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Achieving High Performance in Silicone Rubber Dielectric Elastomers via Synergistic Layer System

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Abstract—Polydimethylsiloxane (PDMS) elastomers are attractive for soft actuation but their intrinsically low permittivity demands high electric fields and promotes premature electrical failure. To address this limitation, this study implements a Synergistic Layer System (SLS) in which a hard filler (TiO₂) and soft fillers (high-permittivity silicone oil (HPSO) and glycerol (Gly)) are co-embedded in PDMS to raise dielectric response while moderating stiffness. A commercial PDMS A/B (1:1) pre-blend was formulated as single-filler films (TiO₂, HPSO, Gly; 15 wt%) and SLS hybrids (TiO₂ + Gly and TiO₂ + HPSO; 1:1, total 15 wt%), then degassed, cast in glass Petri dishes, and oven-cured at 80 °C ($\approx$ 40–80 μ m). Mechanical properties (Young's modulus, tensile strength, elongation at break) were obtained by quasi-static uniaxial tensile testing on a universal testing machine. Breakdown strength followed IEC 60243-1/-2 using a step-up high-voltage setup with semi-spherical electrodes. Relative permittivity (ϵ_r) was measured on an impedance analyzer (20 Hz – 30 MHz) using carbon-grease circular electrodes. Relative to pure PDMS, single-filler films improved either dielectric response or compliance but introduced clear trade-offs. In contrast, the SLS hybrids delivered balanced gains- TiO₂ + Gly increased ϵ_r while tempering stiffness and TiO₂ + HPSO provided the most balanced combination of ϵ_r , modulus, and breakdown strength. These results show that co-embedding a hard filler with a soft filler in a single layer complements interfacial

polarization and plasticization, enabling higher-performance PDMS actuators without excessive stiffness.

Keywords—Polydimethylsiloxane, Dielectric elastomer actuators, Synergistic, Breakdown strength.

I. INTRODUCTION

Dielectric elastomers (DEs) are widely studied for their potential use in soft actuators, artificial muscles, and adaptive devices due to their ability to convert electrical energy into mechanical motion. Among available elastomers, silicone rubber based on polydimethylsiloxane (PDMS) is one of the most attractive materials because of its thermal stability, transparency, biocompatibility and mechanical flexibility [1-4]. The chemical structure of PDMS, built around a flexible Si–O–Si backbone with pendant methyl groups, provides a high degree of molecular mobility that allows large reversible deformations under an applied field. However, despite these advantages, PDMS suffers from one fundamental weakness: its dielectric permittivity is very low ($\approx$ 2.7) [5], which forces devices to operate at very high voltages. Such high voltages are not only inefficient but also accelerate dielectric breakdown, which reduces device reliability and limits practical applications.

To address this challenge, researchers have focused on modifying PDMS with fillers that enhance

its dielectric and mechanical performance [6-9]. Fillers can generally be divided into two groups: hard fillers such as titanium dioxide (TiO_2) and soft fillers such as glycerol and high-permittivity silicone oil (HPSO). Hard fillers increase dielectric permittivity and breakdown strength but also make the polymer matrix stiffer and less flexible [10]. Soft fillers, in contrast, improve elasticity and strain at break but often reduce dielectric stability by introducing defects or phase separation [11]. Most previous approaches examined these fillers individually, which resulted in a trade-off: hard fillers improved electrical performance but reduced mechanical compliance, while soft fillers improved elasticity but weakened electrical endurance.

In this work, we propose the Synergistic Layer System (SLS) as a design approach where different fillers are incorporated together in a single layer of PDMS. The key principle of SLS is the completing each other effect. Hard fillers of TiO_2 provide high permittivity and stronger charge-trapping ability, while soft fillers of glycerol and HPSO offset the stiffening effect by lowering modulus and maintaining flexibility. When combined in appropriate ratios, the strengths of one filler type compensate for the weaknesses of the other. This synergy allows the material to achieve a balanced performance, simultaneously improving permittivity, breakdown strength, and mechanical compliance. The SLS concept therefore represents a pathway to overcome the long-standing trade-off that has limited the performance of PDMS-based dielectric elastomers.

