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Remarkable improvement of the electro-mechanical properties of polydimethylsiloxane elastomers through the combined usage of glycerol and pyridinium-based ionic liquids

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Remarkable improvement of the electro-mechanical properties of polydimethylsiloxane elastomers through the combined usage of glycerol and pyridinium-based ionic liquids

Abstract

The issue of high driving voltages limits the commercial application of dielectric elastomer transducers, but increasing dielectric permittivity and reducing the Young's modulus of elastomers is a means of lowering this driving voltage. However, dielectric elastomer fabrication, which fulfills simultaneously high dielectric permittivity and a low Young's modulus, is difficult. In this study, we explore the combined usage of ionic liquids and glycerol as soft fillers with high dielectric permittivity in silicone-based elastomers. We also fabricate a foamed structure in silicone elastomers to lower further the Young's modulus of the elastomers. Surprisingly, it is found that nitrogen atoms in the cations of ionic liquids (ILs) do not inhibit the platinum (Pt)-catalysed hydrosilation of silicone elastomers when ILs are dissolved in glycerol. The effect of molecular structure and the amount of ILs on the electrical breakdown strength, rheological, dielectric and mechanical properties of silicone-based elastomers is analysed herein. Compared to the pure elastomer, the combined usage of 5% 1-butyl-4-methylpyridinium tetrafluoroborate with 10% glycerol can improve the relative ratio of dielectric permittivity to the Young's modulus almost eleven-fold at a given electrical field below the electrical breakdown field.

Keywords: silicone elastomers; ionic liquids; glycerol; rheology; dielectric permittivity; electrical breakdown strength

1. Introduction

Dielectric elastomers (DEs) have gained a reputation as one of the most promising materials for electro-mechanical transducers, because these lightweight materials

produce linear motion rather than the rotary motion usually produced by heavy electric motors.[1-2] Dielectric elastomer-based actuators have many advantages, such as their ability to handle large strains, along with short response times, comparable to those of human muscle.[3-5]

The main issue that limits the application of this technology is low achievable strain below the breakdown field. Consequently, there is a need to optimise the formulation, in order to overcome the shortcomings of this technology. According to actuation Equation (1)[1], the thickness of strain (S_z) is correlated to driving voltage (V), relative dielectric permittivity (ϵ_r), Young's modulus (Y) and film thickness (d). Vacuum permittivity (ϵ_0) is a constant equal to approximately $8.854 \times 10^{-12} \text{ F m}^{-1}$.

$$S_z = \frac{\epsilon_0 \epsilon_r}{Y} \left(\frac{V}{d} \right)^2 \quad (1)$$

In order to increase the achievable strain in a relatively low electrical field at the given thickness, either dielectric permittivity should be enhanced or the Young's modulus should be decreased.[1] Equation (1) was further elaborated into a figure of merits (F_{om}) for actuators and generators by Sommer-Larsen and Larsen and McKay et al., respectively.[6-7] Equations (2) and (3) show the figure of merits for actuation (DEA) as well as for elastomer energy generation (DEG):

$$F_{om}(\text{DEA}) = \frac{3\epsilon_r \epsilon_0 E_{BD}^2}{Y} \quad (2)$$

$$F_{om}(\text{DEG}) = \frac{\epsilon_r \epsilon_0 E_{BD}^2}{2\phi} \quad (3)$$

where E_{BD} represents the electrical breakdown strength of the elastomer, and ϕ represents the energy density function of the given elastomer. In a relative comparison, it is usually regarded as a constant for most silicone elastomers, due to the scarcity of available data.[8]

In practical uses, the voltage applied to transducers is always below the electrical breakdown strength of the elastomers. Thus, the figure of merit for actuators in Equation (2) is simplified to Equation (4), in which the relative ratio of dielectric permittivity to the Young's modulus is used to evaluate the improved actuation strain at a given electrical field below the electrical breakdown field of elastomer composites.[9]

$$F'_{om}(DEA) = \frac{\epsilon}{Y} / \frac{\epsilon_{ref}}{Y_{ref}} \quad (4)$$

where ϵ_{ref} and Y_{ref} stand for dielectric permittivity and the Young's modulus of the reference materials, respectively, and ϵ and Y stand for dielectric permittivity and the Young's modulus of the tested materials, respectively.

The materials used most commonly to prepare dielectric transducers are acrylic-based, silicone-based, polyurethane-based and ethylene propylene-based elastomers.[10-13] Among these examples, silicone elastomers are chosen most often to fabricate transducers, because they have a long lifetime, excellent durability and high breakdown strength.[14]

In order to improve the $F_{om}(DEA)$, studies have focused on increasing the dielectric permittivity of elastomers by adding fillers with high dielectric permittivity, such as barium titanate, zinc oxide, carbon nanotubes, carbon blacks and graphites.[15-20] Although such rigid fillers can increase dielectric permittivity, the Young's modulus of composites is always increased at the same time, except if fillers are incompatible with the matrix.[16] Increased dielectric permittivity can in turn increase actuation strain, while an increased Young's modulus can in fact decrease actuation strain. In this way, any improvements in actuation strain are minimal.[18] Furthermore, inorganic fillers tend to aggregate in the polymer matrix, which then increases stress concentration points and reduces the mechanical properties of composites.[19] Therefore, in order to improve the performance of actuators, it is important to increase dielectric permittivity and reduce Young's modulus simultaneously.[21]

