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Mechanical stretch influence on lifetime of Dielectric Elastomer Films

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ABSTRACT

Film pre-stretching is a widely adopted solution to improve dielectric strength of the DEA systems. However, to date, long term reliability of this solution has not been investigated. In this work it is explored how the dielectric elastomer lifetime is affected by film pre-stretching. The dielectric loss of soft polydimethylsiloxane (PDMS) films is studied for different stretch ratios by measuring $\tan\delta$. Additionally, time-to-breakdown was measured at DC electric stress for different stretch ratios. For this purpose, accelerated life test (ALT) were performed. The results obtained are compared with non-pre-stretched samples. This study suggests that no additional dielectric losses are caused by film stretching up to 80% of original dimensions.

Keywords: pre-stretching, lifetime, dielectric loss, time-to-breakdown, PDMS, reliability

1. INTRODUCTION

Dielectric elastomer actuators (DEA) have been widely investigated in the last decade and have attracted scientific community for its potential and scalable application [2].

Unfortunately, due to its intrinsic operation nature which requires high electric stresses, reliability of these actuators is limited by dielectric breakdown of the elastomer. This issue can be partially solved by applying internal tension (pre-stretching) to the sample: in fact, it has been extensively shown[1][4][5][7][8][9][10] a higher dielectric strength for stretched specimen. Although this solution is effective on the short-term time-scale [10], its behavior on longer working period cannot be stated a priori. Mechanical tension may in principle change material's permittivity and morphology facilitating current leakages or partial discharges phenomena, to mention some. The effect of such ageing mechanism is more evident on long-lasting electric stress rather than short time test.

Thus far, few investigations were conducted on the long-term reliability of this solution [12] [11] and very little is known on the life of the actuator on a long time-scale term.

This study is an attempt to fill the knowledge gap just mentioned by measuring the lifetime under high DC electric stress of silicon dielectric elastomer membranes with different pre-stretch ratio. Lifetime measurements are highly-time-demanding tests: one has actually to wait the unit under investigation fails to estimate its life; and this, for a statistically relevant number of units. It comes that such tests require long observation times, even years. To shorten the overall test duration, Accelerate Life Testing (ALT) methods are often used, instead. One of this has been adopted and is presented in this work.

Lifetime analysis has been performed on three sets of thin PDMS silicon membranes each at a different stretch ration. Results coming from each set have been compared to quantify the pre-stretch influence on the expected lifetime given a specific electric stress level.

In addition, $\tan\delta$ (dielectric losses) measurements were performed on a wide frequency spectrum for each membrane set to evaluate any dielectric changes related to pre-stretch action.

2. DIELECTRIC SPECTROSCOPY OF PDMS SILICON MEMBRANE

2.1 Sample preparation

Samples are cut in circle shape from commercially available Wacker Elastosil[®] Film 2030/50 silicone elastomer sheet, whose nominal thickness is 50 μm . Three sets specimen have been prepared, every set with a different stretch ratio ($\lambda_{r,0}$, $\lambda_{r,1}$, $\lambda_{r,2}$). The membranes of set 2 and 3, have been radially extended using the Open-Source Radial Stretching System [13] at the desired amount $\lambda_{r,1}=1.5$ and $\lambda_{r,2}=1.8$, for a total radial elongation of 50% and 80%, respectively. To constrain the radial tension, the stretched membranes have been then anchored to rigid ring-shaped PMMA frames of 42 mm

diameter through double-side polyamide silicon tape. Moreover, the rigid frames avoids the membrane wrinkling and allows easy handling and storage of the specimen.

The thickness of the samples has been measured by mean of infrared spectroscopy. This was done with Thermo Nicolet 6700 spectrometer. Long wavelength spectroscopy is a reliable contactless method that avoids systematic errors of mechanical measuring tool that can contaminate and even deform the samples.

Table 1. Membrane final thickness relative to stretch ratio. The values have been estimated by considering the silicone elastomer as incompressible mean.

