Page 1 of 8

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CNT aggregation mechanisms probed by electrical and dielectrical measurements

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Aggregation of carbon nanotubes in silicone oil was studied by electrical and dielectric measurements. The use of a liquid media allowed to slowly dilute and precisely vary the filler contents. With this method, the percolation thresholds were measured with a great accuracy. Filler aggregation process was also probed as a function of time, showing a decrease in the percolation threshold over time after agitation was stopped. In contrast with previous studies on dynamic percolation, in a liquid media a progressive separation between fillers and matrix is evidenced at long time. In addition, no permittivity divergence was measured at the transition, this also results from the liquid nature of the matrix.

1 Introduction

The addition of carbon particles is a common way to improve physical properties of polymer matrices, especially to make lightweight materials with controlled electrical properties[1–5]. Since 1991[6,7], carbon nanotubes (CNT) have shown their great applicability for new technologies. These carbon particles are characterized by unique mechanical[8–11], thermal[12,13] and electrical properties[14–17] and due to their high aspect ratio, a low amount of CNT is necessary to change the electrical properties of an insulating host matrix[18–21].

In the case of a stochastic distribution of CNT in the matrix, which results in a homogeneous system, the electrical conductivity depends on the CNT content and essentially follows the statistic percolation theory[22,23]. For low quantity of filler, CNT are isolated from each other and the electrical properties of composite remain unchanged. Above a critical filler content, called percolation threshold (V_c), a connected path is established between CNT. Consequently, the conductivity of the composite gains several orders of magnitude.

It was recently demonstrated that CNT are not in an equilibrium state just after being dispersed in a matrix. In liquid media, that mimics a molted polymer, CNT tend to aggregate over time. After a certain time, an interconnected cluster is formed within the matrix and similarly to the classic percolation process, an insulating-conductive transition is observed. This phenomena is known as the dynamic per-

colation[24–29]. One of the main interest behind this process is to control the fillers aggregation in order to reduce the amount of CNT necessary to obtain a conductive composite[30].

Dynamic percolation mechanisms have been discussed in the literature. Cipriano showed that polymer chains relaxation plays an important role in the filler aggregation[31]. Skipa[32], Cao[33,34] and Krueckel[35] considered that the dynamic percolation follows a cluster aggregation model. Combessis demonstrated that dynamic percolation is firstly due to macromolecule relaxation followed by a cluster diffusion to a larger scale[30,36]. Most studies were realized in polymer materials, in which long chains have a strong influence on the aggregation process. The use of a liquid matrix instead of a molten polymer appears as an interesting approach to understand the basic dynamic percolation mechanisms: the CNT structuration will not be perturbed by the interactions with long polymeric chains.

To further study and understand dynamic percolation processes, we used insulating liquid matrices. Fillers could indeed be easily and quickly dispersed in liquids with sonication method[37]. Dynamic percolation in liquids at room temperature has been rarely investigated in the literature. Lima[38] studied different carbon fillers in chloroform after sonication and observed an aggregation over time. This was further confirmed by electrical measurements that showed very low percolation thresholds. Similar results were found by Liu in chloroform[39]. Lisetski[40] and Schulz[41] also observed a dynamic percolation respectively in liquid crystals and epoxy before curing. By the means of dielectrical measurements, Poulin investigated dynamic percolation in water filled with CNT[42]. The author demonstrated, by using different surfactant con-

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tents, that percolation threshold is strongly dependent of the attraction between CNT. These results were further explained by the theory of continuum percolation[43].

In this paper, dynamic percolation of carbon nanotubes in silicone oil was studied by means of dc (direct current) conductivity and impedance spectroscopy. Conductivity and permittivity changes were measured over time for different filler contents. The composite were produced by gradual dilution in a stepwise manner, followed by a time dependent electrical measurement for each filler content. This method presents two advantages: the process is fast and easy to carry out; and it is possible to obtain a large amount of data of conductivity *versus* filler content. Consequently, percolation thresholds were determined with an extreme accuracy.

