Filled liquid silicone rubbers: Possibilities and challenges

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ABSTRACT

Liquid silicone rubbers (LSRs) have been shown to possess very favorable properties as dielectric electroactive polymers due to their very high breakdown strengths (up to 170 V/μm) combined with their fast response, relatively high tear strength, acceptable Young's modulus as well as they can be filled with permittivity enhancing fillers. However, LSRs possess large viscosity, especially when additional fillers are added. Therefore both mixing and coating of the required thin films become difficult. The solution so far has been to use solvent to dilute the reaction mixture in order both to ensure better particle dispersion as well as allowing for film formation properties. We show that the mechanical properties of the films as well as the electrical breakdown strength can be affected, and that the control of the amount of solvent throughout the coating process is essential for solvent borne processes. Another problem encountered when adding solvent to the highly filled reaction mixture is the loss of tension in the material upon large deformations. These losses are shown to be irreversible and happen within the first large-strain cycle.

Keywords: elastomers, silicones, composites, electroactive, strain displacement

1. INTRODUCTION

Elastomers from polydimethylsiloxane (PDMS) polymers cross-linked via silylation with low-functional [1] or even high-functional cross-linker [2] have poor mechanical properties when no reinforcing particles are added. Therefore silicone networks used in dielectric electroactive polymer (DEAP) applications - and most other applications - are reinforced with silica fillers for mechanical strength as well as to obtain better ultimate properties. [3] The extent to which the fillers influence the elastomer properties depends on filler size, filler treatment, how well the fillers are dispersed in the elastomer matrix, and the interfacial interaction between the matrix and fillers. [4]

Classically silicone elastomers are filled with silica in order to improve the mechanical properties but fillers greatly complicate the mechanical response by including numerous phenomena such as stress softening and mechanical hysteresis. [5] The nonlinearity of elastomers is commonly characterized by large strains under static conditions, a non-linear stress-strain response and strain rate dependency. Finally, under cyclic actuation, hysteresis and stress-softening are observed. [6] Hysteresis is related to the dissipative nature of the elastomer i.e. the viscoelasticity [7, 8] and the viscoplasticity [9]. It is characterized as the difference between the loading and the unloading paths during a cycle. Additionally the so-called Mullins effect is the stress-softening taking place after the first cycle. It can be assumed to be instantaneous and irreversible due to rearrangements in the microstructure occurring whenever the load increases beyond its prior all-time maximum value. The Payne effect is the stress-softening occurring at low strains and is manifested as a decrease of the shear modulus with strain amplitude at around 0.1% strain amplitude [10].

There are two types of commercial silicone formulations, which are based on curing temperatures, namely 1) high temperature vulcanizable (HTV) and 2) room temperature vulcanizable rubbers (RTVs). HTV rubbers can be further classified as 1) liquid silicone rubbers (LSRs) and 2) millable silicone rubber depending on the degree of polymerization. LSRs are low viscosity silicone (low molecular weight) which is pourable or self-leveling in consistency with a viscosity of 300,000 to 1,000,000 cPs. They are based on platinum curing which offers curing time of a few seconds at elevated temperatures, and some of the compositions may also be additionally peroxide cured. LSR premixes are more viscous than RTVs, and in general the cured elastomers have much higher Young moduli (Y) than RTVs. [11, 12]

In some transducer applications such as wave harvesting, films with high Y are favorable, whereas for actuation purposes a low Y is desired. [13] The thin films with high Y are easier to handle process-wise mainly due to easy peel from substrates. [14] Hence, LSRs are chosen in the present work and the properties of LSR-filler composites along with breakdown strength of these composites are reported. To our knowledge, there is no detailed literature on LSR-filler composites for DEAP application. Previous studies on optimization of various types of titanium dioxide silicone composites as candidates for DEAP materials focused on silicone RTV elastomers. [15-17]