	$\lambda_{r,0}$	$\lambda_{r,1}=1.5$	$\lambda_{r,2}=1.8$
Final thickness (μm)	48±2	21.95±1.00	17.6±0.57
Volume under test (Fractions of V_0)	1	0.45	0.36

Due to the highly electrostatic nature of silicon, it tends to attracts micrometer-size particle on its surface that cannot be removed without risk of damaging the specimen. For this reason, all the preparation and dielectric spectroscopy characterization have been performed in a ISO 7 clean environment to avoid silicon membranes' surfaces contamination from normal atmosphere particulate. Authors of this work found in preliminary tests (non-reported), that dielectric spectroscopy measurements may differ of orders of magnitude when the samples are prepared in room environment. Moreover it has been found that even if alcohol cleaning (isopropyl or ethanol) is an effective method for particulate removal, it permanently modify material's dielectric response. Resulting values from room environment particulate pollution would lead to wrong conclusions.

2.2 $\tan\delta$ characterization

The capacitance and dissipation factor $\tan\delta$ of all the bare membranes were measured with Megger Idax 300 Insulation Diagnostic analyzer. To avoid surface leaking current reading, a guarded bottom electrode cell was used in this study, as shown in Figure 1. The same cell was used all over the tests and its total active sampling surface is $A=490\text{ mm}^2$. The characterization were performed at constant room temperature of $T=23^\circ\text{ C}$ and the rms voltage applied is $V_{rms}=140\text{ V}$.

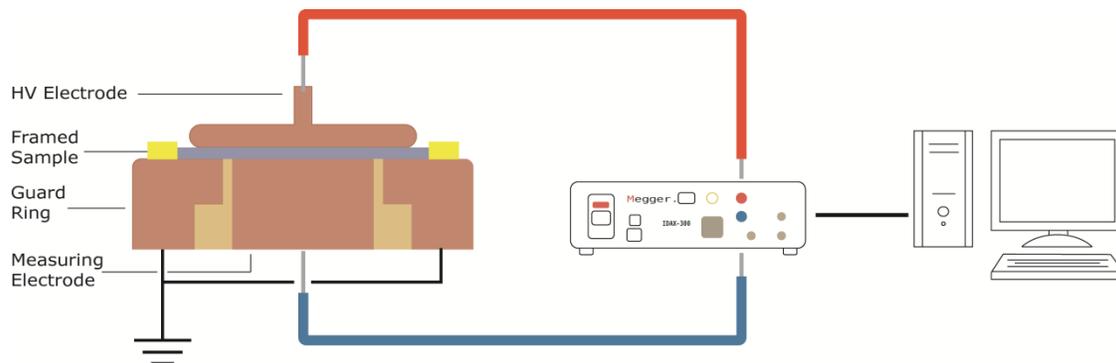


Figure 1. Dielectric spectroscopy setup. Measuring cell is depicted on the left-side. Top electrode, guard ring and bottom electrode are brass-made. The insulation between the guard ring and the measuring bottom electrode is epoxy-based. The scale is exaggerated for readability purpose.

For each set of membranes, a prior measurement of the capacitance was performed. This, measurement gives a baseline for the sample capacitance at each stress level. Specimen of a set, with a capacitance deviation larger than 5% (thickness deviation) from the corresponding baseline were rejected and not tested further, Figure 2a. The dielectric loss spectrum over a range of frequencies spanning from 10^{-2} Hz to 10 kHz of the specimen was then averaged over each set.

The sets containing expanded membranes $\{\lambda_{r,1}, \lambda_{r,2}\}$ demonstrate a slightly lower $\tan\delta$ compared to the relaxed $\lambda_{r,0}$ (see Figure 2b) ones. The lower values of dielectric loss for the highly stretched membranes would suggest improving dielectric properties with increasing stretch ratio. By the way, the amount of change is within the measurement error. Stretching may anyway play a role in altering the dielectric loss spectrum, but its contribution is negligible with respect to the achieved measurement error. From experiment, an evident dependence of dielectric loss with film stretching can't be speculated at this point.