Having said that, the present study evaluates the SLS approach using TiO_2 , glycerol, and HPSO fillers in both single-filler and hybrid combinations. The systems are analyzed in terms of relative permittivity, Young's modulus and breakdown strength. Special attention is given to the hybrid formulations, where the synergistic role of hard and soft fillers demonstrates the completing-each-other effect most clearly. This study therefore evaluates the performance of these hybrid systems against their single-filler counterparts, with the goal of determining if such a synergistic approach can successfully balance the key properties necessary for advanced actuator applications.

II. MATERIALS AND METHOD

A. Polymer Matrix

The silicone elastomer used as the base polymer matrix was ELASTOSIL® LR 3003/60 A/B (Wacker Chemie AG). This material is a two-part system, where Part A contains polydimethylsiloxane (PDMS) and a platinum catalyst, and Part B contains PDMS with a cross-linking agent. Mixing the two parts in a 1:1 ratio initiates the curing reaction and ensures uniform crosslinking across the elastomer matrix. This grade of PDMS was selected because of its high thermal stability, transparency, and flexibility, which make it suitable for dielectric elastomer applications.

B. Fillers

To enhance dielectric performance, two categories of fillers were used: Hard filler: Titanium dioxide (TiO_2) powder with an average particle size of 250 nm (Sigma-Aldrich, Merck). This filler was chosen for its high dielectric permittivity and ability to strengthen the polymer matrix. Soft fillers: Glycerol oil ($\geq 99\%$ purity, Sigma-Aldrich) and high-permittivity silicone oil (HPSO, Gelest Inc.) were selected as polar and silicone-compatible additives, respectively. Both fillers were intended to increase permittivity and improve flexibility.

C. Processing Aids

Analytical-grade heptane ($\geq 99\%$ purity, Thermo Fisher Scientific) was used as a solvent to reduce viscosity and improve filler dispersion during mixing. Carbon grease with high electrical conductivity (MG Chemicals) was applied to sample surfaces during dielectric testing to ensure stable electrode contact. Films were cast in standard glass Petri dishes (90 mm diameter), producing uniform thicknesses of < 1.0 mm.

D. Preparation of Synergistic Layer System (SLS)

The preparation method was adapted from Skov *et al.* [12] with modifications. Equal amounts of PDMS Part A and Part B were weighed and mixed thoroughly with heptane using a magnetic stirrer at 2000 rpm for 30 minutes. The selected filler was then incorporated into the mixture and stirred for an additional 5 minutes to achieve uniform dispersion. The premix was degassed in a vacuum desiccator for 15 minutes to remove trapped air bubbles. Approximately 10 mL of the degassed mixture was poured into a Petri dish and left for 1 hour at room temperature to allow partial solvent evaporation. The films were then cured in an oven at 80°C for 30 minutes. After curing, the films were carefully peeled from the Petri dish and stored between polyethylene sheets until further testing.

The effect of filler concentration on sample integrity was carefully evaluated. Preliminary studies were done with samples prepared with filler loadings below 15% showed lower dielectric and mechanical property values, indicating that improvements were less pronounced at these concentrations. Conversely, films prepared with filler loadings above 15% lost structural integrity as they adhered strongly to the Petri dish and could not be peeled without tearing or cracking. Therefore, 15% loading was identified as the most suitable concentration to balance processability, stability, and property enhancement.

Based on this criterion, five formulations were selected for detailed investigation: T15: PDMS with 15% TiO_2 . G15: PDMS with 15% glycerol. HPSO15: PDMS with 15% high-permittivity silicone oil. T-G15: PDMS with a total of 15% filler, comprising TiO_2 and glycerol in a 1:1 mass ratio (7.5% each). T-HPSO15: PDMS with a total of 15% filler, comprising TiO_2 and HPSO in a 1:1 mass ratio (7.5% each).

E. Characterisation

All samples were characterized for both dielectric and mechanical properties.

Mechanical Characterization: Quasi-static uniaxial tensile tests were performed using a universal testing machine (UTM) to determine the Young's modulus, tensile strength at break, and elongation at break. Samples were cut into a dumbbell shape with a gauge width of 6 mm. The thickness of each sample was measured using a vernier calliper prior to testing. Tests were conducted at a constant crosshead speed of 20 mm/min, with strain applied up to 120%.