The addition of soft or liquid fillers with high permittivity into silicone elastomers can lower the resulting Young's modulus. It has been reported that silicone/glycerol elastomers have been prepared with glycerol droplets homogeneously dispersed in silicone as soft fillers.[22] However, only high amounts of glycerol can significantly increase dielectric permittivity and reduce the Young's modulus of the obtained elastomers.[23]

Apart from glycerol, ionic liquids (ILs) with high dielectric permittivity are also candidates for soft fillers.[24] Ionic liquids are molten salts which are in a liquid state

near room temperature and consist entirely of anions and cations.[25] In ionic liquids, the interaction force between anions and cations is coulombic in nature, the magnitude of which is correlated to the radii of the anions and cations.[26]

These non-volatile ionic liquids have the advantages of low melting points, excellent thermal conductivity, high stability and selective solubility.[27] However, the majority of ionic liquids commonly inhibit the platinum (Pt)-catalysed cross-linking reaction (hydrosilation) of silicone elastomers,[28] and so it is very important to choose the right ionic liquid, in order to avoid this issue. To date, there are no reports regarding the addition of pyridinium-based ionic liquids in Pt-catalysed cross-linked silicone elastomers.

The aim of this work is to fabricate a new type of soft silicone-based elastomer with high dielectric permittivity and a low Young's modulus. Instead of using solid fillers, this work is based on a foamed silicone elastomer containing glycerol and ionic liquids as soft fillers. We report herein on two pyridinium-type ionic liquids which do not inhibit the platinum (Pt)-catalysed hydrosilation of silicone elastomers when ILs are dissolved in glycerol. The addition of ionic liquid and glycerol allows for the rise of dielectric permittivity and a decrease in the Young's modulus of elastomer composites. Furthermore, the foamed structure can also contribute to the reduction of the Young's modulus. The effect of molecular structure and the amounts of ILs on the electrical breakdown strength, rheological, dielectric and mechanical properties of foamed silicone composites is investigated. Moreover, the effect of the separate and combined usage of ILs and glycerol on the electro-mechanical properties of silicone composites is evaluated by a figure of merits and the relative ratio of dielectric permittivity to the Young's modulus. We also determine the optimal formulation of the silicone-based elastomer with a significant improvement in the relative ratio of dielectric permittivity to the Young's modulus.

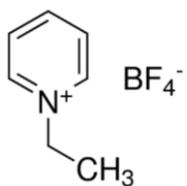
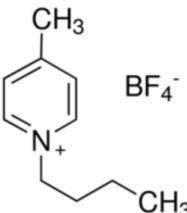
2. Experimental

2.1 Materials

The silicone elastomer used in this study was the two-component Sylgard 184

silicone (abbreviated as S184), consisting of polymer base (vinyl-functional PDMS and platinum catalyst) and curing agent (vinyl-functional PDMS and crosslinker), purchased from Dow Corning, US. Glycerol (abbreviated as gly) was provided by Emmelev A/S, Denmark. Sodium hydroxide (NaOH), 1-ethylpyridinium tetrafluoroborate (abbreviated as ILA) and 1-butyl-4-methylpyridinium tetrafluoroborate (abbreviated as ILB) were purchased from Sigma Aldrich. The ILA and ILB are solid and liquid at 25°C, respectively. The molecular structures of ILA and ILB are listed in Table 1.

Table 1. Structures of the two ionic liquids used herein.

	1-ethylpyridinium tetrafluoroborate
	1-butyl-4-methylpyridinium tetrafluoroborate

2.2 Sample preparation

Glycerol with 0.8wt% NaOH was first prepared by dissolving 0.8g NaOH in 99.2g glycerol. Then the obtained glycerol, ILA (or ILB), curing agent and S184 polymer base were mixed according to the formulations illustrated in Table 2. The loading of glycerol-NaOH solution into the silicone elastomer composites was kept constant at 11.2 phr.

