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Molecular strategies for improved dielectric elastomer electrical breakdown strengths

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Keywords: soft transducer, dielectric elastomer, silicone, electrical stability

Abstract:

Dielectric elastomer transducers (actuators and generators) possess great commercial potential since they allow for novel transducer designs and applications due to – amongst others – their flexibility and low weight. On the other hand, the flexibility and inherent softness of dielectric elastomers also put restrictions upon their use, since the thin elastomers may undergo destructive deformations under large loads or in large electrical fields. In order to design better dielectric elastomers it is crucial to understand the underlying phenomena of how thin and elastic dielectric elastomer films undergo electrical breakdown. This understanding will allow for design of dielectric elastomers with high electrical breakdown strength and thus open up for use of films in transducers at higher electrical fields and forces. Here, the study couples intrinsic electrical breakdown strengths with well-described polymer and network characteristics, namely Kuhn parameters and cross-linking density. The universality of the developed model is illustrated by comparison over a wide range of silicone-based elastomers, such as pre-stretched elastomers and synthesized cross-linked bottlebrush polymers, representing both filled and unfilled elastomers. This study paves a robust way for the molecular design of elastomers into high intrinsic electrical breakdown strength dielectric elastomers.

Dielectric elastomers are competitive electro-mechanical transducers, due to their favourable combination of low weight, inherent softness and compliance with, for example, the human body – and thus for soft robotic applications amongst many others.^[1,2] Nevertheless, the underlying physics behind the intrinsic electrical breakdown field of dielectric elastomers is not yet fully understood. The electrical breakdown strength of elastomer represents the upper limit for some dielectric elastomer applications, due to the premature failure and poor reliability of polymer films instantaneously or over time. In the early stages of dielectric elastomer exploitation, it was found that pre-stretching dielectric films enhanced the ultimate properties to a very large extent.^[3,4] For the commercially available VHB acrylic-based adhesive (3M), pre-stretching is required in order to achieve the reported large actuations of several 100% strains,^[3,4] which can be explained partly by the shape of the stress-strain curve. Pre-stretching ensures that the elastomer is in its softest state, due to the initial stress-softening effect. Subsequent strong stress hardening then ensures that there is no snap-through or electro-mechanical instability.^[5,6] However, the pre-stretching of VHB leads to transient properties due to subsequent stress relaxation and thus product reliability issues. For silicone elastomers, pre-stretching is also very favourable with respect to electrical breakdown strength, but this cannot be explained by the shape of the stress-strain curve, which usually is almost linear.^[2,7-10] Furthermore, silicone elastomers are almost exempt from any stress-induced anisotropy for the investigated strains, so the favourable electrical properties of the stretched elastomer cannot be explained by anisotropy.^[11] Therefore, it is counterintuitive that silicone elastomers should benefit electrically from pre-stretching. Various explanations for why pre-stretching is favourable have been proposed, such as the elimination of air voids and imperfections along with the decreased thickness – and thus a decreased probability of defects – within the given measurement volume.^[7] Moreover, experimental artefacts have been discussed as a means of making these observations, but the current standards for dielectric elastomers should ensure that reliable measurements are achieved as long as experiments are

conducted carefully.^[12] However, these reasonable explanations are not sufficient to describe the observed improved electrical properties of the order twice the level of initial electrical breakdown strength with 50% biaxial pre-stretching for most silicone elastomers, as illustrated in **Figure 1A**. In addition, the influence of the Young's modulus on electrical breakdown strength has been discussed by various authors, but results are neither ubiquitous nor coherent when compared across various silicone elastomer formulations (**Figure 1B**).^[10,13,14] To a large extent, this is understandable, since silicone elastomers come in many shapes and forms, with and without fillers and oils,^[13] with varying cross-linking density^[15] and with varying substitutions along the siloxane backbone.^[16] As a result, there is a great degree of complexity across the whole range of elastomers, though the underlying physics behind electrical breakdown must be identical for at least some of the formulations, such as the silicone elastomers filled with silica only or with other non-conductive fillers. For elastomer formulations with conductive fillers, other effects may contribute to electrical breakdown, such as space charges and local field enhancement.^[17-20]