2 Experimental

2.1 Materials and processing

a) A silicone oil from Bluestar Silicones (reference 47-V100), has been used as a matrix. MultiWall carbon nanotubes Nanocyl 7000, with a diameter of 10 nm and a length of 1500 nm, have been used as fillers. 90mg of CNT were dispersed in 30g of silicone oil by an ultrasonic probe at 20 kHz (Fisher Bioblock Scientific 750 watt) during 5 minutes. An iced bath was used to prevent the overheating induced by the ultrasound step. A mother suspension containing 0.048 vol % of carbon nanotube was obtained after the sonication step (this amount was chosen as the upper limit for the studied filler content).

Then, the suspension was diluted with neat solution in order to obtain lower filler contents. CNT distribution was then homogenized by mean of a magnetic stirring at 1000 rotations per minute. Electrical properties were measured over time, both under agitation (induced by the magnetic stirring) and after that agitation was stopped. A hot plate was used to control the temperature (most of the experiments were done at room temperature, except for the last one, which was performed 210°C).

2.2 Electrical characterization

Electrical measurements were performed with two electrodes with an area S of 4.5 cm^2 (width 2 cm, height 2.25 cm) and spaced of 2 mm (e) ($S/e = 0.2m$). The electrodes were settled at the shallow of the CNT suspension in silicone oil and connected to a dielectric spectrometer (Modulab, Solartron Analytical, see Fig. 1). For each filler content, the electrical measurements were carried over 700 seconds in successive cycles of 17 seconds constituted by a D.C. step (Direct Current) of 4 seconds followed by a A.C. step (Alternating Current) of 13 seconds (in other words one DC value and one AC value

are collected every 17 seconds). The D.C. step consists of a voltage sweep from 0 to 400 mV at the rate of 100 mV/sec. The A.C. step is a 13 seconds frequency sweep from 10^5 Hz to 1 Hz with a voltage peak at 400 mV. As the accuracy of collected data is dependent of length of the electrical measurement, 17 seconds per cycle has found to be a good compromise for a large number of experimental data with significant accuracy. After these characterization cycles, the CNT suspension was diluted in order to obtain a lower CNT percentage, and an other characterization step was performed.
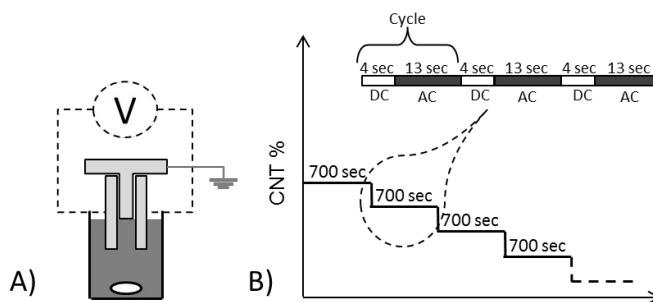

Fig. 1 A-Scheme of the electrodes immersed in silicon oil and connected to the dielectric spectrometer. B- Scheme of the whole experimental process. Successive steps of electrical measurements, dilutions and agitations are performed over time. Each step of electrical measurement is constituted of successive cycle of DC (4 seconds) and AC (13 seconds) over 700 seconds.

3 Results

3.1 DC conductivity after 17 sec

A silicone oil suspension filled with 0.048 vol% CNT was first prepared by sonication. The subsequent compositions were obtained by successive dilution with silicone oil, as described in the previous section. Fig. 2 shows the conductivity *versus* weight fraction of nanotube measured 17 seconds after agitation induced by the magnetic stirring was stopped. The conductivity change *versus* filler content follows the percolation theory as a power law relationship is observed. Close to V_c, the percolation transition from insulating to conductive behavior is observed: the conductivity increases by six orders of magnitude.