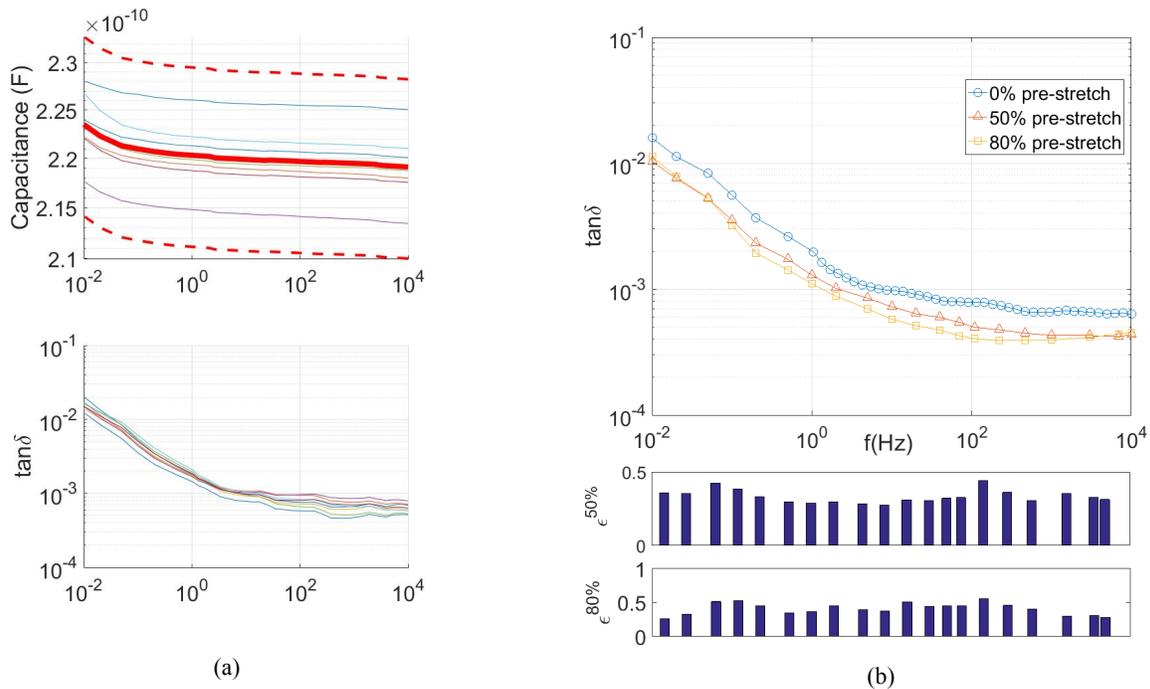


Figure 2. (a) Top Capacitance spectrum for specimen set 1. Thick line represent is the average value, dashed line the rejection bounds. Bottom: $\tan\delta$ for the same set. (b) Top: average $\tan\delta$ measurements for each membranes set. Bottom: absolute value of the relative difference by $\tan\delta$ of non-stretched membrane and the 50% and 80% stretched ones, respectively.

3. DC ACCELERATED LIFE TEST (ALT)

Accelerated life testing (ALT) is a useful method for assessing lifetime of an electrical insulation on short time scale. The general idea is to test at high levels of the accelerating variable (or variables) to speed up failure processes and form the results extrapolate to lower levels of the accelerating variables [16].

3.1 Cumulative exposure ageing model

Simple ALT test run at constant, high stresses: the disadvantage of this method is that it may still run too long because of the great scatter in failure times.

In this work, a cumulative exposure ALT test has been used: the stress is increased by steps over time, following a defined stress-pattern (multi-Step stress accelerated life test, SSALT), until the test specimen fails. Besides substantially shorten the overall duration test without affecting the accuracy of lifetime distribution estimates [17] this method is especially effective for newer material, when little information about appropriate test stresses [18], and for small test sample population [19].