Alternative explanations to why pre-stretching is favourable are that the dielectric constant of the elastomer varies with deformation and depends on the sort of pre-stretching being utilised. Unfortunately, there is conflicting information in the literature as to whether the dielectric constant of VHB materials depends on pre-stretching. Kofod et al., for instance, reported that the dielectric constant only slightly dropped with increased pre-stretching,^[22] but another study concluded that it decreases from its nominal value of 4.7 to 2.6.^[23] For silicone elastomers we have found no indications of dielectric permittivity stretch dependency for a wide range of investigated silicone elastomers. Yet another possibility is that in stretching the elastomer, the vibrational amplitudes of the individual molecules are reduced and consequently the activation volume is decreased for the process in which mobile charges can migrate under the influence of an electrical field. Such an activated state description applies to the electrical breakdown of many amorphous polymers, such as linear polyethylene and

polypropylene.^[24] However, this type of thermally activated breakdown is usually applicable to breakdown under a sustained electrical field rather than short ramping conditions such as those for which electrical breakdown strengths are measured.^[23] The irrelevance of thermal breakdown for common, single-layer thin film silicone-based dielectric elastomers, based on non-conductive fillers, has been confirmed by a series of non-isothermal breakdown measurements in which deviations in electrical breakdown strengths with increasing surrounding temperatures were within experimental uncertainty.

Understanding the underlying mechanisms of dielectric elastomer electrical breakdown is necessary for designing elastomers capable of operating reliably at high voltages and thereby realising their full potential. Proceeding to the molecular level – despite the complexity of silicone elastomer formulations – is obviously required to facilitate a design guide for dielectric elastomers with improved electrical properties. Silicone polymers are characterised by very long chains of the repeating unit $[-\text{Si}(\text{R}_1\text{R}_2)_2\text{O}-]$, where R_1 and R_2 are different or identical groups are attached to the silicon atom. The most common type of silicone is polydimethyl siloxane (PDMS), for which $\text{R}_1=\text{R}_2=\text{methyl}$. Each Si-O bond has its own electronic state. Due to differences in the electronegativity of Si and O, the monomer orbitals will degenerate slightly. These monomer molecular orbitals form a series of extended electronic states, i.e. energy bands, within the molecule, which extend along the polymer backbone in what is usually denoted as ‘anisotropic mobility’. Generally, the inter-molecular forces between apolar polymers are limited to van der Waals forces, and thus there is no overlap of molecular orbitals between polymers, only along the backbone.^[24] In other words, if this statement holds, electrical breakdown will take place via the shortest path of cross-linked polymers through the elastomer film, in which case the maximum voltage of defect-free films can be written as:

$$V_B = c_1 N_{b,min} \quad (1)$$

where c_1 is a constant describing the maximum sustainable voltage per bond and $N_{b,min}$ is the number of Si-O bonds taking the shortest path through the film along the polysiloxane backbone.

The shortest path of cross-linked polymers through the elastomer film should preferably be described by molecular simulation of the specific polymer network, but by assuming that the cross-linker is of high functionality and that the extent of reaction is large, the determination can be simplified to be independent of functionality and extent of reaction.^[25] More details on these assumptions are given in Electronic Supplementary Information (ESI). Consequently, path length can be determined from the number of bonds in a Kuhn step ($N_{b,Kuhn}$) and the number of projected Kuhn steps to the thickness of the film (d), as illustrated in **Figure 2**:

$$N_{b,min} = N_{b,Kuhn} \frac{d}{c_2 \langle r^2 \rangle^{1/2}} \quad (2)$$

where c_2 is the average projection of the chosen mean square radii into the thickness plane. c_2 approaches 1 for ideally cross-linked networks when the cross-linker has an infinite number of functionalities, i.e. from a given cross-linker there will always be a cross-linked polymer pointing in the direction of the field. This limit is, however, valid for unfilled elastomers only since the dimensions of silica (or other) particles will be much larger than the dimensions of the polymer chains ($\langle r^2 \rangle^{1/2}$), and thus the particles will hinder certain directions for certain chains. $\langle r^2 \rangle^{1/2}$ is the average end-to-end distance (mean square radius of a statistical polymer segment, ensuring that there is full permissible rotation around the given segment) and is given by:

$$\langle r^2 \rangle^{1/2} = b N_{b,Kuhn}^\nu = b \left(\frac{M_c}{M_{Kuhn}} \right)^\nu \quad (3)$$

where $N_{b,Kuhn}$ is the number of Kuhn steps of the given polymer chain, and ν is an exponent that is determined for a self-avoiding random walk to $\nu = 0.588$.^[26] b is the length of the

Kuhn step. M_{Kuhn} is the molecular weight of one Kuhn step, and M_c is the molecular weight between crosslinking sites, i.e. the un-crosslinked polymer molecular weight.

Following the derivations given in ESI, electrical breakdown strength as function of polymer and network characteristics can be derived from Equations 1 and 2 and be written solely as a function of polymer characteristics as:

$$E_B = \frac{c_1 b^{0.191}}{c_2 l_B^{1.191}} \left(\frac{M_o}{2M_c} \right)^{0.588} \quad (4)$$

where M_o is the monomer molecular weight, M_c is the molecular weight between crosslinking sites, b is the length of the Kuhn step, namely $\sim 25\text{\AA}$ for PDMS,^[27] and l_B is the bond length of Si-O (1.63 \AA).

In other words, electrical breakdown strength is proportional to Kuhn step characteristics, with bond length playing the most significant role. The rigidity of the polymer is expressed in terms of the Kuhn step length. For PDMS-based silicone elastomers, b and l_B are constants (except for bottlebrush polymers, where b and M_o can be varied interdependently) and the variables in network design reduce to the distance between cross-links, i.e. the polymer's molecular weight. A fit of Equation 4 to data from five well-defined PDMS-based elastomers, based on a high-functionality cross-linker, is shown in **Figure 3A**. The model fits to the data excellently and certainly verifies the validity for simple network structures without any reinforcing fillers. Details on the investigated elastomers are summarized in **Table 1**.

Table 1. Details on the investigated elastomers of Figure 3.

Sample	E_{BD} (V/ μm)	Y @ 5% strain (MPa)	Tensile strength (MPa)	Strain at break (%)
DMS-V35 (HMS-301, r=1.5)	87 \pm 6	0.81	3.07	274
DMS-V41 (HMS-301, r=1.5)	66 \pm 4	0.53	2.82	402
Elastosil LR3043/30	126 \pm 10	2.00	14.8	582

Elastosil LR3043/30+10phr silicone oil TR 50	113±2	1.91	11.7	612
Elastosil LR3043/30+25phr silicone oil TR 50	99±3	1.82	9.91	673
Elastosil LR3043/30+30phr silicone oil TR 50	87±6	1.69	7.55	710
Elastosil LR3043/30+50phr silicone oil TR 50	81±2	1.50	5.22	586
Elastosil LR3043/30+75phr silicone oil TR 50	72±6	1.23	3.84	417
Elastosil LR3043/30+25phr chloropropyl oil LMS-152	106±5	1.80	10.8	700
Elastosil LR3043/30+50phr chloropropyl oil LMS-152	100±5	1.30	5.92	599
Elastosil LR3043/30+75phr chloropropyl oil LMS-152	79±8	1.15	4.12	473
Elastosil LR3043/30+100phr chloropropyl oil LMS-152	80±2	0.75	1.35	269
SE60	94±3	1.32	0.82	129
SE30	77±5	0.79	0.91	271
SE10	68±8	0.58	0.17	163
SE5	50±6	0.46	0.15	122
SE1	30±4	0.37	0.09	107
SE0	25±4	0.21	0.06	92
SEL	33±5	0.27	0.07	110

The developed model can likewise predict electrical breakdown strength as a function of pre-stretching from:

$$E_{B,pre} = E_B \lambda_x \lambda_y \quad (5)$$

where $\lambda_{i,i=x,y}$ is pre-stretching in the i -direction and $E_{B,pre}$ and E_B are electrical breakdown strengths in the stretched and unstretched states, respectively. Derivations are shown in ESI.