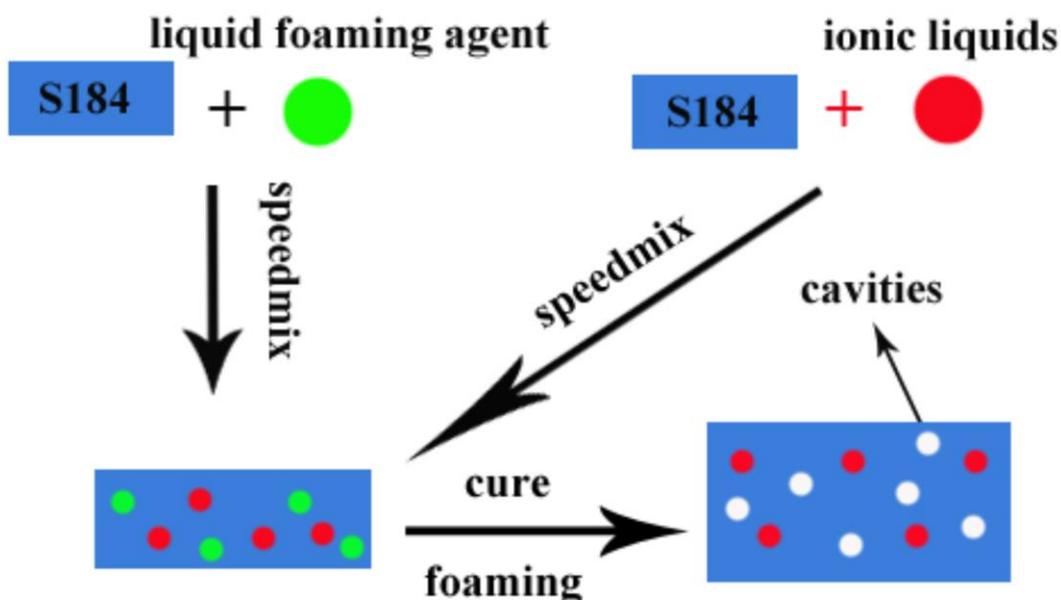
The process employed to fabricate silicone foams with ionic liquids is illustrated in Scheme 1. 50 phr S184 with liquid foaming agents and 50 phr S184 with ionic liquids were mixed separately by a SpeedMixerTM DAC 150 FVZ-K at 3500 rpm for 5 min, following which the two obtained mixes were mixed at 3000 rpm for 1 min.

The mixtures were then coated onto a glass substrate by an Elcometer 3540 film applicator with a 200 μm blade. The films were cured in an oven at 120°C for 1 h. The obtained mixtures were also cast onto a mould with a 1 mm-thick spacer and cured at 120°C for 1 h. The 1 mm-thick sample was then cut into a disc with a diameter of 20 mm.

Table 2. Formulation of silicone with ionic liquids (phr*).

Sample No.	Sample name	S184 polymer base	Curing agent	Glycerol with NaOH	ILA	ILB
1	S184	100	12	-	-	-
2	S184-gly	100	12	11.2	-	-
3	S184-gly-ILA-1.25%	100	12	11.2	1.40	
4	S184-gly-ILA-2.5%	100	12	11.2	2.80	-
5	S184-gly-ILA-3.75%	100	12	11.2	4.20	-
6	S184-gly-ILA-5%	100	12	11.2	5.60	-
7	S184-gly-ILB-1.25%	100	12	11.2	-	1.40
8	S184-gly-ILB-2.5%	100	12	11.2	-	2.80
9	S184-gly-ILB-3.75%	100	12	11.2	-	4.20
10	S184-gly-ILB-5%	100	12	11.2	-	5.60
11	S184-ILB-5%	100	12	-	-	5.60

*phr= parts per hundreds of polymer base and curing agent.



Scheme 1. Mixing silicone foams with ionic liquids at room temperature and curing the obtained mixes at 120°C.

2.3 Characterisation

2.3.1 Rheological measurements

Rheological properties of the prepared silicone films were determined with a TA Instruments Discovery HR-1 Rheometer with a 2% controlled strain within the linear viscoelastic regime. Samples 20 mm in diameter and 1 mm-thick were placed between two parallel plates at 25°C, with a normal force of 7 N and in the frequency range 0.1 ~ 100 Hz.

2.3.2 Dielectric properties and electric conductivity testing

According to IEC 60250, the dielectric properties and electric conductivity of the silicone films were tested on a Novocontrol Alpha-A high-performance frequency analyser in about a 1 V/mm electrical field, in the frequency range 10^{-1} to 10^6 Hz at 25°C. The diameter and thickness of the tested samples were 20 mm and 1 mm, respectively.

2.3.3 Tensile strength and Young's modulus measurements

The tensile strength of the silicone films was tested by a material tester (Instron 3340 materials testing system, INSTRON, US) at 25°C. The sample of 60 mm length, 6 mm width and 0.1 mm thick was placed between two clamps and initially separated by a distance of 30 mm. The testing rate was 10 mm/min. Young's moduli were obtained from the tangent of the stress-strain curves at 5% strain. Each composition was taken at least three measurements which were then averaged.

2.3.4 Optical microscopy observation

The morphology of the cross-section of silicone films, before and after 72 h immersion in water, was observed by DMLB-type optical microscopy from Leica Microsystem, Germany. One end of the 0.1 mm-thick, 6 mm-wide and 15 mm-long film was fixed to the bottom of a plastic cup to ensure the immersion of samples in 20mL deionised water for 72 h. The samples were then dried in an oven for 24 h at 80°C. The water was changed every 12 h.

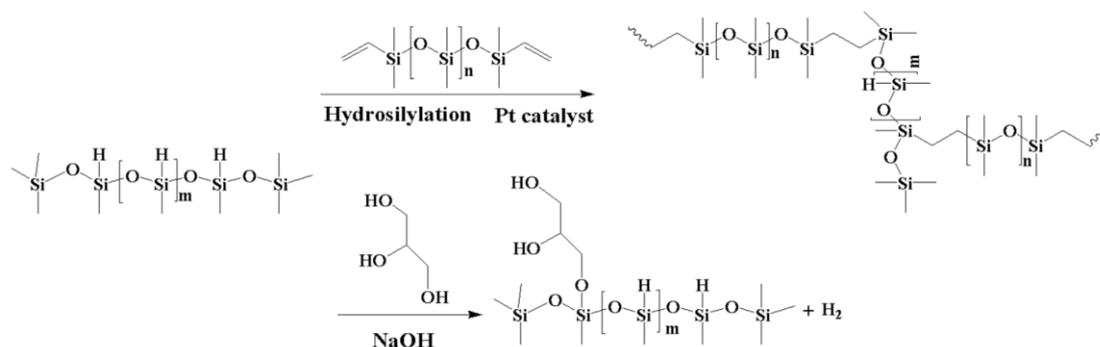
2.3.5 Electrical breakdown strength measurement