Pure LSR elastomers and LSR composites are evaluated based on two figures of merit, namely with respect to actuation $F_{om}(DEA)$ and to generation $F_{om}(DEG)$. The theory and derivation of F_{om} to assess the elastomer performance as a dielectric elastomer actuator (DEA) has been performed by Sommer-Larsen and Larsen [18] and based on this F_{om} for a dielectric elastomer generator (DEG) has been presented by McKay et al [19].

$$F_{om}(DEA) = \frac{3\varepsilon_r \varepsilon_0 E_{breakdown}^2}{Y} \qquad (1)$$

$$F_{om}(DEG) = \frac{\varepsilon_r \varepsilon_0 E_{breakdown}^2}{2} \qquad (2)$$

Where E_{BD} is the electrical field at which electrical breakdown occurs. ε_r is the relative dielectric permittivity and ε_0 is the permittivity of free space. The failure of the films as DEA or DEG is predominantly caused by factors like dielectric breakdown and natural wear whereas the ultimate elongation does not play a significantly role for silicone elastomers which usually exhibit elongations of more than 100-200%. Therefore elongation at break becomes a negligible parameter and is not taken into consideration in this work.

2. EXPERIMENTAL

2.1 Materials

Five liquid silicone rubber (LSR) formulations have been tested.

Elastosil® LR 3043/30, Elastosil® LR 3043/50, Elastosil® LR 3005/50, POWERSIL® XLR® 630 A/B and MJK® 135 5/13 were obtained from Wacker Chemie AG, Germany. The LSRs are supplied as premixes A and B. The premix A contains vinyl terminated PDMS and platinum catalyst, and premix B contains vinyl terminated PDMS and the hydride cross-linker. The mixing ratio of premix A and B is 1:1.

Solvent OS-20 (an ozone-safe volatile methylsiloxane (VMS) fluid) was obtained from Dow Corning, USA.

The nanofillers are:

Aeroxide® TiO_2 T805, hydrophobic metal oxide from Evonik Industries AG, Germany. Primary particle size is ~21 nm. Specific surface area (BET) is 35-55 m^2/g.

Aeroxide® STX801, hydrophobized fumed silicon-titanium mixed oxide with a titanium dioxide core and a silica shell from Evonik Industries AG, Germany. Specific surface area (BET) is 40-70 m^2/g.

HOMBITEC® TiO_2 RM130F, hydrophobic rutile titanium dioxide from Sachtleben Chemie GmbH, Germany. Primary particle size is ~15 nm.

2.2 Sample preparation

Nanofillers were mixed into the silicone premix A using Speedmixer DAC 150FVZ (Hauschild Co., Germany) for 2 minutes at 3500 rpm. The premix B and solvent were then mixed into the previous mixture for another 2 minutes at 3500 rpm in the Speedmixer. The films were coated with a thin film applicator 3540 bird film applicator (Elcometer, Germany) on a suitable substrate and cured in an oven at 100°C for 7-10 minutes. For the rheological and dielectric permittivity experiments the thickness of the film was 0.5-1.0 mm. And for the breakdown, strain displacement and tear tests it was 30-90 μm.

2.3 Characterization

2.3.1 Rheological measurements

Rheological characterization of the prepared films was performed with an ARES-G2 rheometer (TA Instruments, USA) set to a controlled strain mode at 2% strain, which was ensured to be within the linear viscoelastic regime (LVR) that was determined as the region where the response of storage and loss moduli (G' and G'') were constant over the entire range of strain for the two applied frequencies. The measurements were performed with parallel plate geometry of 25 mm at room temperature, with a normal force of 6-8 N, in the frequency range of 100-0.01 Hz. From the shear modulus (G) the Young's modulus (Y) is calculated:

$$Y = 2G(1+\upsilon) \tag{3}$$

$$G = G'(\omega \rightarrow 0) \tag{4}$$

The value of G ($G = G'(\omega \rightarrow 0)$) is obtained from the frequency sweeps of the rheological tests. The Poisson ratio (υ) for silicone elastomers is taken as 0.5 since the silicone elastomers are assumed to be incompressible.