A fit of Equation 5 to data from four different types of PDMS-based elastomers, pre-stretched equibiaxially, to a different extent, is shown in **Figure 3B**. The model catches the overall

features of the data. Electrical breakdown strength derived from the model is independent of thickness, since it does not take into account imperfections in film formation. Data on electrical breakdown strengths are based on 10-14 measurements for each sample, and thus minute film defects are likely to be caught in any given determination; the model will most likely always overestimate the measurements, since it predicts intrinsic breakdown strength. In the same manner, the model can predict the electrical breakdown strength of swollen elastomers:

$$E_{B,swollen} = E_B \frac{1}{(1+\varphi)^{1/3}} \quad (6)$$

where $\varphi = V_{sol}/V$, and V_{sol} and V are the solvent and pristine elastomer volumes, respectively. A fit of Equation 6 to data from a commercially available PDMS-based elastomer ELASTOSIL[®] LR 3043/30, diluted with two types of PDMS-compatible silicone oil, is shown in **Figure 3C**. In contrast to mechanical properties, where the resulting properties depend on whether the solvent/oil was added before or after cross-linking, electrical properties are not affected by the method of introduction.^[29] This is due to physical entanglements not leading to any energy bond overlap.

With respect to a correlation between intrinsic electrical breakdown strength and the Young's modulus, the model cannot in the present state provide a universal, coherent picture of the relationship, since filler-filler and filler-polymer interactions, as well as polymer entanglements, are not accounted for in the model. Entanglements do not contribute to any changes in the derived equations, but they do contribute strongly to the Young's modulus in a non-linear way.^[30] However, for entanglement- and filler-free elastomers such as the recently developed cross-linked polymer brushes, a relationship can be derived.^[31] Equation 4 can be expressed in terms of the Young's modulus (Y) by assuming that the network deforms affinely such that $Y = 3\rho RT/M_c$ and electrical breakdown strength becomes:

$$E_B = \frac{c_1 b^{0.191}}{c_2 l_B^{1.191}} \left(\frac{M_o Y}{6\rho RT} \right)^{0.588} \sim Y^{0.558} \quad (7)$$

where ρ is the density of the elastomer, R is the gas constant and T is the absolute temperature. This relationship must be used carefully, since entanglement and filler contributions to elasticity are ignored such that Equation 7 holds only for elastomers without fillers and cross-linked from polymers with molecular weights below the molecular entanglement weight, such as for the investigated bottlebrush polymers. The model prediction of data for cross-linked bottlebrushes is shown in **Figure 3D**, from which it is obvious that the model can account for the behaviour of cross-linked bottles.

The overall conclusion is that the model indeed predicts a wide variety of silicone elastomer data and thus helps understand the effect of any change in network structure on electrical breakdown strength. Traditional silicone elastomers from linear, unimodal polymers all suffer from the conflicting role of decreasing molecular weight between cross-links, since the Young's modulus also increases with decreased molecular weight, and thus increased inherent electrical breakdown strength is achieved by sacrificing softness. Tailor-made elastomers with complex but controlled network structures, such as heterogeneous bimodal elastomers and bottlebrush polymer-based examples, have emerged recently as excellent candidates for soft actuators.^[31,32] Both types of elastomer are very soft and possess good actuation properties. Bimodal elastomers have low electrical breakdown strength compared to filled elastomers but high electrical breakdown strength compared to non-filled elastomers. Bottlebrush elastomers have overall low electrical breakdown strength. Cross-linked bottlebrush polymers remain relatively uninvestigated, due to their recent emergence, but from an electrical point of view these materials will never excel, due to their inherent dilution, which at the same time is the reason for their unmatched elastic properties. High electrical breakdown strengths, however, may not be needed for bottlebrush elastomers in order to realise their potential as dielectric elastomers. Yet, bimodal elastomers also remain relatively uninvestigated as soft elastomers and they may hold great promise if the elastomer structure is carefully designed.

