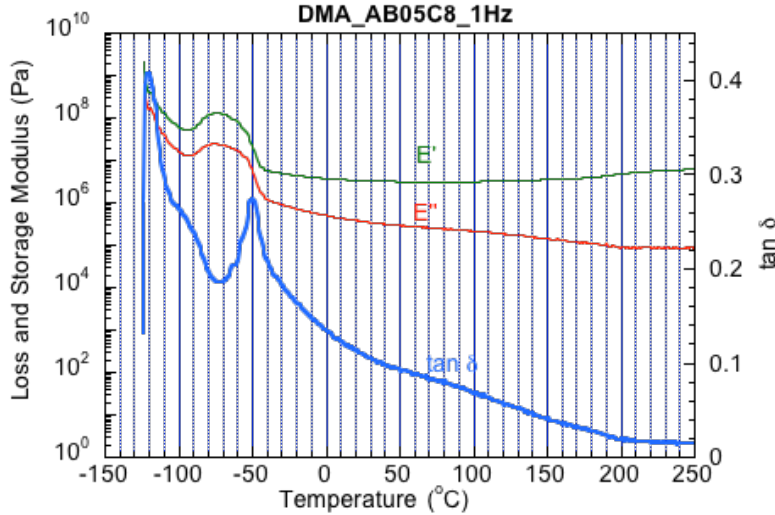


Figure 1: Variation of storage modulus (E'), loss modulus (E'') and loss tangent ($\tan \delta$) with temperature for an AB05C8 sample (8% MWCNT content and 5:1 cross-link content). Measurement at 1Hz

As shown in Figure 1 (and Figure 2) , E' and E'' follow a similar variation with the temperature, nearly stabilizing above ca. -40°C . Two main transitions, associated to peak values of $\tan \delta$, are identified. The first, around -122°C that we associate to the glass transition, and the second one at around -50°C , associated to the melting process. These results show that PDMS forms a semicrystalline polymer network, consistent with previous reports [16]. The loss modulus is lower than the storage modulus across the entire temperature range, evidencing that the elastic component prevails over the viscous component. It should be noted that the determination of T_g , under the present conditions, has a significant error as this temperature is close to the lower limit of the accessible temperature interval.

The stabilization of both E' and E'' after the melting process is a signature of the molecular structure of the material (polymeric network). The feature in both E' and E'' curves below the melting (at ca. -90°C), consisting on a dip, was found in all samples. We consider that the most likely explanation is the existence of a crystallization of the sample, resulting from a two fast cooling process to allow the attainment of thermodynamic equilibrium, that is, without time to reach the proper crystallization. This is a well-known observation in polymers, as, for instance, semicrystalline polymers can be obtained as amorphous materials if they are fast cooled (quenched) from the “melt” to a low temperature, preventing the polymer chains to reorganize from the melt into

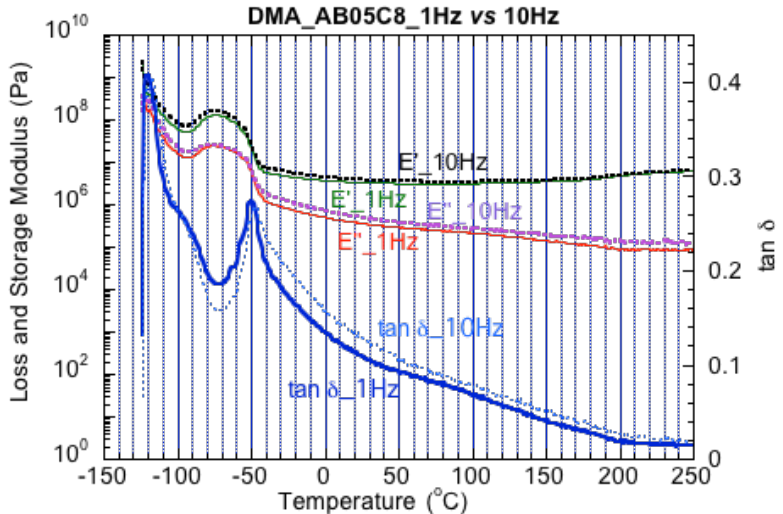
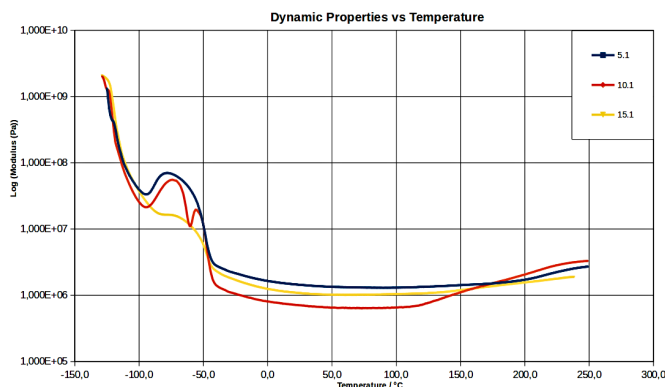