2.3.2 Dielectric characterization

Dielectric relaxation spectroscopy (DRS) was performed on a Novocontrol Alpha-A high performance frequency analyzer (Novocontrol Technologies GmbH & Co. KG, Germany) operating in the frequency range 10^{-1}-10^{6} Hz at 23°C. The diameter of the samples tested is 25 mm.

2.3.3 Breakdown strength test

Electrical breakdown tests were performed on an in-house built device based on international standards (IEC 60243-1 (1998) and IEC 60243-2 (2001)). The film thicknesses are measured by microscopy of cross-sectional cuts and the distance between the spherical electrodes is set accordingly with a micrometer stage and gauge. An indent of less than 5% of sample thickness is added to ensure that the spheres were in contact with the sample. The polymer film (30-90 μm) is slid between the two spherical electrodes (radius of 20 mm) and the breakdown is measured at the point of contact with a stepwise increasing voltage is applied (50-100 V/step) at a rate of 0.5-1 steps/s. Each sample is subjected to 12 breakdown measurements and an average of these values is indicated as the breakdown strength of the sample.

2.3.4 Strain displacement measurement

The strain displacement of the prepared films has been investigated in a simple manner by clamping the films in one end and then strain the films to 120% and allow the films to relax. This is repeated 5 times for each film. The staying deformation (strain displacement) is measured after each cycle.

2.3.5 Tear test

The tear strength of the elastomers was measured according to specification ASTM D 624 B (2007) - Type B Tear Strength (nicked tap end) standard for the determination of the Tear Strength of Conventional Vulcanized Rubber and Thermoplastic Elastomers using a Zwick/Roell Zmart.pro material tester (Zwick GmbH & Co. KG, Germany). The sample was first placed between clamps initially separated by a distance of 180±1 mm, the test specimen was lengthened at 500±50 mm/min until failure. Each sample is subjected to 4 tear measurements and then averaged.

3. RESULTS AND DISCUSSION

In the following two categories of composites are investigated. The composites are split according to whether they possess little/no strain displacement (low loadings of fillers) or significant strain displacement (higher loadings). For the particular composites consisting of LSR elastomers and various types of titanium dioxide the Mullins effect causing the strain displacement starts to become significant above 10phr titanium dioxide loading.

3.1.1 Low concentrations of fillers

First of all composites with low concentrations (up to 10phr) of titanium dioxides of various types were investigated. The films were all homogeneous and a selection of data is given in Table 1. No significant evidence of the Mullin effect was measured since all films had a strain displacement of less than 0.5% after 120% strain. These small losses were shown to be irreversible and happen within the first large-strain cycle. The observation on the irreversibility was also valid for the films showing a large Mullin effect.

However, as can be seen in Table 1 the obtained increase in dielectric permittivity at high frequencies was not as large as anticipated but as will be discussed in section 3.2.1 higher loadings of all investigated types of titanium dioxide caused significant strain displacement. The Young's moduli (Y) are calculated based on the assumption of incompressible and isotropic composites such that the Young's modulus is 3 times the shear modulus, which has been measured for all the films. This assumption holds for neither the pure LSRs or the composites and all checked samples rather had a proportionality factor of 9-10 than the theoretical factor of 3 (approximately 10 films were tested in both shear and elongation and all confirmed this trend). This may be due to the Payne effect.

Table 1. Experimental data and figures of merit for pure LSR elastomers and composites thereof.