Finally, it is obvious that pre-stretching is very favourable for any dielectric elastomer and may be used as a method to produce electrically stable transducers; however, it needs to be kept in mind that pre-stretching may lead to transient properties over time, if elastomers are highly filled despite being well-cross-linked.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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The low intrinsic electrical breakdown strength of soft silicone elastomers remains a challenge in the commercialisation of dielectric elastomers. A theoretical model, employed to describe electrical breakdown strength based on the molecular structure of the polymer network structure, is developed and shows excellent predictions in relation to the electrical breakdown strengths of a wide range of silicone-based dielectric elastomers.

Keywords: soft transducer, dielectric elastomer, silicone, electrical stability, Kuhn statistics

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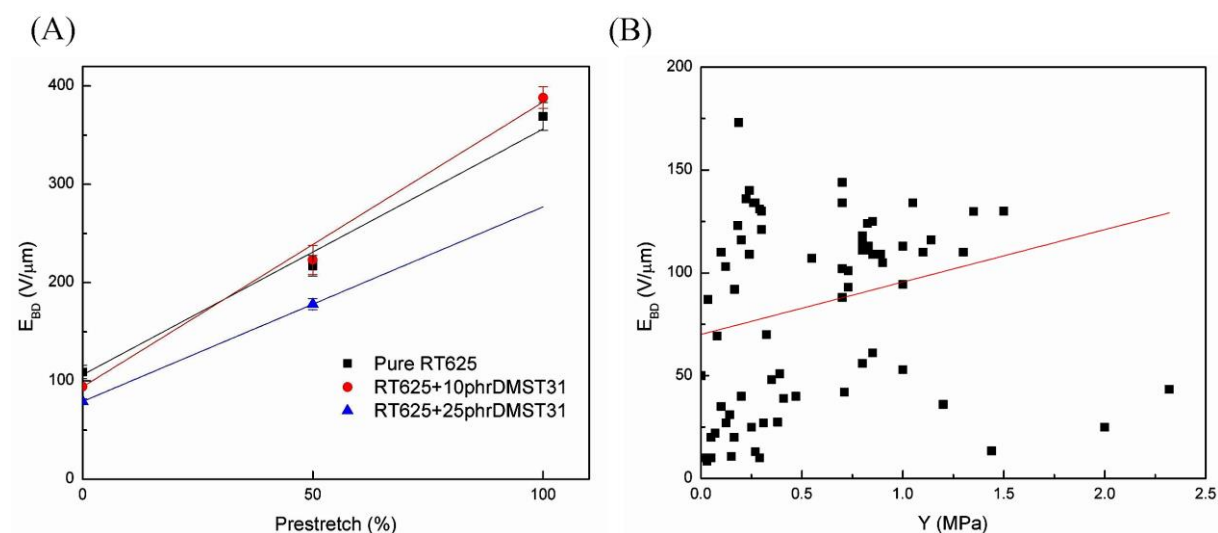


Figure 1. (A) The influence of pre-stretching of three plasticized silicone elastomers on intrinsic electrical breakdown strength. The amount of oil is stated in phr (parts per hundred rubber). The plasticizer is a silicone oil (DMST31) compatible with the Elastosil RT625 (Wacker Chemie). The addition of silicone oil to silicone elastomers in ratios of 10-30 phr is a common method for diluting and thereby softening the elastomer. (B) Electrical breakdown strength as a function of the Young's modulus of 50 different silicone elastomer formulations with no pre-stretch. The red line results from a least-square fit to the scattered data.^[21]