According to IEC 60243-2 (2001), electrical breakdown strengths of the developed elastomers were tested by an in-house-built device by increasing voltage (50 ~ 100 V per step) at a rate of 0.5 ~ 1 steps/s. The cross-sectional thicknesses of samples were measured by optical microscopy. The distance between the spherical electrodes was set to 95% of the obtained thickness, in order to ensure the electrodes made contact with the samples, each of which was tested 12 times to obtain average electrical breakdown strength values.

3. Results and discussion

Foamed S184-gly-ILs elastomer composites were prepared according to Scheme 2, in order to lower the Young's modulus of silicone-based elastomers. The silicone hydrides reacted with glycerol in the presence of sodium hydroxide (NaOH) to generate

hydrogen gas, thus producing bubbles.[29] Meanwhile, silicone hydrides also reacted with the vinyl groups of silicones to form a cross-linked network.[30]



Scheme 2. Reaction of silicone hydrides with vinyl groups of silicones and NaOH.

3.1 Rheological properties

The effect of the concentration and type of ionic liquid on the storage modulus (G') and loss factor ($\tan\delta$) of S184, S184-gly, S184-ILB-5% and S184-gly-ILs was determined by a rheometer. The results are shown in Figure 1 and Figure 2. As evident from Figure 1, the addition of 11.2 phr glycerol lowers the storage modulus of S184, because glycerol is a liquid and can be regarded as a soft filler. Furthermore, the addition of ILA and ILB increases and decreases, respectively, the storage modulus of S184-gly, because ILA (1-ethylpyridinium tetrafluoroborate) and ILB (1-butyl-4-methylpyridinium tetrafluoroborate) are in a solid and a liquid state, respectively, at the testing temperature used for rheological properties. During the rheological test, the solid ILA can withstand more external force than elastomer matrix, while the liquid ILB cannot sustain that force, leading to their different contribution to the storage modulus of elastomer matrix.[31-32]

It is evident from Figure 2 that the loss factors ($\tan\delta$) of S184, S184-gly, S184-ILB-5% and S184-gly-ILs are all lower than 0.1 at 10^{-1} Hz, indicating the good cross-linked structure of silicone elastomers.[33] Generally, the majority of ionic liquids containing nitrogen inhibit the Pt-catalysed curing reaction.[34] However, in this work,

the $\tan\delta$ of S184-ILB-5% is lower than 0.1 at 10^{-1} Hz, indicating that ILB with pyridinium structure do not inhibit the Pt-catalysed cross-linking reaction of silicone elastomers to a significant extent. The S184-ILA without glycerol has not been tested due to the complete immiscibility between solid ILA and liquid S184.

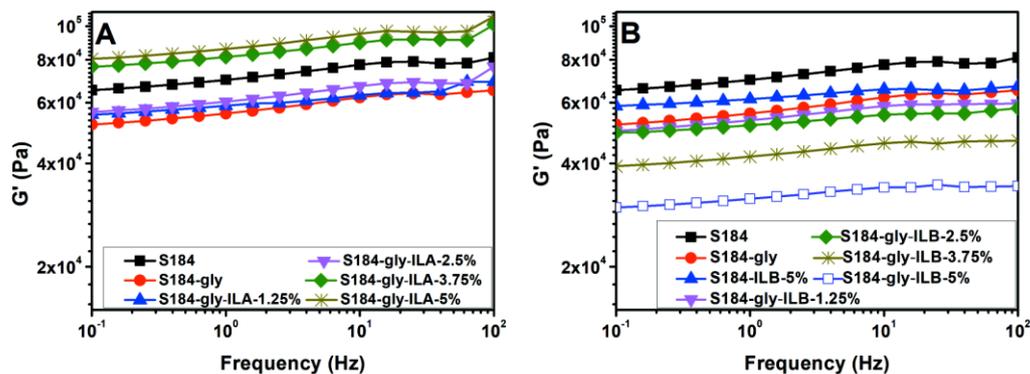


Figure 1. Storage modulus of silicone with ionic liquids determined at 2% strain and room temperature.