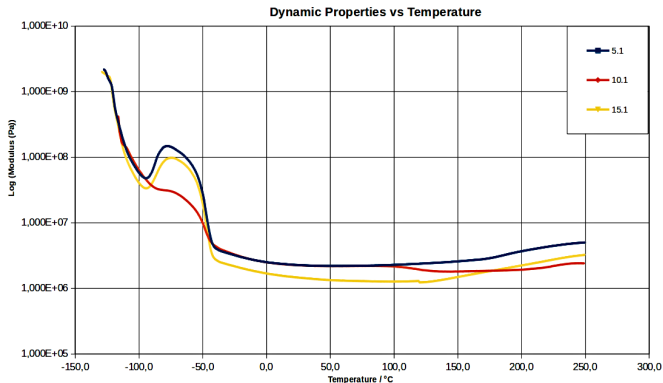
Figure 2: Effect of the DMA test frequency (1 Hz (continuous line) vs 10 Hz (dotted line)) on the loss and storage moduli and loss tangent of sample AB05C8.

an ordered state. Figure 2 compares the DMA results obtained for sample AB05C8 at 1Hz with those obtained at 10Hz. We find that the results at 10Hz, corresponding to shorter relaxation times, lead to a shift of the curves to higher temperature and to slightly higher values of both the storage and loss moduli. The effects are, however, small and, in the remaining of the text, we will refer only to the DMA data obtained at 1Hz.

We find a general trend of increased modulus with increased MWCNT content, as shown in Figure 3. However, we did not find a monotonic increase of the modulus upon increase of the cross-link density. We believe that MWCNT aggregation and the consequent samples' composition heterogeneity are at origin of this behaviour. We note that the MWCNT were not chemically modified. Therefore, we expect that the interaction between the MWCNTs and the PDMS matrix is weak, leading to a small and non-monotonic effect. We would expect that an increase of Part A: Part B ratio, where Part B represents the cross-linking agent (see Table 1) should lead to a decrease of the storage modulus. That is, the AB05 samples (possessing the highest cross-link content) should possess a higher storage modulus, while the AB15 samples should present the lower storage modulus. This prediction is not confirmed as shown in Figure 3, which we attribute to samples inhomogeneity.



(a) Storage modulus for 5:1, 10:1 and 15:1 with 4% CN



(b) Variation of storage modulus with temperature for 6% samples

Figure 3: Variation of storage modulus with temperature for 4% and 6% CN

In addition to this somewhat erratic variation of the storage modulus with both the MWCNT load and the crosslink content, we found that above ca. 100°C an increase of the storage modulus occurs. This is a surprising result, for which we miss, at present, an explanation.

Figure 4 shows the variation of $\tan \delta$ with temperature for all samples. Analysing the position of the highest intensity peak, the one associated to the glass transition temperature, we conclude that it is harder to establish a relationship between either the MWCNT content or the cross-link content and the temperature at which that peak is observed and that we associate to the glass transition temperature (T_g). It is worth pointing out that that such T_g values are close to the temperature at which the temperature scanning started and, therefore, thermal stabilization effects may have a significant impact. Samples heterogeneity, as mentioned above, are likely a contributing factor for such spreading of values. The melting temperatures of all samples, corresponding to the temperature at which the second highest $\tan \delta$ peak occurs, are in range -46.4 °C to -49.5 °C which is a quite narrow range. We could not identify a clear effect of either the MWCNT content or the cross-link content on the melting temperature (T_m).