Dielectric Breakdown Strength: The dielectric breakdown strength was measured in accordance with IEC 60243-1 and IEC 60243-2 standards using an in-house-built testing apparatus. Semi-spherical electrodes with a radius of 20 mm were used. The electrode gap was set to match the sample thickness (40 – 80 μm), applying a minimal indent of less than 5% to ensure firm contact. A stepwise increasing AC voltage (50 – 100 V per step at a rate of 0.5 – 1 steps/s) was applied until failure occurred. The breakdown strength for each sample was taken as the average of ten individual measurements.

Dielectric Permittivity: The relative permittivity was characterized using a Keysight E4990A Impedance Analyzer over a frequency range of 20 Hz to 30 MHz. Prior to measurement, circular electrodes 20 mm in diameter were applied to both sides of the samples using conductive carbon grease to ensure effective electrical contact.

III. RESULTS AND DISCUSSIONS

Relative Permittivity of SLS: The relative permittivity of the SLS samples demonstrates clear distinctions between the different fillers and their combinations (Table I). Pure PDMS, with a value of approximately 2.8, reflects the inherent limitation of the silicone backbone, whose non-polar $-\text{Si}-\text{O}-\text{Si}-$ chains and methyl side groups restrict dipolar orientation [13]. This low permittivity is the principal reason PDMS requires high operating voltages in actuator devices. With the addition of 15% titanium dioxide, the T15 system achieved a notable improvement, reaching 4.17 ± 0.17 . The inclusion of the hard filler introduces pronounced interfacial polarization at the polymer–filler boundary, where differences in permittivity and conductivity promote localized charge accumulation [14]. The strong interaction of TiO_2 with the PDMS matrix, despite some degree of aggregation, provides a robust enhancement of dielectric storage capacity.

Table I. Relative permittivity of SLS samples.

Sample	Relative Permittivity (ϵ_r)
T15	4.17 ± 0.17
G15	4.87 ± 0.36
HPSO15	4.33 ± 0.24
T-G15	4.98 ± 0.38
T-HPSO15	4.21 ± 0.25

By contrast, G15 yielded the highest permittivity among the single-filler systems, at 4.87 ± 0.36 . This significant increase arises from the highly polar hydroxyl groups of glycerol, which exhibit orientational polarization under an electric field. The presence of glycerol molecules within the PDMS matrix disrupts the uniform hydrophobic environment, introducing dipolar heterogeneity that intensifies polarization processes [15]. However, the dispersion of glycerol is imperfect, as indicated by micro-voids and phase separation reported in the morphological analysis (Fig. 1). These structural inhomogeneities suggest that, although the dielectric constant rises substantially, the system may sacrifice long-term stability and electrical performance.

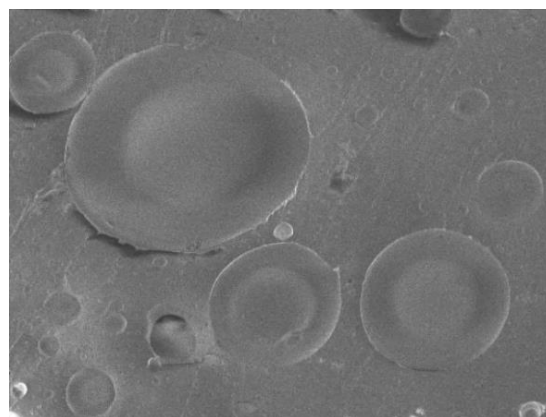


Fig. 1. Formation of glycerol voids due to dispersion of glycerol for G15 (200 μm ; mag. 200x).

The HPSO15 system displayed an intermediate performance, with a relative permittivity of 4.33 ± 0.24 . This value, though lower than G15, surpasses T15, reflecting the advantageous compatibility of HPSO with the PDMS network. The trimethylsiloxy-terminated HPSO molecules interact favorably with the PDMS backbone, embedding themselves within the polymer matrix without severe phase separation [16]. The increase is attributed to both interfacial and orientational polarization, though less pronounced than in glycerol due to weaker dipolar moments.

The most compelling outcomes were obtained from the hybrid formulations. T-G15 exhibited the highest permittivity at 4.98 ± 0.38 , surpassing both T15 and G15 individually. This synergistic behavior arises from the complementary mechanisms of the fillers: TiO_2 introduces interfacial polarization and moderates charge transport, while glycerol contributes strong dipolar alignment. Importantly, TiO_2 also acts as a dispersant, reducing the extent of glycerol clustering and enhancing the homogeneity of the dielectric network [17]. T-HPSO15, on the other hand, reached 4.21 ± 0.25 , which, although lower than the glycerol hybrid, remains higher than T15. Here, the compatibility of HPSO with PDMS promotes uniform dispersion, while TiO_2 stabilizes the dielectric response by reducing excessive chain mobility. Collectively, these results confirm that the completing each other effect is operative: rigid fillers elevate

permittivity while soft fillers ensure homogeneity and prevent excessive aggregation.