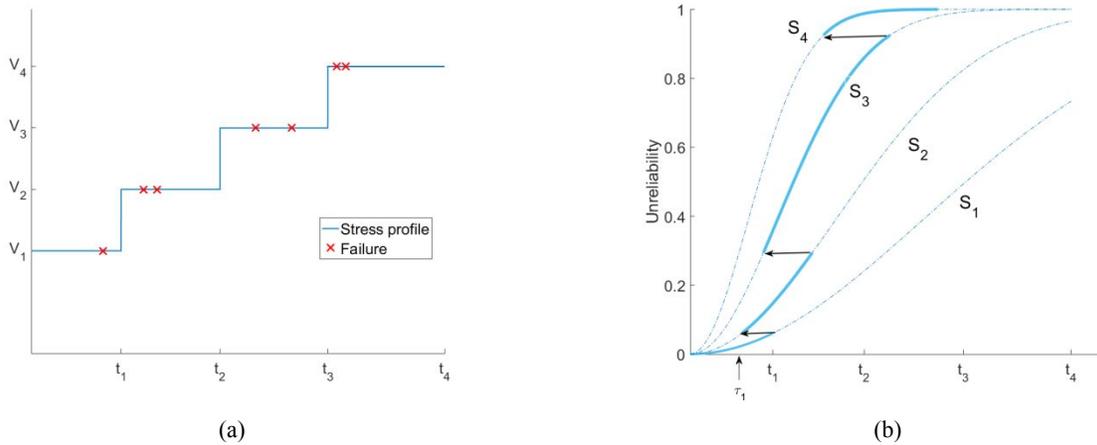


Figure 3. (a) Step-stress example pattern over time and (b) resulting cumulative Weibull cdf.

Given the following assumptions:

- Though several failure mechanisms potentially play role in insulation ageing (Partial discharge, electromechanical, thermal etc.), the aging mechanism is classified as uniform and strictly depending on the stress magnitude S_i
- The remaining life of specimens only depends on the current stress and the cumulative fraction failed, but not on how the stress was accumulated
- The failure time t^* follows a Weibull distribution at each i -th stress level S_i
- The Weibull shape parameter β is constant for all the stresses

the cumulative density function for this model becomes [15][14]

$$F_0(t^*, S_i) = 1 - \exp \left\{ - \left[(t^* - t_{i-1} + \tau_{i-1}) \left(\frac{S_i}{S_0} \right)^p \right]^\beta \right\}, \quad t_{i-1} \leq t^* \leq t_i \quad (1)$$

where t_{i-1} is the time at which the stress level is raised to S_i ,

$$\tau_{i-1} = (t_{i-1} - t_{i-2} + \tau_{i-2}) \left(\frac{S_{i-1}}{S_i} \right)^p \quad (2)$$

is the equivalent time spent at step $(i-1)$ -th, $\{S_0, p, \beta\}$ are positive parameters characteristic of the specimen and test method. Note that Eq.(1) is implicitly assumed that the Weibull time-scale parameter

$$\alpha(S) = \left(\frac{S_0}{S} \right)^p \quad (3)$$

Follows inverse power law relation.

Given a real stress pattern and the corresponding $\{t_j^*\}$ failure time measurements, parameter set $\{S_0, p, \beta\}$ can be estimated by fitting Eq. (1) with experimental data and, eventually, $F_0(t)$ can be calculated for any stress.

3.2 Parallel DC lifetime testing

The starting value for high DC stress level of ALT test was determined by prior DC breakdown voltage characterization for each set. According to IEC 60423-1 standard, the characterization was done with a minimum of 10 samples per set. Thinner sample will suffer of lower breakdown voltages compared to the relaxed (thicker) membranes ones.