No.	Material	Filler	ε_r (f = 1MHz)	ε_r (f = 0.1Hz)	E (V/μm)	Y * (Pa)	Fom (DEA) (f = 1MHz)	Fom (DEA) (f = 0.1Hz)	Fom (DEG) (f = 0.1Hz)
1		LR3043/30	2.80	2.80	122.8	253260	4.43	4.43	1.87
2		3% T805	3.40	3.76	90.3	234810	3.10	3.45	1.35
3		6% T805	3.64	3.79	103.4	179070	5.75	6.02	1.80
4	LR3043/30	9% T805	3.68	5.31	97.0	208470	4.43	6.37	2.21
5		3% STX801	3.29	3.21	123.8	207480	6.46	6.28	2.17
6		6% STX801	3.72	3.43	124.2	220800	6.90	6.37	2.34
7		9% STX801	2.84	3.80	133.2	167820	7.97	10.7	2.98
8		LR3043/50	2.80	2.80	144.5	536000	2.92	2.92	2.58
9		3% T805	2.16	2.72	129.0	283140	3.36	4.25	2.00
10		6% T805	3.63	5.36	123.3	288000	5.13	7.52	3.60
11	LR3043/50	9% T805	3.35	5.63	141.4	304800	5.84	9.82	4.98
12		3% STX801	3.20	3.32	153.3	384300	5.22	5.40	3.45
13		6% STX801	3.40	3.59	156.8	306000	7.26	7.70	3.91
14		9% STX801	3.36	3.80	167.1	261900	9.47	10.7	4.69
15		LR3005/50	2.90	2.90	115.0	280770	3.63	3.63	3.39
16		3% T805	3.52	3.50	109.7	179040	6.28	6.20	1.86
17		6% T805	3.48	3.18	118.1	255000	5.04	4.60	1.96
18	LR3005/50	9% T805	3.27	3.18	135.9	223950	7.17	6.99	2.59
19		3% STX801	3.54	3.36	108.0	155070	7.08	6.73	1.73
20		6% STX801	3.77	3.27	138.0	255180	7.43	6.46	2.75
21		9% STX801	2.85	3.39	143.0	363600	4.25	5.04	3.06
22		XLR 630	2.90	2.90	134.3	244620	5.66	5.66	4.63
23		3% T805	3.26	3.05	118.0	296100	4.07	3.81	1.88
24		6% T805	3.44	3.16	138.0	483000	3.63	3.27	2.66
25	XLR630	9% T805	3.56	3.32	107.0	549300	1.95	1.86	1.68
26		3% STX801	3.25	3.19	108.0	291900	3.45	3.36	1.64
27		6% STX801	3.88	3.44	173.0	186630	16.55	14.7	4.55
28		9% STX801	3.76	3.50	126.0	206220	7.70	7.17	2.45

* The Young moduli are calculated based on the relation between shear and elastic moduli.

The relatively low loading of TiO_2 does not cause great improvement in dielectric permittivity. The best results are increases of 33% at 0.1Hz and 100% at 1MHz. However, all improvements are obtained without compromising the breakdown strength of the pure LSR elastomer significantly, in some cases actually improving it. The trends in the breakdown strengths will be discussed in the following section.