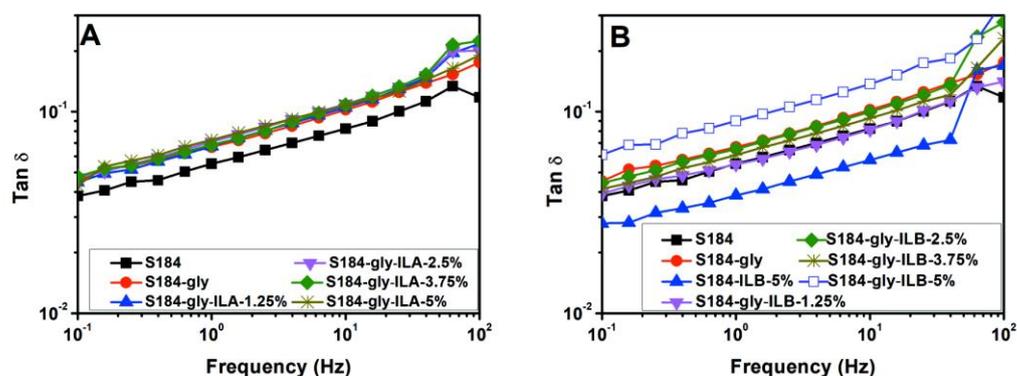


Figure 2. Loss factor ($\tan\delta$) of silicone with ionic liquids determined at 2% strain and room temperature.

3.2 Optical microscope images

The morphologies of S184, S184-gly, S184-gly-ILA-5% and S184-gly-ILB-5%, before and after immersion in water, were investigated by optical microscopy and are shown in Figure 3. The morphologies of S184-gly with different amounts of ILA and ILB are listed in Figure S1-S4 in ESI. The solid ILA is observed as particles in Figure 3D. Due to the brown colour of ILB, droplets thereof are observed in Figure 3F. There

is no obvious agglomeration of 5% solid ILA and 5% liquid ILB observed in Figures 3D and 3F, indicating the homogeneous dispersion of ILA and ILB in S184-gly.

It is evident in Figure 3C that after being immersed in water and dried, micro-sized cells are still separated in the elastomer. According to Scheme 2, the silicone hydrides react with glycerol in the presence of sodium hydroxide (NaOH) to generate hydrogen gas, which produces cells.[29] Meanwhile, silicone hydrides also react with the vinyl groups of silicones to form a cross-linked network.[30] Furthermore, after S184-gly with ILA and ILB have been immersed in water for 72 h, there are still residues of ILA and ILB observed in the matrix, because both open and closed cells have formed in the silicone matrix. The ILA and ILB in interconnected open cells can be washed away by water, while this cannot be done for those in closed cells.

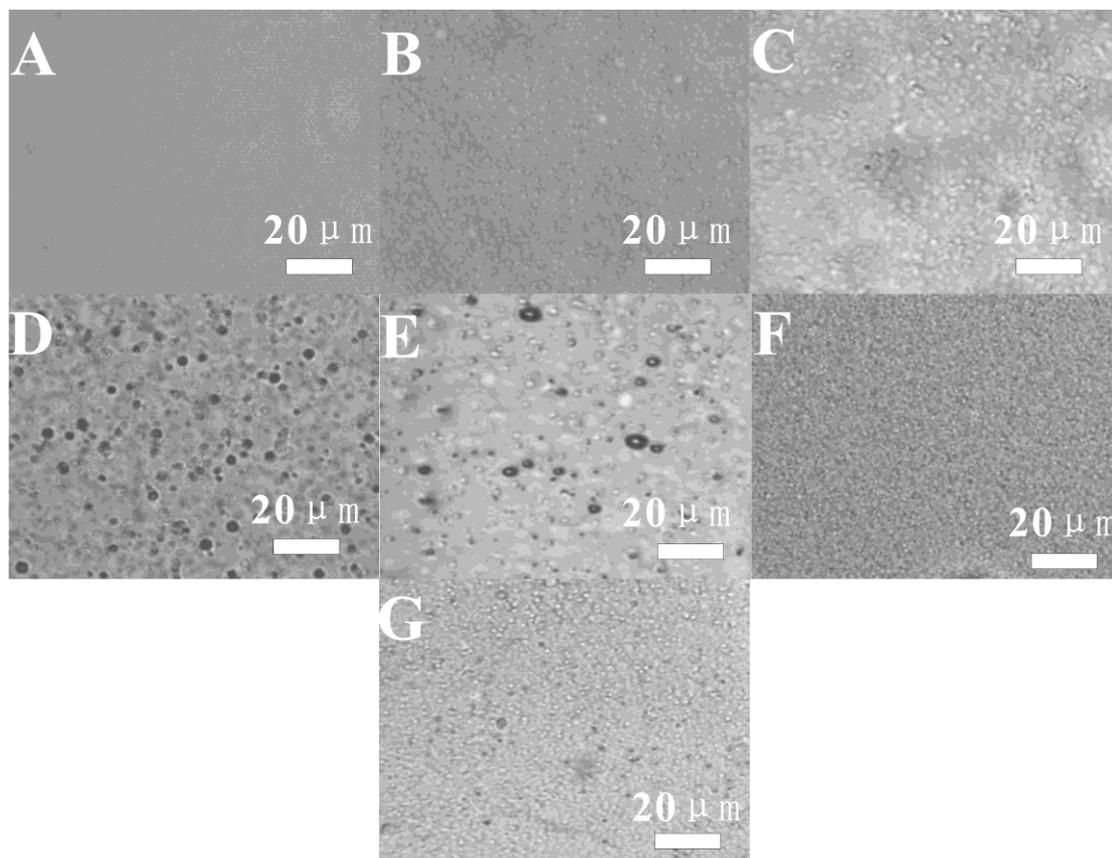


Figure 3. Optical microscope images of S184 elastomers, before and after immersion: A. S184; B. S184-gly; C. S184-gly-after immersion; D. S184-gly-ILA-5%; E. S184-gly-ILA-5% after immersion; F. S184-gly-ILB-5%; G. S184-gly-ILB-5% after immersion.

