Effect of the field frequency during treeing tests in silicone polymers with different degree of crosslinking

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***Abstract*— Electrical treeing in silicone polymers has been investigated using a typical pin-plane geometry. The purpose of this work was to investigate the effect of ac field frequency upon the structure of treeing damage in a range of silicone dielectric materials. The liquid components were cured using different crosslinking ratios and hence both gels and elastomers were formed possessing a range of mechanical stiffness. These materials were characterized mechanically with an AR-G2 Rheometer. The results suggest that there is a clear dependence of the tree fractal dimension upon the applied ac voltage frequency and the sample's microstructure as characterized by oscillating rheometry.**

Keywords—Silicones; Electrical Trees; Cross-linked insulation; Elastic Modulus; Streamer; Fractal Dimension.

I. Introduction

Silicones are polymers consisting of silicon and oxygen atoms covalently bonded in long linear chains of inorganic siloxanes monomers (-Si-O-Si-). All of the crosslinking takes place at the ends of the polymer chains and the reactive groups are either hydroxyl (-OH) or vinyl (-CH=CH_2). They react with the crosslinking agent and a catalyst to give a cured elastomer. The chemical structure of silicones allows them to be produced in a number of variations [1-3]. By using siloxane units with different functionalities, products can be made with oily, polymeric, resinous or rubbery properties. This versatility of silicones makes them ideally suited for a variety of applications. Silicone elastomers are important materials for many application areas such as electrical insulation. They are increasingly being used to substitute for organic rubbers, because of their advantageous properties, such as high temperature and chemical resistance, combined with excellent electrical properties [1-3].

In this work we present preliminary results about the effect of the applied frequency upon the structure of treeing damage in dielectric silicone samples. These were prepared by varying the ratio of the two components of a commercial product that is offered as a dielectric silicone gel used as a potting or encapsulating compound. From the modification of this ratio, we could obtain a wide range of microstructures and crosslinking densities, from pure liquid components, gel-like samples and even a densely cross-linked elastomer [4]. Electrical trees were grown in the various samples and it was shown that there was a clear dependence of the tree fractal dimension upon the ac frequency and the microstructure as characterized by oscillating rheometry.

II. Experimental

A. Sample Preparation

A two part transparent dielectric silicone gel from RS Components Ltd. UK was used. According to the supplier's data sheet, when the two liquid parts (A and B) are mixed in one-to-one ratio, the final state obtained after the curing process will be a sticky gel-like material, semi-liquid, semi-elastomeric. However, we found that we could also obtain different microstructures (crosslinking densities) by modifying the mixing ratio [4].

Liquids A and B were manually mixed and degassed in a vacuum oven at room temperature (RT=20°C) for 5 minutes to form samples of each composition. The degassed mixtures were cast in transparent cells for treeing tests or placed in the plate of a rheometer for mechanical characterization.

The treeing cells were polystyrene cuvettes of 10mm x 10mm x 40mm of which the base had previously been removed and replaced by a plane film metal electrode (aluminum foil). Ogura Jewels Ltd. steel needles of 5µm tip radius were embedded in each fully filled liquid cuvette and positioned with a fixed gap of 3mm between the pin tip and film metal electrode. The specimens were cured for 4 hours at 65°C in an inert atmosphere.

B. Electrical Treeing Tests

A typical experimental setup for electrical treeing was used. The voltage source was a high voltage amplifier from TREK Inc. (model 20-20C-HS), controlled by a digital wave function generator. A Faraday cage was used to reduce the external electrical interference over the sample surrounding and also allow electroluminescence images. Tree growing videos were recorded with a full HD CMOS camera. A more detailed description can be found in [4].

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The fractal dimension was estimated as the tree grows by box-counting, which in the literature is one of the most widespread methods used to estimate the fractal dimension of electrical trees [5]. In practice, a square grid with box length *r* is used to cover the entire tree structure and the number of boxes containing a tree segment is counted. Thus for each box length *r* we have M(*r*) boxes. The fractal dimension, *Df*, is obtained using the expression $M(r) = M_0\, r^{-Df}$. Computational software has been used to obtain binary images of the tree structure from the decompiled video and to calculate the fractal dimension for each frame.

C. *Mechanical Tests*

Mechanical characterization was performed with an AR-G2 Rheometer from TA Instruments. The samples were prepared in the same way and cured in-situ in the rheometer. They were measured isothermally at 65ºC during curing with a maximum shear strain of 1%.