In Figure 1 the figures of merit for the four different LSR elastomers as well as their composites are shown. It is obvious that the LSR composites investigated here are not suitable for candidates for DEAP actuation but are very suitable as materials for generators.
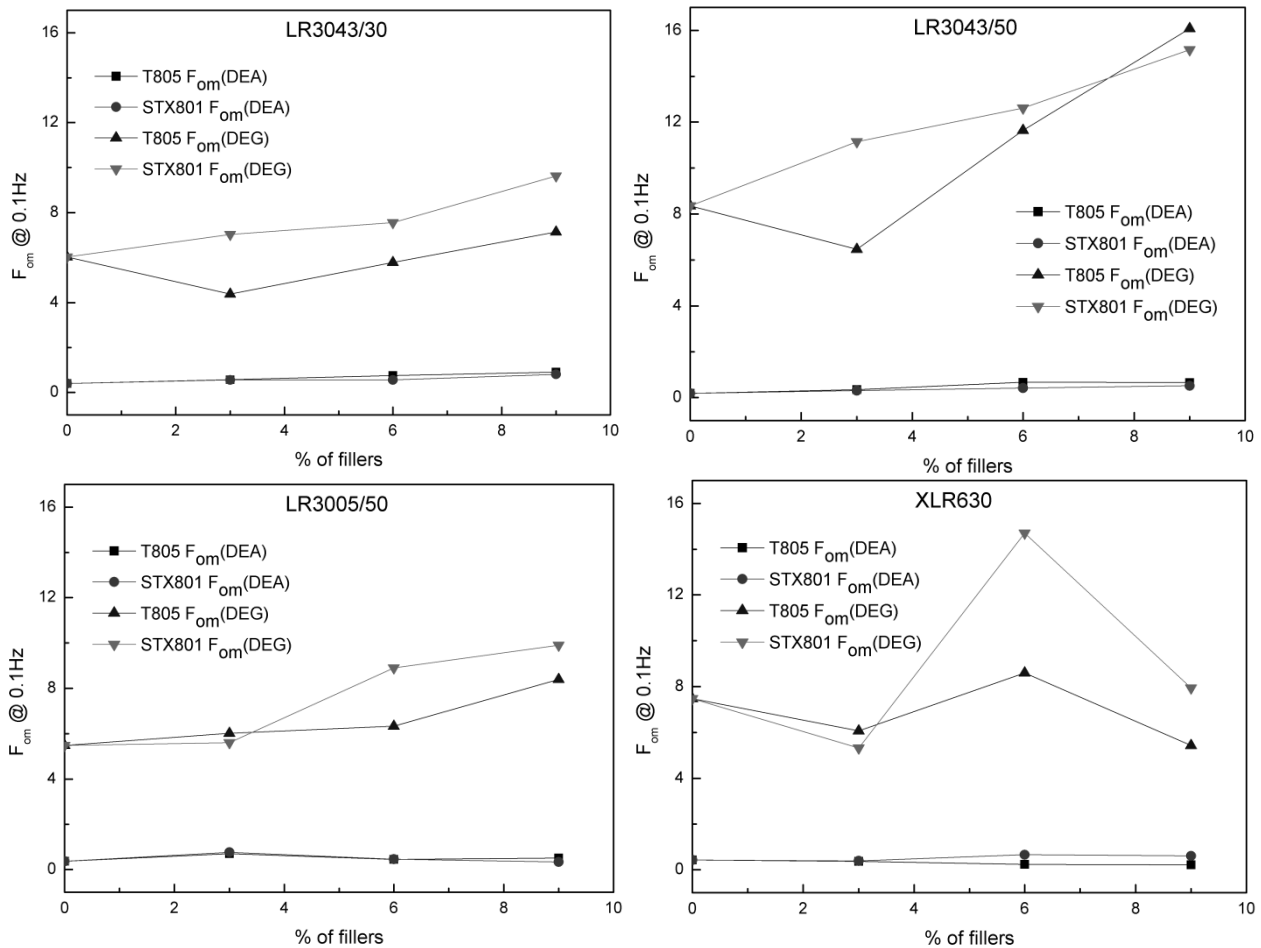

Figure 1. F_{om}(DEA) and F_{om}(DEG) @ f = 0.1 Hz for the pure LSRs and their composites. The benchmark material, which is used for normalization, is the pure ERT625 such that a value of 1 or less corresponds to no improvement of the actuation/generation properties. All figures of merit for actuation are lower than 1.

3.1.2 Effect of Young's modulus on electrical breakdown strength

It has previously been shown that the electrical breakdown strength of polyethylene was increased with increasing Y. [20] In Figure 2 the breakdown strength as function of Y is plotted. The results indicate that the resistance of the elastomers to dielectric breakdown is indeed enhanced with the increase of Y but not in a trivial or universal way. The dielectric strength is affected by various factors, including the polymer structure and the sample geometry. [21, 22] The sample geometry is here kept constant so the results in this study are directly comparative. The films with high Y have compact polymer chains which can trap charges. [23] Trapped charges cannot flow easily and hence a higher electric field is needed to induce electron avalanches and thus the films possess high breakdown strength. [24]