Kirkpatrick's equation[23] describes the conductivity variations *versus* filler content as:

$$\sigma = \sigma_0 . (V - V_c)^{\mu} \tag{1}$$

With σ_0 the theoretical macroscopic conductivity of fillers, V_c the percolation threshold, V the filler content and μ the critical exponent, usually close to 2 for 3D system.
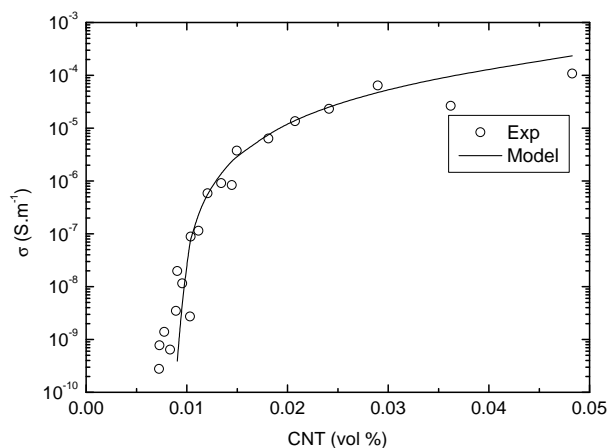
Fig. 2 Electrical conductivity *versus* filler content 17 sec after stopping the agitation. Experimental results (scatter) are fitted with Kirkpatrick's model (line).

Experimental data were adjusted to the percolation model of Kirkpatrick. A good agreement was observed. The value of percolation threshold and critical exponent μ were respectively determined to be 0.0087 ± 0.0005 vol% and 2.32 ± 0.16. The low standard deviation in the vicinity of percolation threshold is due to the large amount of data the dilution technique allowed to collect in the transition zone.

V_c is close to the value determined by Lima[38] for CNT aggregation in chloroform. This seems to indicate that V_c values in liquids are one order of magnitude lower than the ones commonly observed in polymer matrices. This is in a good agreement with the percolation threshold reviewed by Bauhofer *et al* for carbon nanotubes.[22] in different polymers. The authors reported lower V_c in epoxies than in other matrices, probably due to the fact that epoxies have been processed in a liquid state. This low V_c in liquid must be explained by the higher mobility of fillers in low viscosity media. Here we note that 17 seconds after agitation was stopped, critical exponent μ was found close to the universal value, which corresponds to a statistical CNT distribution.[44]

3.2 AC conductivity after 17 sec

Electrical measurements performed in A.C. mode bring further detailed informations on the CNT structuration. The conducting part of the composite is associated to the CNT involved in the percolation path. The capacitive part corresponds to the CNT cluster which are not participating to the transport of electrons. This includes isolated clusters and dead arms. Impedance *versus* frequency is plotted on Fig.3, 17 seconds after the agitation induced by the magnetic stirring was
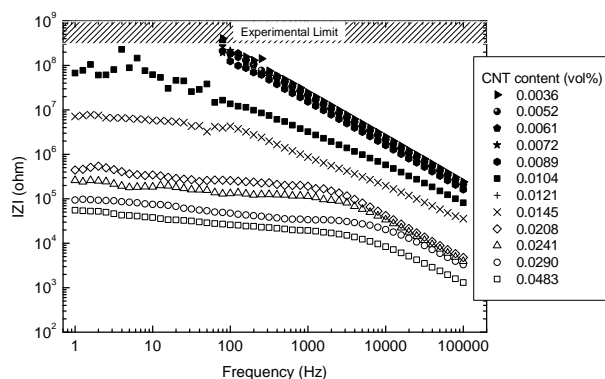
stopped.

Fig. 3 Impedance *versus* frequency for several filler contents. Measurements were performed 17 sec after stopping the agitation.