3.3 Dielectric properties and conductivity

The relative dielectric permittivity and dielectric loss tangents ($\tan\delta=\varepsilon''/\varepsilon'$) of S184, S184-gly, S184-ILB-5% and S184-gly-ILs were determined through dielectric spectroscopy, the results for which are illustrated in Figure 4 and Figure 5, respectively. With increased amounts of ILA and ILB, the dielectric permittivities of S184-gly composites are enhanced. In the range 10^{-1} to 10^6 Hz, 5% ILA and ILB increase the relative dielectric permittivity of S184-gly from 3.2 to 4.5 and 5.2 at 10^{-1} Hz, respectively, since the asymmetrical structure of cations endows ionic liquids with polarity.[35] This shows that ILB, aligned with cations with larger radii, possesses higher polarity than ILA, and so in this way, the dielectric permittivity of S184-gly-ILB is higher than that of S184-gly-ILA.

As for S184-gly and S184-ILB-5%, the separate addition of 10% glycerol and 5% ILB can increase the dielectric permittivity of S184 from 2.6 to 3.2 and 2.9 at 10^{-1} Hz, respectively. In comparison, in terms of S184-gly-ILB-5%, the combined usage of 10% glycerol and 5% ILB enhances the dielectric permittivity of S184 from 2.6 to 5.2 (100% increase) at 10^{-1} Hz, as the presence of glycerol lengthens the distance between ILB cations and anions. In this way, the interaction between cations and anions is weakened and the alignment of ILB in the excited electrical field is enhanced since the backbone of silicone consists of silicon-oxygen bonds which are not affected by ILB and glycerol. Thereby, the increased dielectric permittivity is ascribed to the combined usage of ILB and glycerol.[36-37] According to Equation (2), the 100% increase in dielectric permittivity of S184-gly-ILB-5% is beneficial, in that it increases the $F_{om}(DEA)$ of transducers. Variations in dielectric losses are presented in Figure 5. It can be observed that dielectric loss increases with increasing IL loading. Moreover, the loss tangent of pure S184 is almost frequency-independent, while the values of S184-ILs and S184-gly-ILs increase in line with decreasing frequency, which is an evidence of dipole relaxations of ionic liquids and glycerol. It is worth noting that the dielectric losses of S184-gly-ILs are lower than S184-gly, indicating that the ionic liquids might act as inhibitors for the energy dissipation caused by glycerol. Consequently, the level of all

the elastomers is still acceptable (generally below 1%) for the DEs applications.

In addition, the electric conductivities of S184, S184-gly, S184-ILB-5% and S184-gly-ILs are shown in Figure 6 and Table 3. With the increased amount of ionic liquids, the amounts of cations and anions are increased, which can facilitate the transfer of electrons. In this way, the increased content of ILA and ILB increases the electric conductivity of S184-gly. Moreover, at the same concentration, the conductivity of S184-gly-ILB is a little higher than that of S184-gly-ILA, which is ascribed to the fact that the large radii of ILB cations makes them, and anions, move easier than ILA, which facilitates the transfer of charges.

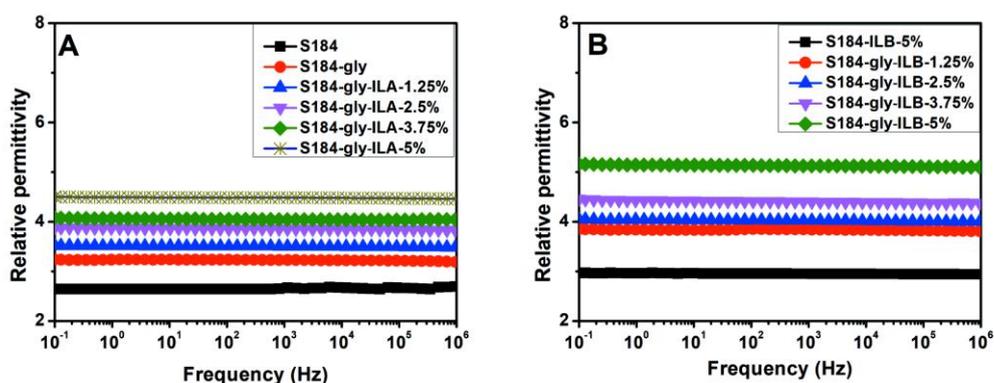


Figure 4. Dielectric permittivity of S184-gly with varying ionic liquid loads at room temperature.