It is not possible to increase Y unlimitedly for a given composite since the formulations start to become inhomogeneous with the result that the opposite effect is encountered, i.e. premature electrical breakdowns. Such highly filler loaded films are the scope of the next part of the article. From Figure 2 it is seen that it is not only the Young's modulus that influences the breakdown strength but that it plays a significant role. It can be seen that as a guideline the breakdown

strength of silicone elastomers with TiO_2 fillers scales linearly with the Young's modulus. Other factors may be additives varying from one commercial silicone to another as well as the type of filler used to reinforce the LSR. In this study we have limited ourselves to nonconductive particles and the picture may be different for LSR composites with conductive particles below the percolation threshold such as silicones with carbon nanotubes [25] and exfoliated graphite [26]. Furthermore the type of elastomer may also influence.
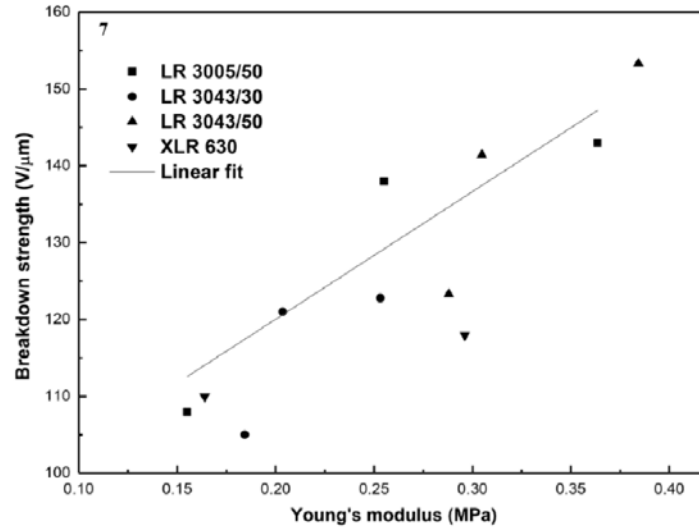

Figure 2. Correlation between Young's modulus (determined from the relation between the shear and elastic modulus) and the electrical breakdown strength for pure LSRs and the titanium oxide loaded composites. Data covers loadings up to 10 phr TiO_2 only. The line shown is a linear fit added as guideline to the eye.

3.2.1 High concentrations of fillers

In order to achieve even higher dielectric permittivities large concentrations of various types of titanium dioxides were applied. A custom-made LSR was applied where the amount of silica had been lowered and thereby the total solid content of the composites should be lowered. It was hypothesized that by lowering the total amount of solid fillers, the Mullin effect could be reduced while maintaining the favorable mechanical properties of the LSR. However, the Mullin effect was still encountered when large concentrations of the slightly incompatible titanium dioxides were applied. The first large-strain deformation caused irreversible extension of the elastomer films but after the first destructive cycle, the films could be strained reversibly with no further deformation as long as the initial applied strain was not exceeded.

A summary of high concentration results are shown in Table 2 and will be discussed in more detail in the following.

Table 2. Strain displacement and dielectric permittivity for various composites based on the low content silica LSR.

Composition	Strain displacement	Permittivity ε_r @ 0.1Hz / @ 1kHz
MJK 135 5/13 reference	0%	3.0 / 3.0
20% T805 + 29% OS20	1.0%	11.0 / 4.9
30% T805 + 38% OS20	2.7%	16.0 / 5.1
35% T805 + 45% OS20	3.9%	13.0 / 4.3
20% RM130F + 17% OS20	4.1%	6.6 / 4.9
30% RM130F + 30% OS20	6.9%	9.1 / 5.4
35% RM130F + 33% OS20	8.3%	8.0 / 4.7
40% RM130F + 34% OS20	6.8%	11.0 / 5.7
10% T805 + 10% RM130F + 23% OS20	4.4%	7.2 / 4.4
15% T805 + 15% RM130F + 30% OS20	6.8%	8.4 / 4.0
17.5% T805 + 17.5% RM130F + 39% OS20	6.8%	11.0 / 4.6
20% T805 + 20% RM130F + 43% OS20	8.1%	19.0 / 5.6