For filler contents below 0.01 vol% (considered as below V_c), the impedance is frequency dependent and the composite exhibits a capacitive behavior. For higher filler contents, the capacitive behavior is solely observed at high frequencies and a constant impedance related to a resistive behavior is evidenced at lower frequency. The material may thus be viewed as a parallel combination of a capacitor and a resistor. Similar results have been observed close to the percolation threshold in solid material.[45]

3.3 Permittivity measurements after 17 sec

The permittivity ε of a composite is correlated to the number and the size of isolated clusters and dead arms. The following equation describes the theoretical permittivity dependence to the filler content[46,47]:

$$\varepsilon \propto |V - V_c|^{-s} \qquad (2)$$

With s a critical exponent close to 0.7 for 3D systems. For filler contents close to V_c, a divergence of permittivity is expected[47]. According to the theory, below the percolation threshold, the increase of the permittivity is attributed to the growth of CNT clusters. Above Vc, permittivity decrease is correlated to the diminution of the number of isolated clusters and dead arms as clusters are participating to the percolating path[42].

Relative permittivity *versus* filler content is presented in Fig. 4 and adjusted with the Eq. 2 with the following parameters: $V_c = 0.0087$ vol % and $s = 0.7$.

As expected, a the divergence of the permittivity seemed to occur below the threshold as revealed by the sharp increases for $V \sim 0.1$ vol%. Above V_c however, a decrease of permittivity to an asymptotic value was expected. Instead, permittivity

kept on increasing with increased filler content. Similar results have been described in polymers filled with carbon black and carbon nanotubes[47–50].

Two hypothesis may explain these results. The first has been proposed by Pötschke[48] and Lorenz[50]: non-divergence may result from polymer layers between fillers. The lack of direct contact in the filler network creates, according to these authors, microcapacitors that prevail over structural formation of the network. In other words, the percolating network also induces a capacitive effect that is stronger than the loss of capacitance associated with the reduction of the correlation length.

The second hypothesis is based on a different filler organization in liquid. Close to V_c, divergence could be a consequence of the presence of isolated cluster and dead arms near the percolation path. In solid polymer matrices, this is possible because the system is embedded and submitted to a lower mobility.

In a liquid material, CNT clusters present a certain mobility. Without an embedded state of filler and polymer chains, permittivity divergence must be impossible to observe. Then, we assume that above V_c a part of these clusters are not directly integrated to the percolation network and are likely to participate to the permittivity of the composite.
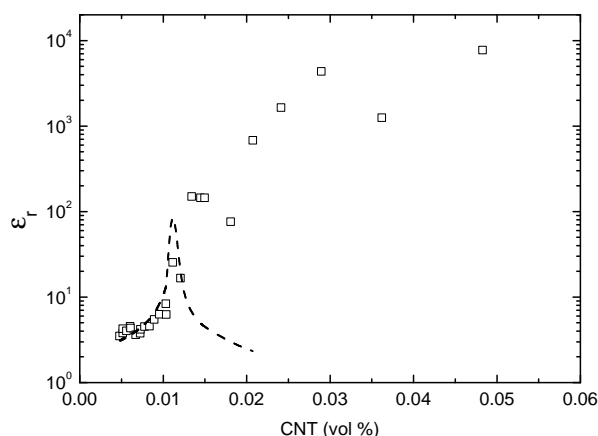
Fig. 4 Scatters represent relative permittivity *versus* filler content 17 sec after stopping the agitation. Dotted line corresponds to the theoretical permittivity described by Eq. 2.