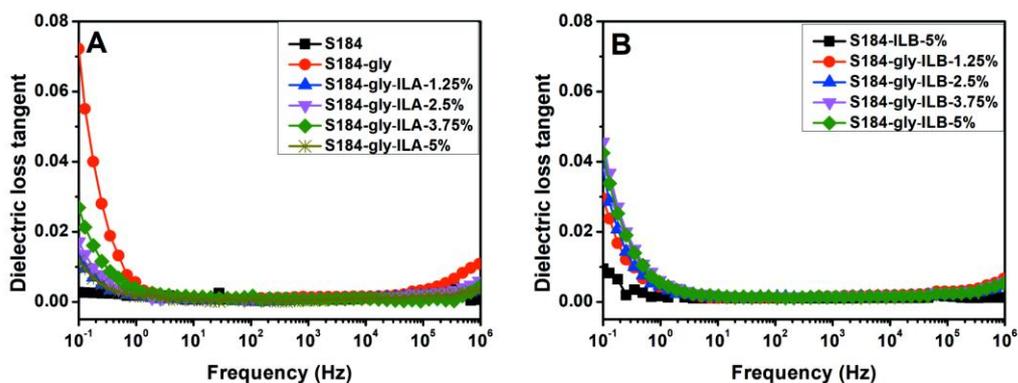


Figure 5. Dielectric loss tangent of S184-gly with varying ionic liquid loads at room temperature.

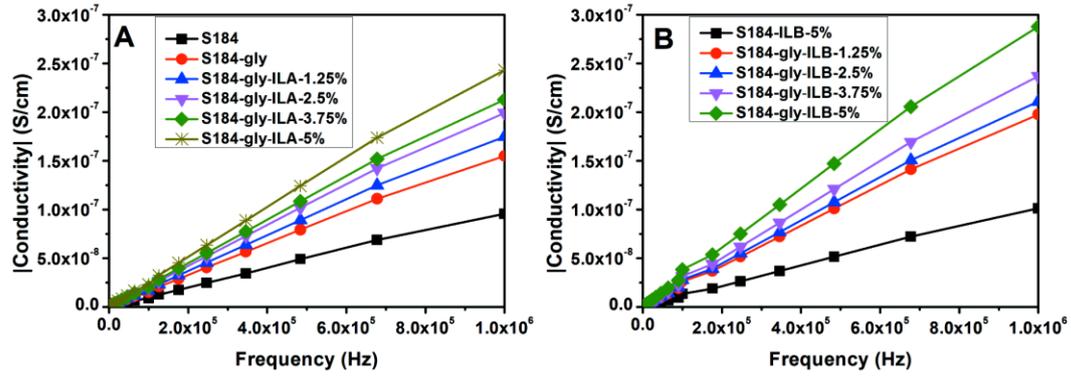


Figure 6. Electric conductivity of S184-gly with varying ionic liquid loads at room temperature.

Table 3. Electrical conductivity of S184-gly with varying ionic liquid loads at room temperature.

Sample name	$ \sigma $ (S/cm)	
	0.1 Hz	1 MHz
S184	5.90×10^{-14}	9.57×10^{-8}
S184-gly	1.25×10^{-13}	1.55×10^{-7}
S184-gly-ILA-1.25%	1.40×10^{-13}	1.74×10^{-7}
S184-gly-ILA-2.5%	1.60×10^{-13}	1.99×10^{-7}
S184-gly-ILA-3.75%	1.70×10^{-13}	2.12×10^{-7}
S184-gly-ILA-5%	1.90×10^{-13}	2.43×10^{-7}
S184-gly-ILB-1.25%	1.58×10^{-13}	1.97×10^{-7}
S184-gly-ILB-2.5%	1.69×10^{-13}	2.10×10^{-7}
S184-gly-ILB-3.75%	1.92×10^{-13}	2.36×10^{-7}
S184-gly-ILB-5%	2.31×10^{-13}	2.87×10^{-7}
S184-ILB-5%	8.18×10^{-14}	1.01×10^{-7}

3.4 Mechanical properties and electrical breakdown

The Young's modulus, tensile stress and strain at break of S184, S184-gly, S184-

ILB-5% and S184-gly-ILs were determined through extensional experiments. The Young's modulus at 5% was used herein, and the resulting data are listed in Table 4. The addition of glycerol and ILB decreases the Young's modulus of S184, while the addition of ILA increases the Young modulus of S184-gly. The reason for this behavior is that during the mechanical test, the solid ILA can withstand more external force than elastomer matrix and increases the stiffness of S184-gly. By comparison, glycerol and liquid ILB are regarded as soft fillers and cannot withstand mechanical stress, leading to the decreased stiffness of elastomers. Compared to S184, the decreased Young's modulus of S184-gly-ILA-5% is favourable for actuation.

In terms of tensile strain, the 5% ILA increases the stress concentration points in S184-gly with ILA and decreases the strain at break of S184-gly. By comparison, the 5% liquid ILB has no such effect, and it increases the strain at break of S184-gly.

The electrical breakdown strength, figures of merit and relative ratios of dielectric permittivity (ϵ) to the Young's modulus (Y) for S184, S184-gly, S184-ILB-5% and S184-gly-ILs are listed in Table 5. As evident from Table 5, the addition of glycerol, ILA and ILB decreases electrical breakdown strength compared to the reference film S184, thereby establishing that the use of glycerol, ILA and ILB as fillers for DEs compromises the dielectric breakdown strength of the films. [38]

Furthermore, only S184-gly, S184-gly-ILA-1.25% and S184-gly-ILB-1.25% exhibit higher figures of merit for actuation than the S184 control. Among them, the largest figure of merit was achieved for S184-gly at 50% higher than S184. Although the further addition of ionic liquids can increase dielectric permittivity, the E_{BD} of elastomers is reduced at the same time, leading to a decrease in figures of merits. Moreover, the addition of glycerol and ionic liquids reduces figures of merit for generation, albeit this is expected, since energy generation favours stiff materials.[39]