Therefore, for quicker aging, the voltage stress $V_{70}^{(i)}$ corresponding to 70% unreliability of the 2-parameter Weibull analysis for each stretch ratio was chosen as starting ALT stress $S_{70}^{(i)}$ (See Eq.(1)).

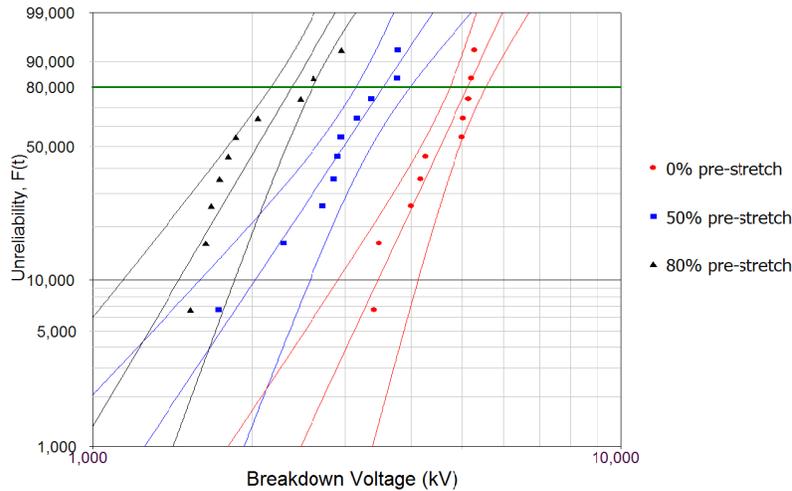


Figure 4 Weibull plot corresponding to preliminary breakdown voltage characterization of the three available set. Specimens with higher stretch and ,thus, thinner, have lower breakdown voltage. The test was executed by ramp voltage on 10 different specimen for each stretch set.

To further reduce total duration of measurements, a custom setup was used for testing several specimens at the same time (See Figure 5). For each of the three set of specimen, 16 samples were selected for the aging test: according the estimated Weibull statistic (Table 2), a surviving population of about 5 specimens is then expected at starting stress $V_{70}^{(i)}$.

Before stress step 1, each set was held at 10%,50%,70% of respective V_{70} . The voltage was then stepped-up every after a defined time period. Table 3 reports the voltage profile over time for each stretch ratio. To reduce the pressure of top electrode's weight, 25 mm rounded light aluminum disks were used as top electrodes and 15 mm steel ones (casted in epoxy ring) as ground electrode. Each electrode was polished to the surface roughness (<200 nm) measured with Dektak 150 surface profilometer.

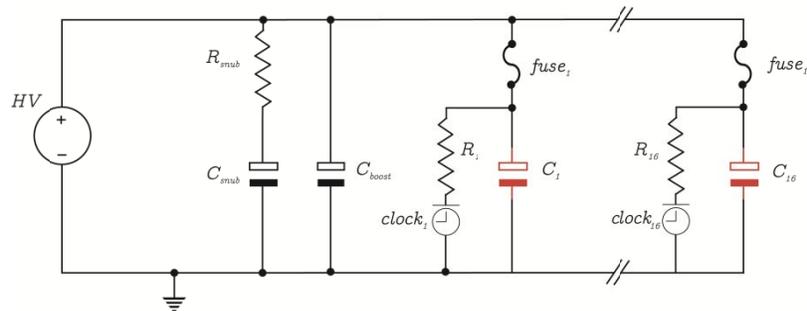


Figure 5. Parallel DC lifetime setup schematics with a single high voltage DC source. The sample-under-test C_i disconnection from main line at breakdown event is done through corresponding fuse $fuse_i$ blowing. At the same time, corresponding timer $clock_i$ stops counting. Up to 16 samples can be tested at the same time with this setup. Timer resolution is 0.01 h. For this work, HV source used is Heinzinger PNC1000-15.

Table 2. Weibull scale (α) and shape (β) parameters measured from breakdown voltage tests of bare membranes at different stretch ratio. Last column is the calculated initial stress level for SSALT corresponding to each stretch-set.