III. Results And Discussion

In Fig. 1 the extent of curing process after 4hs at 65ºC associated with the density of crosslinking created for different samples with mixing ratios varying between 0% and 100% of part A, is illustrated via the variation of the elastic shear modulus (G') against the percentage of A (%A). It can be seen that viscosity increases for samples having more than 20%A. With 45%A the material reaches the *gel point*. From this point up to 60%A, the elastic modulus is low enough for the material to behave as a sticky gel-like compound. The samples in the range from 66%A to 75%A behave like elastomeric rubber. The latter samples show a decrease in G', which can be associated with an excess of part A compared to part B, and therefore with a drop of the curing efficiency [4]. The samples with 80%A and 90%A show a similar behavior to the samples in the first gel region, and they constitute a second gel region. A new *gel point* is to be expected in the region between 90%A and 100%A (pure liquid A).

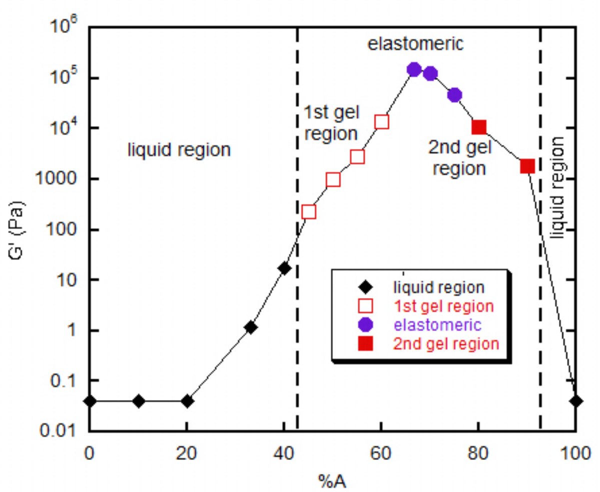

Fig. 1. Final microstructure of samples, as the elastic shear modulus (G') against the percentage of part A (%A), after in-situ curing during 4h at 65ºC. Rheometer setting: 1% strain, 0.2Hz, 65ºC.

Next three figures show the differences in the treeing process between the main regions in Fig. 1, i.e. liquid (Fig. 2), gel-like (Fig. 3) and elastomeric (Fig. 4), at two frequencies of the applied 8kVrms voltage: 50Hz and 6kHz.

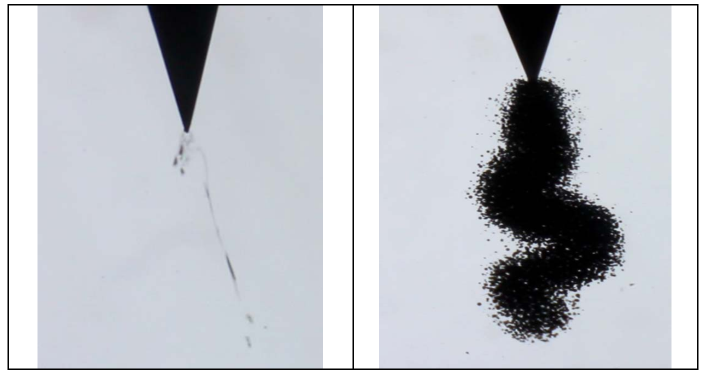

Fig. 2. Streamer projection at 50Hz (left) and "snake" of bubbles at 6kHz (right) for a liquid sample of 33%A.

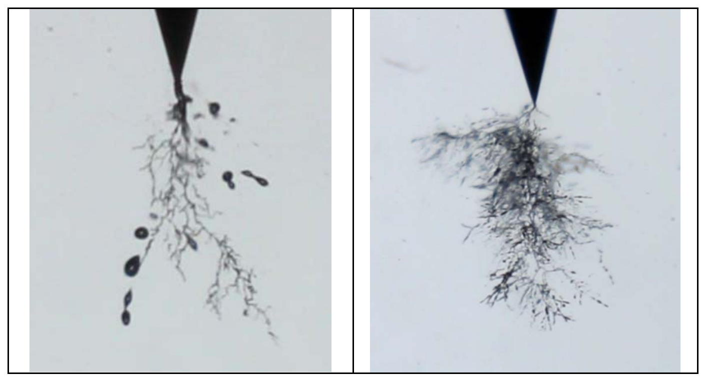

Fig. 3. Treeing comparison between 50Hz (left) and 6kHz (right) of ac field frequency for a gel-like sample of 50%A.