As evident from Table 5, the addition of 5% ILB can increase the relative ratio ($\frac{\epsilon_r}{Y} / \frac{\epsilon_{ref}}{Y_{ref}}$) to 1.56, because the 5% liquid ILB can soften the S184 and increase dielectric permittivity at the same time. By comparison, 10% liquid glycerol not only softens the S184, but also produces a microporous structure, both of which decrease the Young's

modulus of S184. Moreover, 10% glycerol endows S184 with higher dielectric permittivity than 5% ILB. Thereby, the addition of 10% glycerol increases the relative ratio ($\frac{\epsilon_r}{Y} / \frac{\epsilon_{ref}}{Y_{ref}}$) to 3.44.

With the increased amount of ILs, the relative ratio ($\frac{\epsilon_r}{Y} / \frac{\epsilon_{ref}}{Y_{ref}}$) of S184-gly-ILs increases in all cases. Furthermore, the ILB can enhance the relative ratio more remarkably than ILA, due to its softening effect and high dielectric permittivity. It was determined that the combined usage of 10% glycerol and 5% ILB can boost the relative ratio of S184 to 10.57, which is almost an eleven-fold larger actuation at a given field below the electrical breakdown strength.

Table 4. Mechanical properties of S184-gly with ionic liquids.

Sample name	Young's modulus @ 5% MPa	Tensile stress MPa	Strain at break %
S184	3.7±0.3	10.9±0.4	105±13
S184-gly	1.5±0.1	5.6±0.2	180±17
S184-gly-ILA-1.25%	1.6±0.1	5.8±0.2	167±21
S184-gly-ILA-2.5%	1.7±0.1	5.1±0.3	147±24
S184-gly-ILA-3.75%	1.8±0.1	4.3±0.2	132±15
S184-gly-ILA-5%	1.9±0.1	4.1±0.2	107±11
S184-gly-ILB-1.25%	1.1±0.1	3.5±0.2	197±19
S184-gly-ILB-2.5%	1.0±0.1	2.9±0.2	239±27
S184-gly-ILB-3.75%	0.9±0.1	2.9±0.1	261±25
S184-gly-ILB-5%	0.7±0.1	2.6±0.2	302±31
S184-ILB-5%	2.8±0.2	7.9±0.2	163±18

Table 5. Electrical breakdown strengths and figures of merit for the S184-gly with varying ionic liquid loads at room temperature.

Sample name	E_{BD} V/ μm	Thickness μm	$\frac{F_{om}(DEA)}{F_{om}(DEA)_{pure}}$ *	$\frac{F_{om}(DEG)}{F_{om}(DEG)_{pure}}$ *	$\frac{\epsilon/Y}{\epsilon_{ref}/Y_{ref}}$ *
S184	120 \pm 3	120	1.00	1.00	1.00
S184-gly	80 \pm 2	122	1.53	0.62	3.44
S184-gly-ILA- 1.25%	72 \pm 2	110	1.27	0.55	3.53
S184-gly-ILA- 2.5%	46 \pm 2	130	0.53	0.24	3.61
S184-gly-ILA- 3.75%	35 \pm 2	141	0.30	0.15	3.58
S184-gly-ILA-5%	22 \pm 1	133	0.13	0.07	3.82
S184-gly-ILB- 1.25%	56 \pm 2	124	1.21	0.36	4.92
S184-gly-ILB- 2.5%	37 \pm 2	131	0.61	0.17	5.69
S184-gly-ILB- 3.75%	32 \pm 2	122	0.57	0.14	6.69
S184-gly-ILB-5%	21 \pm 1	111	0.36	0.07	10.57
S184-ILB-5%	35 \pm 2	113	0.13	0.10	1.47

*The figures of merit and ϵ/Y are normalised with respect to S184.

4. Conclusions

A new foamed dielectric elastomer with high dielectric permittivity and a low Young's modulus was prepared through the combined usage of glycerol and ILB as soft fillers. It was somewhat surprising to find that S184 cures well in the presence of 5% ILs, indicating that ionic liquids with a pyridinium structure do not inhibit to a significant extent the Pt-catalysed cross-linking reaction of silicone elastomers.

Based on a relative ratio of dielectric permittivity to Young's modulus, improved overall actuation performance could be obtained for S184-gly-ILB-5%, because the combined usage of 10% glycerol and 5% ILB not only provides high dielectric permittivity, but also lowers the Young's modulus of the reference S184 film. Meanwhile, the formed structure was prepared to reduce further the Young's modulus of S184. The increased dielectric permittivity and decreased Young's modulus of S184-gly-ILB-5% improved the relative ratio ($\frac{\epsilon_r}{Y} / \frac{\epsilon_{ref}}{Y_{ref}}$) of S184-gly-ILB almost eleven-fold, compared to the pristine silicone elastomer at a given electrical field below the electrical breakdown strength. By comparison, there was very little improvement in figures of merit, because the effect of decreased E_{BD} counteracted the increased relative ratio ($\frac{\epsilon_r}{Y} / \frac{\epsilon_{ref}}{Y_{ref}}$).

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