Radial Stretch λ_r	α	β	V_{70} (kV)
1	4.40	3.48	4.5
1.5	3.28	4.19	3.4
1.8	2.02	5.7096	2.1

3.3 Results on 50 um thick un-stretched PDMS membrane

A total of 7 samples out of 16 survived to step 1. For this residual specimens, the stress was then increased according to profile described in Table 3 till all the samples in the set failed. Four samples broke down during the first step, a fourth on the third step and 2 samples survived up to the final step for around a minute. The resulting fitted fraction of non-stretched samples that fail by age t is then:

$$F^{no\ stretch}_0(t, V) = 1 - \exp\left\{-\left[t\left(\frac{E}{112.27}\right)^{34.2}\right]^{0.39}\right\}$$

where $\beta_0=0.39\pm0.06$ is the Weibull shape parameter, $p_0=34.2\pm6.22$ is the power in the inverse-power law model (Eq. (3)), $E_0=112.27\pm3.01$, V is the electric stress in kV/mm and t is expressed in hours.

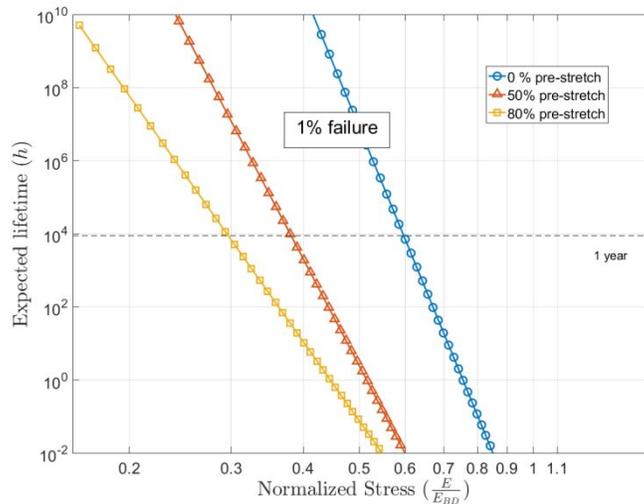


Figure 6. 1% Failure curves for the three membrane set. For better comparison, the stress has been normalized to respective breakdown field.

3.4 Results on 50% pre-stretch membrane

During preliminary test run, it has been observed that by applying a starting stress level of V_{70} (see Table 2) the resulting time-to-failure was shorter than the timer resolution ($0.01\ h$), consequently unusable for the analysis. Therefore a lower starting voltage level was chosen ($V_{start}=1.4\ kV$). From an initial population of 16 samples, a total of 6 survived to step 1 and the electric stress was then increased following the profile in Table 3. None of the specimen failed at the first stress level, but all of them finally broke down within the third step. As non-stretched membrane case, the times-to-failure have been used to fit Eq. (4), giving the following expression for the fraction of samples that fail by age t :

$$F_{0}^{50}(t,V) = 1 - \exp\left\{-\left[t\left(\frac{E}{115.94}\right)^{30.23}\right]^{0.26}\right\}$$

whit fitting parameter $\beta_{50}=0.26\pm 0.04$, $p_{50}=30.23\pm 12.84$, $E_{50,0}=112.94\pm 7.01$ and t expressed in hours. The parameter p_{50} , that play a major role in lifetime estimation of Eq.(3), is smaller compared with the previous p_0 . As can be seen in Figure 6, this results in a shorted expected lifetime.

3.5 Results on 80% pre-stretch membrane

As in 50% stretch case, also 80% stretched membranes suffered a too-short time-to-failure at starting voltage of respective V_{70} . The start voltage $V_{start}=1.4$ kV was, then, used also in this case. For this test, only two steps were needed to break all the 9 survived samples from step 1. The fit of time-to-failure gives

$$F_{0}^{80}(t,V) = 1 - \exp\left\{-\left[t\left(\frac{E}{113.57}\right)^{22.36}\right]^{0.28}\right\}$$

whit parameter $\beta_{80}=0.28\pm 0.23$, $p_{80}=22.36\pm 15.35$, $E_{80,0}=113.57\pm 21.24$ and t expressed in hours. Also in this case, the p_{80} magnitude decreases, and accordingly the expected lifetime for a given stress.