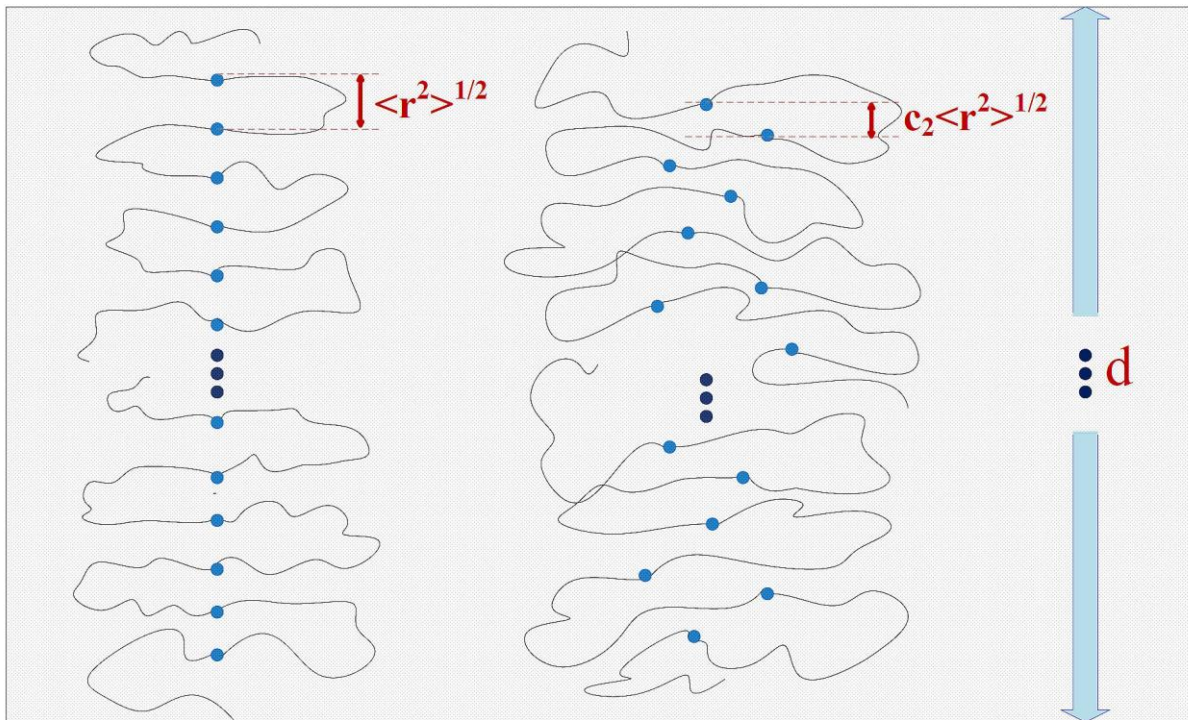


Figure 2. Illustration of the shortest path along the Si-O backbone through the highly cross-linked network from a high-functionality cross-linker. The left part illustrates the fully aligned (parallel to the thickness direction, $c_2 = 1$) situation and the right illustrates the more realistic projection of the radius in the field direction. The remaining polymer chains in the network have been omitted for clarity.