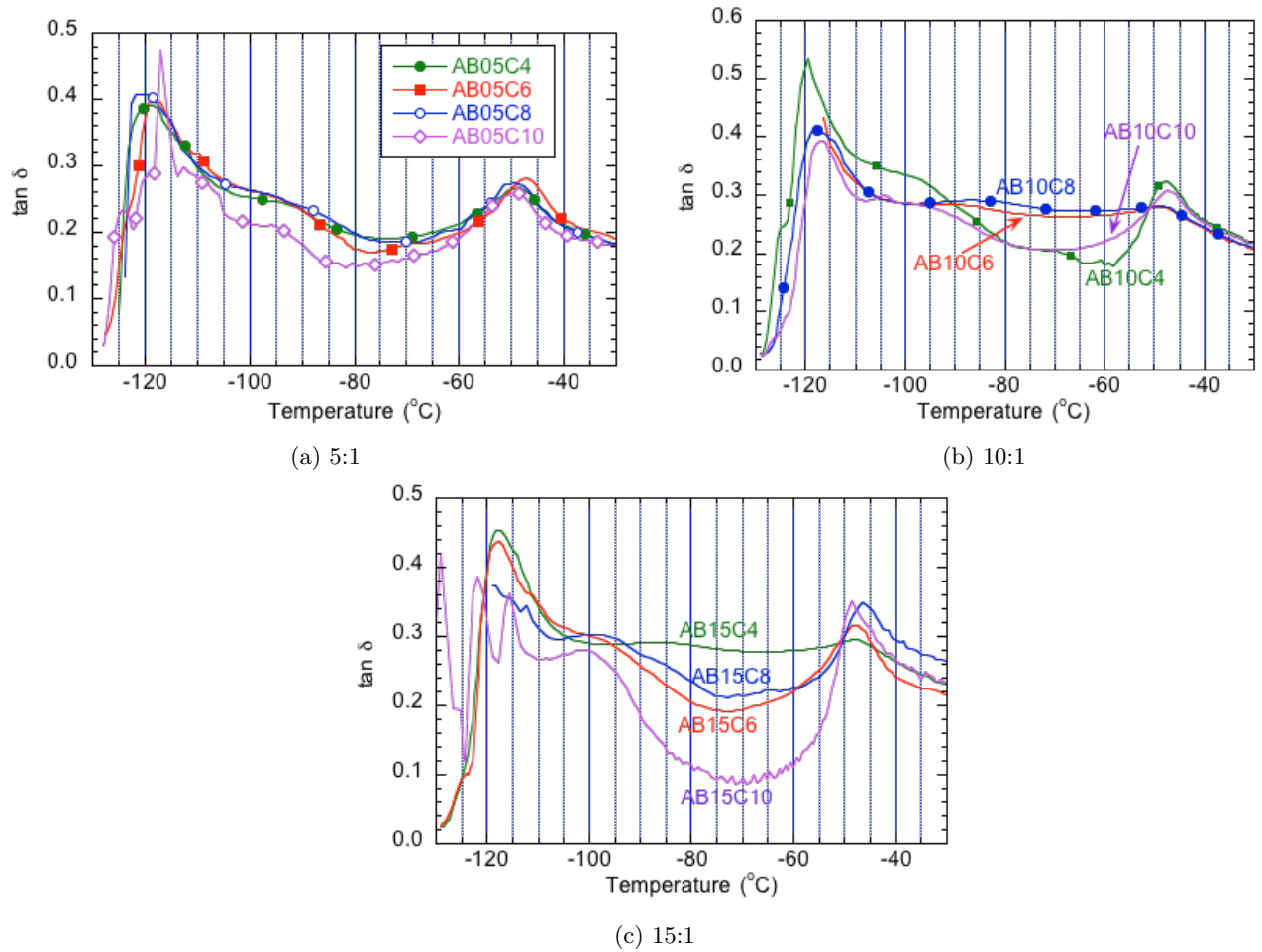


Figure 4: Variation of $\tan\delta$ of the PDMS/MWCNT nanocomposites as a function of the MWCNT content for each cross-link content

In conclusion, both the mechanical properties (namely the storage and loss moduli) and the glass transition temperature present some random dependence on both the cross-link content and the MWCNT load, being this randomness attributed to samples heterogeneity. On the other hand, the melting temperature is approximately the same for all samples, showing a negligible effect of the MWCNT and cross-link content on the crystalline domains.

3.2 Electrical characterization

The electrical conductivity of the four samples containing a 10:1 cross-link content (samples AB10C4, AB10C6, AB10C8 and AB10C10) at room temperature is shown in Figure 5.