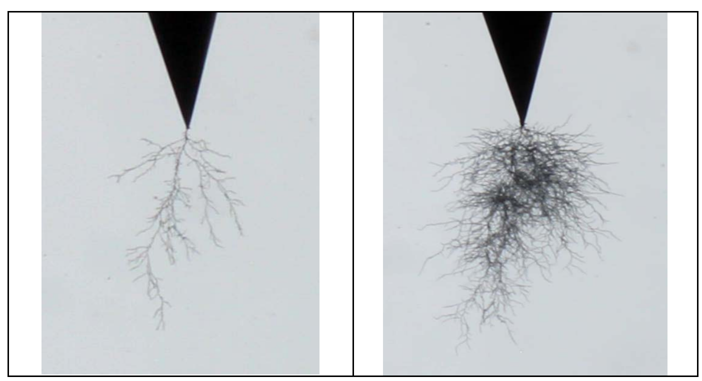

Fig. 4. Treeing comparison between 50Hz (left) and 6kHz (right) of ac field frequency for an elastomeric sample of 70%A.

Liquid samples with 33%A (Fig 2), show very fast long streamers or gaseous projections from the electrode pin tip at low frequencies (50Hz), which form and re-heal in a fraction of second without a permanent damage or formation of a tree structure.

On increasing the frequency to 6kHz, the dynamic is dominated by an abrupt injection of a "snake of bubbles" toward the plane ground electrode. The entire formation develops in less than 2-3 seconds before it reaches the counterelectrode. The "*snake*" structure of these thousands of micro-bubbles might be associated with the high rate of partial discharges and the viscous friction of the liquid, which slides and folds while it is pushed away from the pin tip (i.e. the expansion velocity is higher near the tip). After a voltage shutdown the micro-bubbles collapse more slowly i.e. in the order of a few seconds. Their average size and speed of decay seem to be related to the applied ac field frequency and the viscosity of the sample.

Gel-like samples (Fig. 3) at 50Hz develop slower streamers than liquids which usually end up as bubbles. This can be explained with the sample gelification, i.e. the cross-linked phase dominates the microstructure. The repetition of discharges over the same filament provokes chemical changes over its walls, winning the competition against *stage 1* of the self-healing mechanism [4], which is associated with the instant hydrostatic pressure of the still high liquid fraction and the restoring elastic forces of the expanded reticulated matrix [4]. The result is the creation of a quasi-permanent branch tree (backbone) growing from the tip of the needle. The tree will partially disappear hours or days after the voltage shutdown (self-healing *stage 2*) [4]. In the growing region at the tree tips or tree borders, gas-filled cavities develop during the partial discharge activity, and start to collapse when the expansion forces diminish. This can happen when the cavity is isolated from the tree structure or because the partial discharge activity decreases across the filament [4]. At 6kHz cavities do not develop. The tree backbone grows faster and exceeds the self-healing capacity.

Finally the last sequence (Fig.4) corresponds to an elastomeric sample (70%A). The electrical tree structure developed at 50Hz is similar to those reported in the literature for other common polymers, e.g. XLPE, epoxies, etc. [6,7]. The tree has extremely thin filaments and the cavity formation is inhibited due to the higher elastic modulus G'. Because the liquid faction inside the sample is the lowest of all the mixing ratios, *stage 1* of the self-healing mechanism cannot be observed.

Fig. 5 and Fig. 6 show the fractal dimensions (FD) of the samples belonging to the gel region (50%A and 55%A) and the elastomeric region (66%A and 70%A), at 50Hz and 6kHz, respectively. Fig. 7 summarizes the final values obtained for the tree structures at both frequencies.

At 50Hz, it can be observed that FD increases as the matrix become more cross-linked. In the gel samples, the FD values show oscillation between 0.7 and 1.17. This is associated to the growth mechanism which is based on the formation and collapse of cavities and streamers due to the *stage 1* of self-healing mechanism. For rubbery samples, corresponding to the maximum efficiency of crosslink, FD is around 1.4 and 1.5. At higher frequencies, the FD is around 1.7 for both gel and elastomeric samples.