Reference [10] highlighted that rigid–flexible filler systems improve interfacial compatibility, while ref. [18] demonstrated that polar fillers enhance hard filler dispersion, producing more stable composites. The current results, in line with such reports, emphasize that hybridization is not a simple summation of effects but a synergistic mechanism that provides enhanced dielectric properties with improved quality and stability.

Young's Modulus of SLS: The mechanical performance of the Synergistic Layer System (SLS) samples is markedly shaped by the intrinsic properties of the incorporated fillers (Table II). Pure PDMS exhibited a Young's modulus of 3.64 MPa, which is characteristic of a soft, elastic silicone network with relatively free chain mobility. Incorporation of 15 wt% TiO₂ significantly reinforced the elastomer, yielding T15 with a modulus of 7.30 MPa. This substantial increase can be attributed to the rigid inorganic nature of TiO₂ particles, which act as reinforcing nodes within the polymer network. By restricting molecular motion and reducing free volume, the hard filler generates a stiffer and less deformable composite [17]. While this reinforcement improves dielectric endurance, it simultaneously compromises elasticity leading to diminished compliance under large strains.

Table II. Young's modulus, tensile strength and strain at break values of tested samples in SLS.

Sample	Young's Modulus (MPa)	Tensile Strength (MPa)	Strain at Break (%)
T15	7.30	15.24	552
G15	3.12	5.66	676
HPSO15	3.31	6.14	653
T-G15	4.24	8.92	621
T-HPSO15	4.46	10.14	706

Conversely, the addition of soft fillers produced the opposite effect. The G15 sample recorded a modulus of 3.12 MPa, while HPSO15 yielded 3.31 MPa, both lower than pure PDMS. These reductions are due to the disruption of crosslink density and the introduction of free volume. Glycerol with its polar hydroxyl groups, forms transient hydrogen bonds with the silicone backbone [15], interfering with the uniform crosslinking of the network. HPSO, acting as a plasticizer, weakens intermolecular interactions, allowing chain segments to slide more easily. Both mechanisms enhance molecular mobility, thus lowering modulus and increasing flexibility. However, this gain in elasticity comes at the cost of reduced structural strength, rendering the materials less capable of withstanding stress without deformation.

The hybrid formulations again demonstrated the value of the completing-each-other effect. T-G15 reached 4.24 MPa, while T-HPSO15 recorded 4.46 MPa. These values represent a carefully balanced

middle ground between excessive stiffness and excessive softness. TiO₂ reinforcement ensured that the films retained adequate strength, while glycerol and HPSO counterbalanced the rigidity, preserving elasticity and strain capacity [11]. This synergy creates materials that are neither brittle nor overly pliable but maintain the mechanical compliance required for dielectric elastomer actuation. Importantly, the hybrids retained elongation at break values above 600%, demonstrating that the integration of hard and soft fillers does not compromise stretchability but, rather, optimizes it.

These results resonate with prior studies; ref. [19] observed that TiO₂ fillers, while enhancing stiffness, can reduce elongation due to agglomeration at high loadings. Reference [20] reported that glycerol lowers modulus by disrupting crosslinking, while ref. [21] confirmed the plasticizing role of silicone oil. In light of these findings, the present study demonstrates that hybrid systems integrate these divergent mechanisms into a coherent balance, delivering mechanical properties that are both robust and adaptable. This reinforces the argument that SLS formulations are superior to single-filler systems for applications requiring large, reversible deformations under high electric fields.

Breakdown Strength of SLS: Breakdown strength is an essential measure of dielectric elastomer reliability, as it reflects the ability of the material to resist catastrophic electrical failure. Pure PDMS, with a breakdown strength of 89 V/μm, provides a baseline performance dictated by its uniform and relatively defect-free network. Upon loading with 15 wt% TiO₂, T15 exhibited a remarkable increase to 149 V/μm (Table III). This pronounced improvement is due to the reinforcing effect of rigid hard filler particles, which create localized barriers to charge transport. The high dielectric constant of TiO₂ promotes charge trapping at the polymer–filler interface, delaying avalanche conduction and stabilizing the dielectric response. The dense microstructure formed in T15 provides a compact and robust barrier against premature breakdown, albeit at the expense of flexibility.