The conductivity increases, as expected, upon increase of the MWCNT content. A conductivity of only 8.9×10^{-5} S/m was obtained for the AB10C4 sample, which increases to 4.6×10^{-2} S/m for the AB10C10 sample. The sample with 6% MWCNT content represents a “critical onset”. We anticipate that upon further increase of the MWCNT content the conductivity would further increase. However, for the aimed application, the conductivity values achieved were considered adequate. We believe that, in order to create a system with high resistance sensitivity to deformation, a somewhat incomplete conductive network of MWCNT is preferred. In addition, further increase of the MWCNT content would emphasise even further the aggregation of the MWCNT, leading to highly heterogeneous samples. We also carried out a very simple study of the effect of temperature on the electrical resistance of samples AB10C4 and AB10C8. We used a simple two-contact arrangement to measure directly the resistance of the samples (which will include both the contacts and the sample resistance). As shown in Figure 6, the increase of temperature leads to an increase of the samples’ resistance, being this increase more pronounced for the sample with the lower MWCNT content (sample AB10C4). For a metallic-like system (as would be expected for a compact ensemble of MWCNT), the resistance is in fact expected to increase upon increase of temperature. However, at these low MWCNT content, we expect that the conductivity determining mechanism will be the charge carriers hopping between MWCNTs, a process that is thermally activated. Within this conduction mechanism, the resistance should decrease upon increase of temperature. These are, therefore, two contradicting scenarios. As we consider unlikely that, at these low MWCNT contents, the observed resistance increase upon samples heating simply the reflects the behaviour of a typical metallic-like system, we propose that the expansion of the PDMS matrix could play a major role, as this process would lead to an increased distance between MWCNT that do not touch each other. The fact that the stronger temperature effects occurs in the sample with 4% MWCNT content, below the “critical onset” concentration (of 6%), characterized by an incomplete network of conduction paths, supports this explanation.

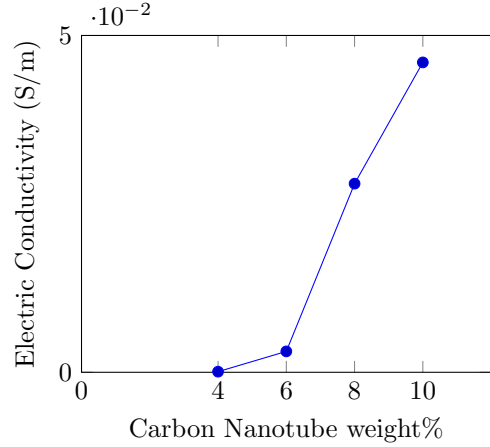


Figure 5: Electrical conductivity of the PDMS/MWCNT nanocomposites as a function of the MWCNT load at room temperature. The line is a guide to the eye.

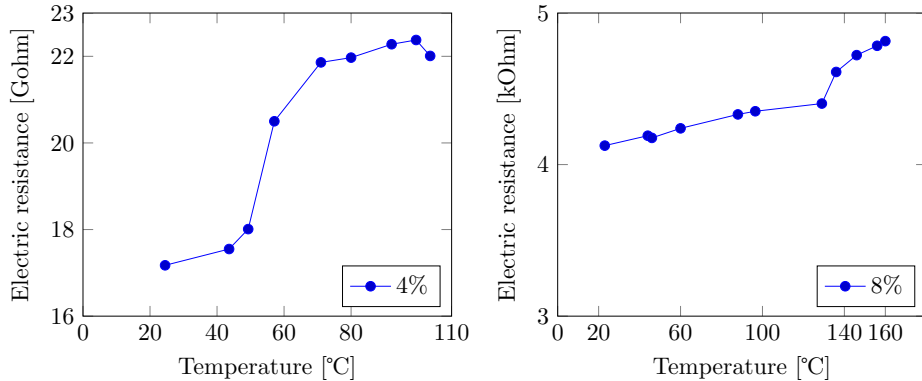


Figure 6: Effect of the temperature on the resistance of samples AB10C4 and AB10C8. The lines are guides to the eye.

Previous studies showed that polymeric composites under tension or compression may suffer reorientation of conductive fillers [17][18]. In order to assess the effect of the DMA studies(cyclic stress/strain and temperature) on PDMS/carbon nanotubes electrical properties of the 12 samples, their electrical resistance was measured before and after Dynamic Thermal Mechanical Analysis, using the two-contact method, where the contacts were made with silver paste. From the resistance values, the electrical conductivity and electrical conductivity variation were calculated. The results shows that there is not a consistent effect of the mechanical and thermal stress impinged on the samples during the DMA investigations on the samples' conductivity. The magnitude of the variation is also very broad. All samples with 6, 8 or 10% MWCNT content undergo a reduction of conductivity, while the samples with 4% show a mixed effect. It is not possible either to conclude about the effect of the cross-link content on the conductivity variation. This observation is likely related to the absence of clear correlation between the Part A:Part B ratio of the samples and the corresponding mechanical properties (as discussed above).