3.4 Conductivity over time

This part presents dynamic percolation of CNT over time after agitation was stopped. Electrical conductivity measurements were carried out after stopping the agitation. Fig. 5 shows the results obtained for 3 filler contents: 0.003 vol%, 0.006 vol% and 0.024 vol%. The first filler content is below the percolation threshold determined previously ($V_c = 0.0087$ vol%), the second one is just below and the third one well
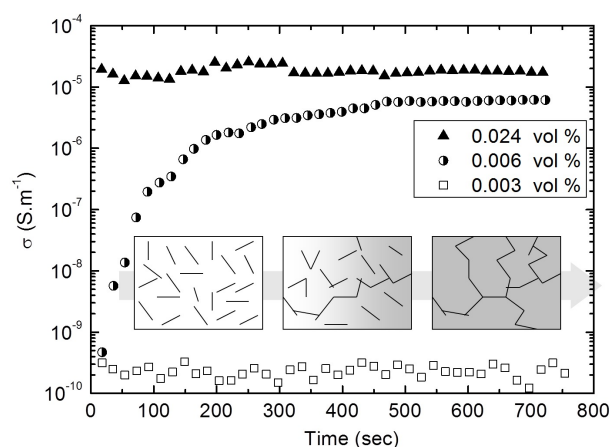
above.

Fig. 5 Conductivity *versus* time measured for 3 different filler contents: 0.024 vol%, 0.006 vol% and 0.003 vol%.

From Fig. 5, it is obvious that variations of electrical properties are solely observable for a given range of filler content. Above V_c, as the infinite cluster is already formed, conductivity doesn't change over time. The lowest filler content exhibits a conductivity independence too: due to the poor quantities of CNT in the liquid, the formation of an infinite network is not possible. In contrast, close to V_c the conductivity is modified, gaining 3 orders of magnitude after 500 seconds. These results are attributed to the aggregation of fillers in the silicone oil and are similar to the ones observed in molten polymers[30–32] and in liquids, such as chloroform[38]. Here we highlight that aggregation seems faster in liquid media than in molten polymers, mostly due to a difference in viscosities[27,51,52]. Finally we note that 10 minutes after that agitation was stopped, the whole suspension keeps homogeneous aspect. In other words, at short times dynamic percolation is not visually observable at macro scale and must occurs at sub-millimeter scale.

3.5 V_c over time

In order to investigate V_c dependence to time, electrical properties were measured for each filler content during the agitation step and over time for 700 sec after agitation was stopped. Results are represented on Fig. 6, describing the conductivity *versus* filler content for 3 different times: during agitation, 10 seconds and 700 seconds after agitation was stopped.

Kirkpatrick's relation (Eq.1) was used to determine the corresponding values of percolation threshold and critical exponent, parameters are indicated on table 1. After stopping
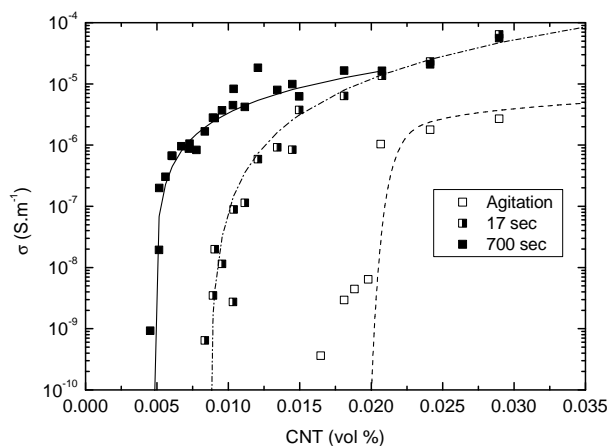
Fig. 6 Conductivity *versus* CNT content for 3 different time after stopping the agitation.

	Agitation	17 sec	700 sec
σ_0	4×10^{-5}	0.4	0.0056
μ	0.5	2.32	1.41
V_c	0.02	0.0087	0.0048

Table 1 Kirkpatrick parameters used to fit the experimental results showed in Fig. 6

the agitation, the self-structuration of CNT over time led to a percolation threshold decrease by a factor 4 (from 0.02 vol% to 0.0048 vol%).

The effect of time on V_c is presented in Fig. 7. After stopping the agitation, V_c decreases quickly and seems to reach a plateau of 0.0048 vol%, after 100 seconds. Close to 0.02 vol % (the percolation threshold measured under agitation), aggregation is really fast: CNT are close to each other and only small motions are required to develop an infinite cluster. In contrast, for filler content close to 0.0048 vol%, there are less particles: the time needed to build the percolation path is longer. Below 0.0048 vol%, there are not enough CNT to form the network, as shown in Fig. 5 for 0.003 vol%.