In general the lower the tree fractal dimension, the more variable the discharges so we would expect that the trees with low FD to have a greater amount of PD fluctuations [8]. The increase observed in FD on going to higher frequency is consistent with the results found in [9] for branched electrical trees in polyethylene.

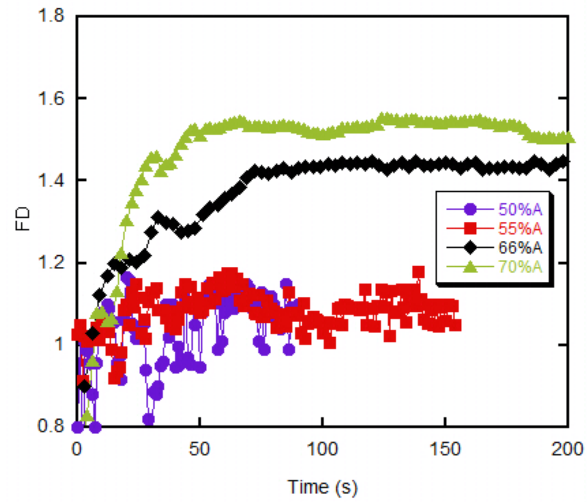

Fig. 5. FD vs. time for gel region samples (50%A and 55%A) and elastomeric samples (66%A and 70%A) at 50Hz.

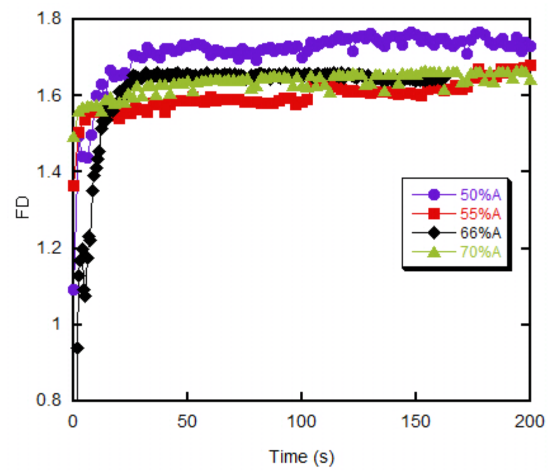

Fig. 6. FD vs. time for gel region samples (50%A and 55%A) and elastomeric samples (66%A and 70%A) at 6kHz.

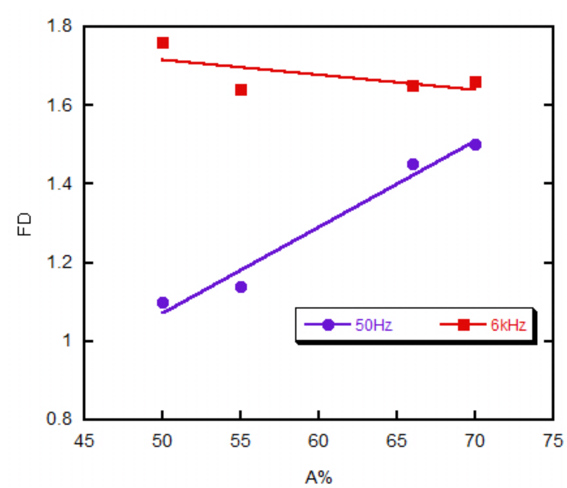

Fig. 7. Final FD values of Fig. 5 and Fig. 6 against the %A and for both frequencies: 50Hz (blue circles) and 6kHz (red squares).

IV. Conclusions

It was found that electrical damage changes with the crosslinking density of the silicone polymer. For gels and viscous-liquid samples, the degradation mechanism changes significantly with the ac field frequency.

In the viscous-liquid samples, filamentary fast streamers are observed at 50Hz frequency. Increasing the ac frequency causes the injection of thousands of micro-bubbles in a swirling structure that moves toward the earth electrode.

Formation of gas-filled cavities at the tree tips can be observed at 50Hz in the gel-like samples. These cavities collapse due to a self-healing mechanism. Increasing the ac frequency prevents the formation of gas cavities and leads to the formation of electrical tree structures with higher fractal dimension.

The effect of increasing the ac frequency in the elastomeric samples is to increase the fractal dimension of the resultant electrical tree structure as also found for trees in polyethylene.

The results of this article are preliminary and more detailed investigation will be performed in the future.

Acknowledgment

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