Table 3. Step-stress pattern and test data for all sets of PDMS membrane. The final step and the corresponding time to failure of the specimens are also reported.

Step #	Pattern & Specimen data											
	$\lambda_{r,0}$				$\lambda_{r,1} = 1.5$				$\lambda_{r,2} = 1.8$			
	Duration (h)	Stress (kV/mm)	Final Step #	Failure time(h)	Duration (h)	Stress (kV/mm)	Final Step #	Failure time(h)	Duration (h)	Stress (kV/mm)	Final Step #	Failure time(h)
1	503.94	90	1	0.35	152.82	63.78			152.82	79.54	1	0.48
			1	8.99							1	1.73
			1	156.53								
			1	161.43								
2	93.86	95			176.2	91.11	2	153.11	3.25	113.63	2	152.85
							2	253.04			2	152.95
							2	302.85			2	153.84
							2	329.74			2	154.12
											2	155.31
				2	155.42							
				2	156.07							
3	71.74	98	3	673.41	4.02	123.01	3	329.74				
							3	331.43				
4	22.7	104										
5	2.06	110										
6	0.02	122	6	694.74								
			6	694.76								

4. DISCUSSION

The stretching of PDMS membrane plays a key role for its long-term dielectric stability. Although this technique provides an immediate enhancement for the dielectric strength, it pays back with a shorter expected lifetime on the long-time scale. It has been found, in fact, that the estimated time-to-failure at a given relative electric stress reduces with the increasing pre-stretch ratio, see Figure 6. This trend is mainly quantified by the parameter p of Eq. (1), which also represents the negative slope of lines in Figure 6. The estimated value of $p_0=34.2$ of the un-stretched membrane reduces to $p_{50}=30.23$ for the 50% pre-stretch case and decrease further for the 80% stretch, $p_{80}=22.36$. These parameters have been calculated by Non-Linear Least Square regression of Eq. (1) that better suits small specimen population and gives the best fit of the data. The testing time and the number of steps used, influenced the error on the parameters' estimation: the last set $\lambda_{r,2}$ only experienced 2 steps and has been tested for a maximum total time of 156.07 hours and its relative parameters' errors are larger compared to the set $\lambda_{r,0}$ which underwent 6 steps and a maximum test time of 694.76 hours. The set $\lambda_{r,1}$ with a total of 3 steps and test time of 331.43 hours sits at an intermediate level of parameters' error magnitude. These results suggest using smaller steps but larger in number and longer in time.

It is worth to remember that this model works under the assumption mentioned in (3.1). It doesn't consider different possible breakdown mechanisms, but both aging and degradation are supposed to be a monotonic function of only applied electric stress. Moreover, lifetime is also very dependent on the geometry and nature of electrodes. Nevertheless, from results of (3.3)(3.4)(3.5) a clear tendency of lifetime with stretching was found.

5. CONCLUSIONS

The influence of pre-stretching dielectric silicon elastomer on both insulating properties and lifetime has been studied. A method for accelerated ageing was also presented and applied for the first time on a dielectric elastomer. From dielectric spectroscopy measurements, an evident dielectric loss trend associated with the increasing stretch ratio couldn't be speculated at the voltages used. However, results from accelerated ageing tests, revealed that the reliability of dielectric silicon membranes is reduced by increasing stretch ratio. Therefore, albeit pre-stretch technique might improve the dielectric's breakdown strength value, it is likely to shorten its total lifetime.