3.6 From homogeneous to heterogeneous system

As mentioned above, 10 minutes after stopping the agitation, filler aggregation is not visually observable. After 24 hours, however, 2 kinds of phases were clearly distinguished: the firsts, transparent, seemed constituted of poorly filled silicone oil, and the seconds constituted of denser CNT clusters, randomly dispersed in the silicon oil. Finally, after 2 weeks we observed a CNT sediment at the bottom of the suspension.
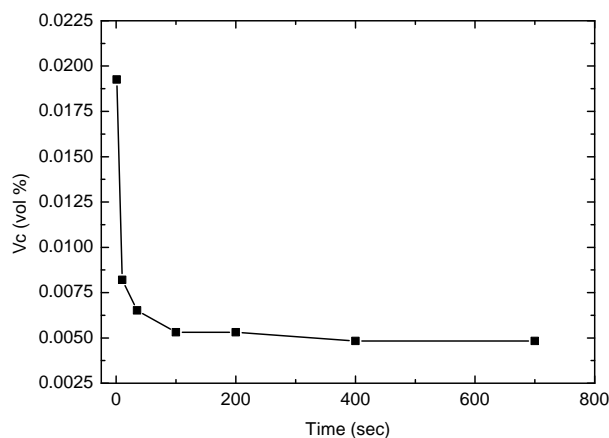

Fig. 7 Percolation threshold *versus* time.

The aggregation process could be divided in three steps: at short time, contacts are established between neighbors and a percolation path is created. However for longer annealing time, aggregation continues at a larger scale and the percolation network turns into floating and isolated macro-clusters: the system becomes strongly heterogeneous, even at macroscopic scale. At last, clusters fall to the bottom of the suspension due to a sedimentation effect. In order to accelerate the filler aggregation, the system was heated at 210^oC. Pictures of silicone oil are presented on Fig. 8. After 180 sec, a progressive separation occurred between oil and carbonaceous fillers. After 300 seconds at 210^oC, a heterogeneous system is clearly observed and is similar to the system observed after 24 hours at room temperature. Due to heat flow convection in the suspension, sedimentation is not observed at 210^oC.