In Figure 3 the strain displacement of all the coatable samples is shown. The pure LSR formulation requires no solvent and possesses no measurable strain displacement. It is obvious that the data is very scattered which would also be expected since the concentration of fillers and the concentration of solvent are interdependent due to the experimental restriction put on the formulations to have as little solvent as possible. It is also clear that each type of filler behaves similarly but with different onsets for the strong dependency of strain displacement. This can also be explained from a network perspective since the upturn when lowering the network fraction (i.e. going from right to left in the figure) is expected due to the elasticity scaling with the network fraction to a complex power function diverging close to the network being destructed. In our case the network is being destructed simply by adding so much filler and solvent that the elasticity is lost.
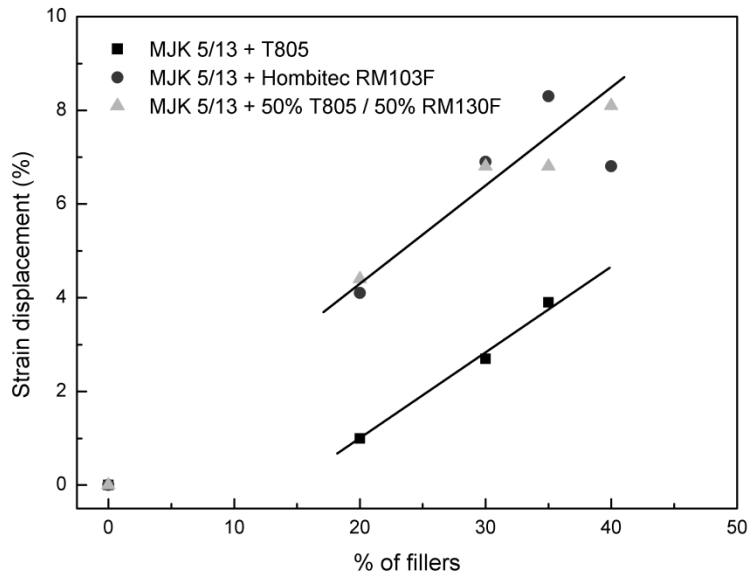

Figure 3. The strain displacement as function of filler loading of both types of filler and mixtures thereof (x-axis percentage of fillers added (phr)).

The LSR elastomer 5/13 allowed for easier processing of the highly loaded films but the breakdown strengths of the composites were significantly lower than that of the commercially available LSR elastomers (typically around 20 V/μm lower). Therefore the composites based on this particular LSR had too many drawbacks to be successful candidates for DEAP elastomers.

3.2.2 The influence of solvent on strain displacement

The strain displacement is furthermore strongly influenced by the amount of solvent applied in the coating process. A recipe of 16% Hombitec RM130F in XLR630 was applied as a recipe that requires solvent for coatability, and the amount of solvent applied to dilute the premixes was altered from 15-40 phr. 15 phr solvent was the minimum amount required for coatability.

Table 3. Strain displacement for composite consisting of 16% Hombitec RM130F in XLR630 with varying amounts of solvent applied in the coating process.

Solvent concentration[phr]	Strain displacement [%]
15	3.1
20	5.5
30	7.4
40	11.0

The results in Table 3 clearly indicate that the use of solvent in the coating process needs to be considered as it influences some of the important properties of the resulting elastomer films. The Young moduli were also slightly influenced with a decrease of Y with increased solvent amounts. This agrees well with the loss of tension in the films. The tear strengths were also considerably reduced at the large amounts of solvent. The dielectric permittivity was not influenced as expected since the solvent will be removed from the composites during the curing.

4. CONCLUSION

Composites of LSR and various types of TiO_2 have been shown to possess favorable properties for DEAP uses, mainly due to their high electrical breakdown strengths (up to 170 V/μm). The LSR composites do, however, possess high Young's moduli and hence are most suitable as elastomers for energy generation rather than actuation. The composites with less than 10phr suffer from lower energy densities than that of the highly loaded composites. However, the latter composites suffer from strain displacement, which is increased with the amount of solvent applied in the coating process. Solvent is also shown to influence the strain displacement negatively so it is important to minimize the use of solvent in the coating of these composites.

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