Carbon nanotubes have a strong tendency to aggregate, forming bundles, due to the strong van der Waals interactions between them. It is reasonable to assume that the dynamic mechanical stress and the temperature cycling could facilitate their aggregation, reducing percolation, and leading to a decreased conductivity. Though observed for most samples, this is not observed for all samples. We should also mention that, being the electrical conductivity determined by the two-contacts method, a possible degradation of the interaction between contacts and the samples surface may occur, thereby increasing the contact resistance, and therefore decreasing the calculated samples conductivity. A preliminary assessment of the sample resistance response to deformation was made, with a view to obtain a first validation of the hypothesis that the PDMS/MWCNT nanocomposites can be used as the active sensing element. Therefore, we analysed the variation of the electrical resistance of the AB10 samples with 4, 6, 8 and 10% MWCNT content upon application of strain of 50% (elongation). As shown in Table 3, a strong resistance variation was observed for all samples.

Table 2: Electrical Conductivity variation for all 12 samples

CNT %	Cross-link	Conductivity Variation %
4%	5:1	-14.06×10^3
	10:1	101
	15:1	98
6%	5:1	-234.02
	10:1	-14.90×10^3
	15:1	-26.91
8%	5:1	-25
	10:1	-88.62
	15:1	-689.6
10%	5:1	-20.64×10^3
	10:1	-33.32×10^2
	15:1	-24.17×10^3

Table 3: Variation of the electrical resistance of AB10 samples upon an elongation strain of 50%

CNT%	Electric Resistance Variation(%)
4%	2677.77
6%	1886.31
8%	1100.52
10%	1265.22

We also find that the variation of the electrical resistance upon deformation is lower for the nanocomposite with higher carbon nanotubes loadings. For higher CNT loadings in a polymeric matrix, the number of contacts between carbon nanotubes increases and so does the electrical conductivity. Upon stretching of the material, contacts between carbon nanotubes are broken and the conductive network is destabilized. Due to the higher density of contacts between carbon nanotubes existing on the 10% MWCNT samples, the impact of 50% deformation on electrical properties is less pronounced. Despite the general decrease of the relative resistance variation upon increase of the MWCNT content, we find that such decrease is slightly lower for the sample with 8% than that of the sample with 10% MWCNT. This result is difficult to explain if we assume that we have samples with homogeneous distribution of the MWCNT and that no degradation of the contact resistance during the deformation occurs. These two assumptions are probably (and most likely) not entirely valid, which may explain the result.

4 Conclusions

In this study, 12 PDMS/MWCNT samples were studied in order to evaluate the effects of cross-linking and MWCNT content on the mechanical, thermal and electrical properties. The adequate (macroscopic) dispersion of the MWCNT in PDMS was a challenging task. Yet, we found that isopropanol was a good solvent and the preparation procedure was optimised. The variation of the cross-link degree and MWCNT content did not show a consistent and monotonic effect on the storage and elastic moduli, as assessed by DMA, despite the observation of a moduli increase with the MWCNT content for a fixed cross-link density but no monotonic variation with the cross-linking degree was observed. We believe that these inconclusive results derive from the weak interactions between PDMS and the MWCNT and a heterogeneous dispersion of the MWCNT within the PDMS matrix due to their aggregation. From DMA we also concluded that the nanocomposites have a semicrystalline nature, with the loss tangent showing two maxima, the first associated to the glass transition and a second associated to the melting. The glass transition temperature values are probably affected by a poor samples thermalisation, as no clear variation with the cross-linking degree or the MWCNT content was observed. The electrical conductivity of the tested samples increases, as expected, with an increase of MWCNT loading, with the samples with 6% carbon nanotubes content showing a critical onset of the steep conductivity increase. This behaviour is typical of nanocomposites with a conductivity explained by the formation of percolation paths between the MWCNT. The electrical resistance of samples AB10C4 and AB10C8 was observed to increase with temperature, a variation that, within the percolation regime, with a conductivity controlled by charge hopping, is attributed to the samples' thermal expansion. The thermal and mechanical stress during the DMA studies was also shown to affect samples electrical resistance. However, no clear relation with the cross-linking degree or the MWCNT content could be established. This set of characterisation studies did not allow us to establish conclusions about the effect of the cross-linking degree and MWCNT content on the nanocomposites' properties, which we attribute to the lack of strong PDMS-MWCNT interactions and to the inhomogeneity of the MWCNT dispersion within the PDMS matrix. Finally, the preliminary tests of the variation of the samples resistance with deformation did show that this is a promising system to be used in pressure sensors. Further work is however required to obtain a better description of the system's properties, which is required to optimise the material and, therefore, increase the possibilities of obtaining a properly working pressure sensor, operating within the intended pressure window required for the specific application.

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