This progressive separation between fillers and matrix was already mentioned in solid compound by Combessis[30]. He showed that dynamic percolation is the result of a transitory state. The final and more stable state for carbon nanotubes in the composite must be a complete separation with matrix, which could be controlled and freezed in polymers at solid state.
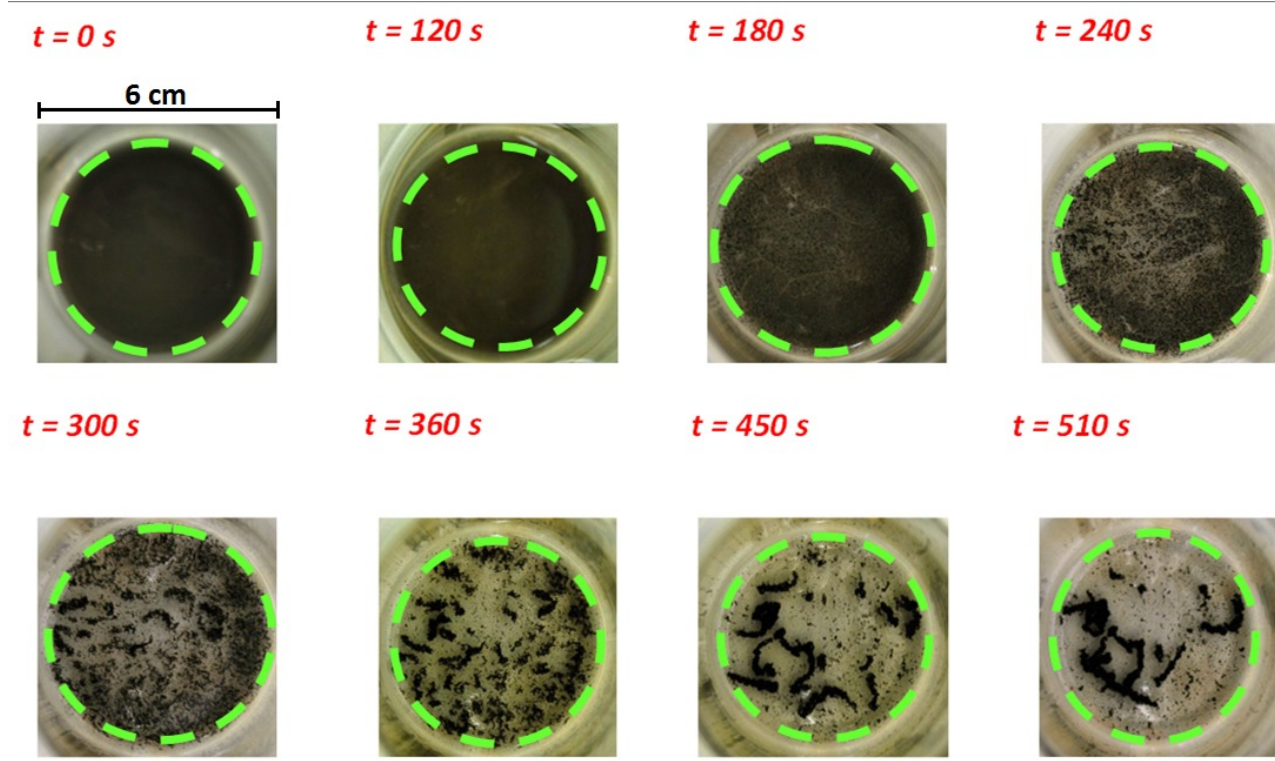
Fig. 8 Pictures of CNT aggregation over time after agitation was stopped, taking from the top of the suspension. After 300 seconds, floating clusters in silicon oil are observed.

Conclusion

Percolation of carbon nanotubes was studied in silicone oil. We proposed a new method which consists in slowly diluting the liquid and perform conductivity measurements. Due to the large amount of data obtained, the percolation threshold was determined with a great accuracy. Compared to the value usually observed in solid compounds, the one found in liquids was one order of magnitude lower. We also noticed that V_c tends to decrease over time after agitation was stopped. The low values were explained by the ease of carbon nanotubes to self-organize in liquid media. However, it was shown that dynamic percolation is just ephemeral and it was followed by formation of macroscopic clusters, which results in heterogeneous system. The final state for the system seems to be a sedimentation of CNT clusters. Dielectric measurement has brought more informations on the network structuration. In contrast with results commonly obtained in solid compounds, a strong capacitive part was kept above V_c. It was assumed that clusters may easily move in the oil and participate to the network permittivity. The new method presented in this paper opens some ways to understand how the properties of both particles and polymer, such as surface tensions, can control the dynamic percolation.

Acknowledgements

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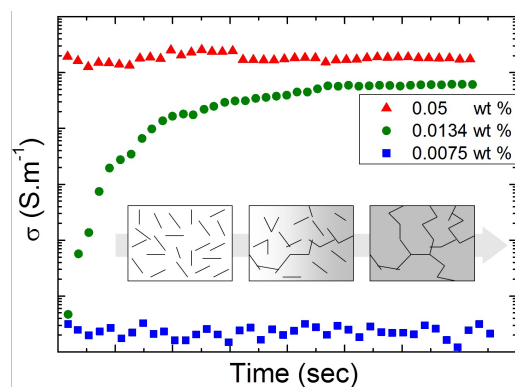
The aggregation mechanisms of carbon nanotubes in silicon oil have been explored in depth by the means of dielectrical measurements.

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