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REFERENCES

- [1] Kofod G., Pelrine R., Kornbluh R., "High-strain actuator materials based on dielectric elastomers." *Advanced Materials*, 12(16), 1223–1225 (2000).
- [2] Shea H., Rosset S., "Small, fast, and tough: Shrinking down integrated elastomer transducers.", 3(3), (2016)
- [3] Pei Q., Joseph J., Pelrine R., Kornbluh R., "High-speed electrically actuated elastomers with strain greater than 100%", *Science*, 287(5454), 836-839(2000).
- [4] Baumgartner R., Suo Z., Bauer S., Keplinger C., Li T., "Harnessing snap-through instability in soft dielectrics to achieve giant voltage-triggered deformation.", *Soft Matter*, 8, 285-288 (2012).
- [5] Kofod G., "The static actuation of dielectric elastomer actuators: how does pre-stretch improve actuation?", *J. Phys. D: Appl. Phys.*, 41(215405), (2008).
- [6] Suo Z., Kofod G., Kollosche M., Zhu J., "Complex interplay of nonlinear processes in dielectric elastomers." *Physical Review E*, 85(051801), (2012).

- [7] Jordi C., Kovacs G., Kovacs R., Clarke D. R., Suo Z., Lu T., Huang J., “Dielectric elastomer actuators under equal-biaxial forces, uniaxial forces, and uniaxial constraint of stiff fibers.”, *Soft Matter*, 8(6167), 6167-6173(2012).
- [8] Trols A., Kogler A., Baumgartner R., Kaltseis R., Keplinger C., Schwodiauer R., Graz I., Bauer S., “Stretch dependence of electrical breakdown strength and dielectric constant of dielectric elastomers”. *Smart Mater. Struct.*, 22(10), (2012).
- [9] Zakaria S., Morshuis P. H. F., Benslimane M. Y., Yu L., Skov A.L., “The electrical breakdown strength of pre-stretched elastomers, with and without sample volume conservation.” *Smart Mater. Struct.*, 24(055009), (2015).
- [10] Gatti D., Haus H., Matysek M., Frohnapfel B., Tropea C., Schlaak H. F., ”The dielectric breakdown limit of silicone dielectric elastomer actuators.”, *Applied Physics Letters*, 104(5), (2014).
- [11] de Saint-Aubin C. A., Rosset S., Shea H., “Aging setup for high cycle tests on dielectric elastomer actuators”, *EuroEAP 2016 - Sixth international conference on Electromechanically Active Polymer (EAP) transducers & artificial muscles*,(2016).
- [12] Matysek M., Lotz P., Schlaak H.F., “Lifetime investigation of dielectric elastomer stack actuators.”, *IEEE Transaction on Dielectrics and Electrical Insulation*,18(1),89-96,(2011).
- [13] S. E. Schausberger , R. Kaltseis, Michael Drack, U. D. Cakmak, Z. Major, S. Bauer, “Cost-Efficient Open Source Desktop Size Radial Stretching System With Force Sensor”, *IEEE Access*, 3,556-561(2015)
- [14] Nelson W., ”Accelerated Life Testing – Step-Stress Models and Data Analyses”, *IEEE Transaction of Reliability*,29(2),103-108(1980).
- [15] Hirose H., “Theoretical foundation for residual lifetime estimation”, *Trans. IEE of Japan*, 116-B(2),168-173(1996).
- [16] Escobar L.A., Meeker W.Q., “A review of Accelerated Test Models.”, *Statistical Science*, 21(4),552-577(2006).
- [17] Zhao W., Elsayed A., “Optimum Accelerated Life Testing Plans Based on Proportional Mean Residual Life.”, *Quality and Reliability Engineering International*,21(7),701-713(2005).
- [18] Hu C.H., Plante R.D., Tang J., ”Step-stress accelerated life tests: a proportional hazards-based non-parametric model.”, *IIE Transactions*,44(9),754-764(2011).
- [19] Tseng S.T., Wen Z.C.,”Step-stress accelerated degradation analysis for highly reliable products.”, *Journal of Quality Technology*, 32, 209–216(2000).