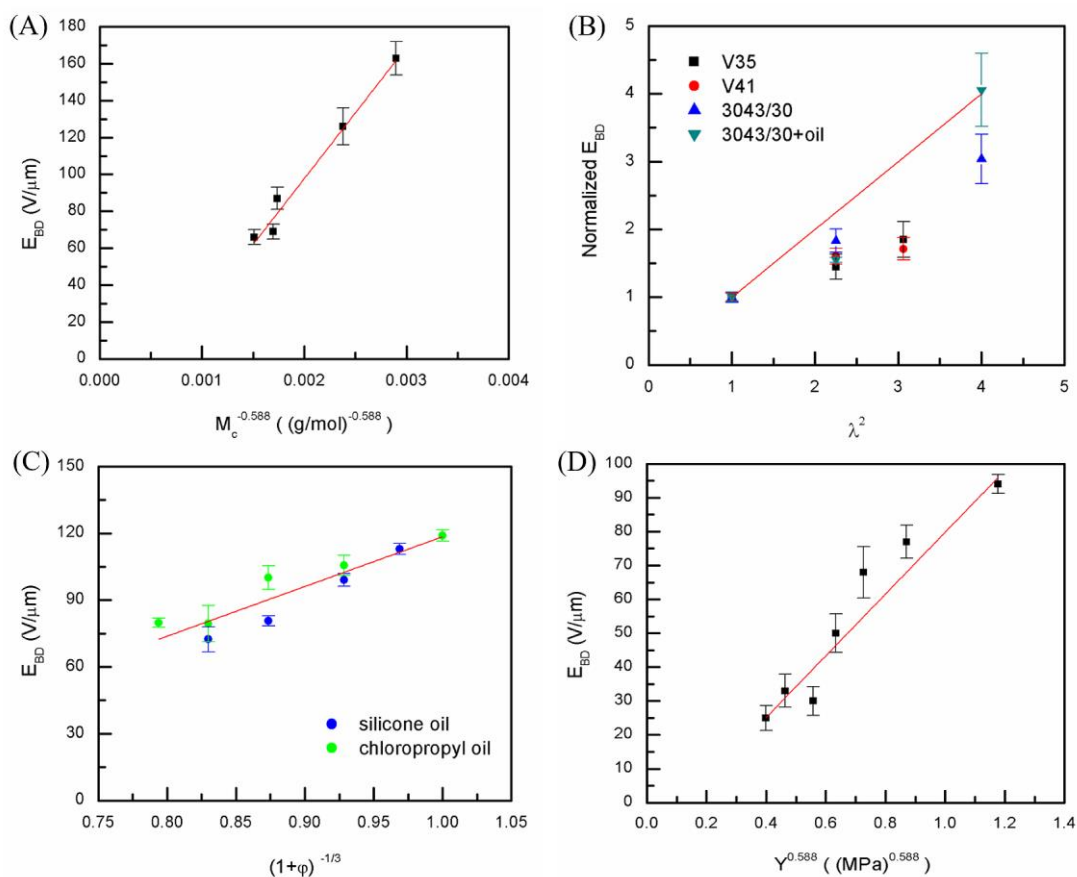
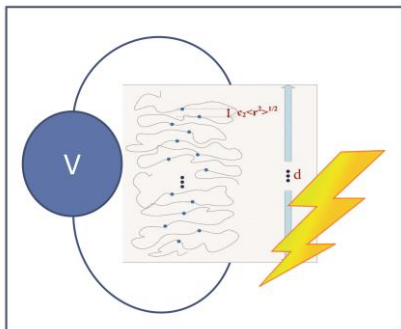


Figure 3. Electrical breakdown strengths of various types of silicone-based elastomers, with and without silica fillers, compared to model predictions. (A) Cross-linked polymer molecular weight (M_c) dependency. The elastomers are synthesized from different molecular weight telechelic vinyl-functional PDMS with constant stoichiometry and silica concentration. The higher the molecular weight of the PDMS, the lower crosslinking degree. (B) The effect of equibiaxial pre-stretching. Electrical breakdown strengths have been normalised with the value at no pre-stretching for easier comparison. The elastomers are four different types of elastomers. V35 and V41 are synthesized elastomers (with different crosslinking degree), whereas 3043/30 is a commercial LR rubber from Wacker Chemie that has been diluted with 25 phr silicone oil (for more details on samples, see ESI). (C) The effect of adding two types of compatible oils, namely the commercial silicone oil utilized before as well as a synthesized chloropropyl functional silicone oil.^[28] (D) The relationship between electrical breakdown strength and the Young's modulus for filler-free bottlebrush polymer-based networks with

varying degree of crosslinking density and thereby varying Young's modulus (the higher crosslinking density, the higher Y).



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Table 1. Details on the investigated elastomers of Figure 3.

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