Table III. Breakdown values of tested samples in SLS.

Sample	Breakdown Strength (V/μm)
T15	149
G15	67
HPSO15	69
T-G15	98
T-HPSO15	119

The behavior of soft-filler systems was less favorable. G15 displayed a reduced breakdown strength of 67 V/μm, while HPSO15 measured 69 V/μm. In these cases, the addition of polar or plasticizing species introduced structural weaknesses. Glycerol's hydrophilic domains were poorly compatible with the hydrophobic PDMS network,

producing micro-voids and weak interfacial regions that acted as preferential breakdown sites [15]. Similarly, HPSO reduced modulus and introduced localized regions of lower density, increasing susceptibility to field concentration and electrical failure [16]. These results highlight the challenge of using soft fillers alone: although they improve elasticity and permittivity, they undermine electrical endurance.

The hybrid systems once again outperformed the single-filler counterparts in achieving a superior balance. T-G15 registered a breakdown strength of 98 V/ μm , higher than G15 but lower than T15, confirming that the introduction of TiO_2 mitigates the structural weaknesses induced by glycerol. More notably, T-HPSO15 achieved 119 V/ μm , approaching the values of rigid TiO_2 composites while maintaining far greater mechanical compliance. This clearly illustrates the completing each other effect: TiO_2 stabilizes charge transport and resists breakdown, while HPSO preserves chain mobility and elasticity. The resulting films combine dielectric resilience with flexibility, qualities rarely found in single-filler systems.

Reference [21] emphasized that hard fillers enhance breakdown resistance by creating dense, charge-trapping networks. Polar additives increase microcracks and reduce electrical endurance. References [6, 7] highlighted the importance of hybrid strategies in balancing mechanical and electrical stability. The current findings corroborate these reports, demonstrating that T-HPSO15, in particular, exemplifies the hybrid design principle, producing a film that is both durable and compliant — essential traits for high-performance dielectric actuators.

To further contextualize the outcomes, the present results can be compared with established findings in the literature. Earlier studies have shown that hard fillers such as TiO_2 improve dielectric permittivity but significantly increase stiffness, while soft fillers such as glycerol or silicone oils enhance elasticity but reduce dielectric endurance [10]. More recent work on hybrid filler systems has demonstrated that combining hard and soft phases can provide a more balanced performance [6]. In this context, the T-HPSO15 formulation in the current study stands out, as it achieved an elevated permittivity (4.21 ± 0.25), improved breakdown strength (119 V/ μm), and a moderate Young's modulus (4.46 MPa), outperforming the single-filler systems. These findings confirm that the synergistic layer system approach delivers competitive and well-balanced dielectric and mechanical properties, consistent with the state-of-the-art in PDMS-based dielectric elastomer research.

IV. CONCLUSION

This study systematically evaluated the dielectric and mechanical performance of Synergistic Layer System (SLS) elastomers at 15 wt% filler loading for five formulations—T15, G15, HPSO15, T-G15, and T-HPSO15—against pure PDMS. Pure PDMS, while

flexible and easy to process, showed low relative permittivity and limited breakdown strength, which restrict actuator use. Single-filler systems changed these properties but introduced trade-offs: T15 achieved higher permittivity and superior breakdown strength through interfacial polarization and charge trapping at the TiO_2 -polymer interface, but the increased Young's modulus reduced compliance; G15 and HPSO15 raised permittivity and kept the modulus low, yet both reduced breakdown strength. The hybrid systems demonstrated the most advantageous balance by applying the completing each other effect between hard and soft fillers: T-G15 improved performance as TiO_2 mitigated the weaknesses of glycerol while retaining permittivity gains, and T-HPSO15 emerged as the best-performing formulation by combining elevated permittivity with a moderate modulus and improved breakdown strength. These results confirm that, by selecting appropriate hard-soft filler combinations within a single layer, SLS can overcome the limitations of single-filler formulations and deliver a more balanced profile, with T-HPSO15 providing the most suitable combination for dielectric elastomer actuator applications.

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