

Article

Design, Manufacturing and Characterization of Stretchable Silicone-Based Conductive Composites

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Abstract

Stretchable conductive composites are important for soft electronics, wearable systems, and adaptive electromechanical devices, yet the mechanisms governing strain-dependent electrical transport remain insufficiently understood, particularly in hybrid filler systems. In this work, the strain-dependent electromechanical behavior of graphite–silicone and hybrid graphite–copper–silicone composites was investigated under uniaxial tensile deformation up to 60% strain. Electrical measurements revealed distinct transport behaviors governed by filler composition and conductive network structure. Graphite-only composites containing 50 wt% and 60 wt% graphite exhibited monotonic resistance increases with increasing strain due to progressive widening of inter-particle tunneling gaps between neighboring graphite platelets. In contrast, hybrid graphite–copper composites showed monotonic resistance decreases under deformation, which is attributed to Poisson-ratio-driven transverse contraction, tunneling-gap reduction, and strain-assisted formation of Cu–Cu and Cu–graphite conductive pathways. Representative volume element (RVE)-based simulations further supported the proposed transport interpretation. From an engineering design perspective, the results show that filler composition and conductive network architecture can be used as design variables to tune strain-dependent electrical responses in stretchable conductive composites. These findings provide design guidance for developing silicone-based conductive composites with tunable electromechanical functionality for soft electronics, wearable sensors, and adaptive devices.

Keywords: stretchable conductive composites; graphite–silicone composites; graphite–copper composites; strain-dependent electrical transport; electron tunneling; contact percolation

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1. Introduction

Stretchable conductive composites have become key material platforms for soft electronics, wearable sensors, flexible energy systems, and human–machine interfaces [1–8]. These applications require materials that can undergo large mechanical deformation while maintaining stable and predictable electrical performance [8–14]. Elastomer-based composites filled with conductive particles offer a promising approach to achieving this combination of flexibility and conductivity. However, the fundamental mechanisms governing strain-dependent electrical transport in these materials remain incompletely understood, particularly in systems incorporating multiple filler types with distinct mechanical and electrical characteristics. Electrical transport in conductive elastomer composites is typically governed by a combination of percolation, inter-particle contact, and electron tunneling

through the insulating polymer matrix [15–20]. When the filler content exceeds the percolation threshold, a conductive network forms, enabling charge transport through both direct particle contacts and near-contact tunneling interactions. Under mechanical deformation, this network evolves through particle translation, rotation, separation, and contact formation or loss [21–25]. These structural changes can produce complex and nonlinear electromechanical responses, including monotonic increases, decreases, or non-monotonic variations in resistance depending on filler composition, morphology, and network stability.

Among carbon-based composites [26–31], graphite is widely used due to its high intrinsic electrical conductivity, chemical stability, and platelet morphology, which promotes efficient network formation at relatively low loadings [20,32–37]. In graphite-filled elastomers, electrical transport under tensile strain is often dominated by tunneling between adjacent platelets separated by thin polymer layers. As deformation increases inter-particle separation, resistance rises exponentially, and models such as the Simmons equation have been used to describe this behavior [38–43]. However, near the percolation threshold, the conductive network remains mechanically sensitive, and deformation significantly influences inter-particle spacing and tunneling behavior. Depending on filler loading and conductive network stability, tensile deformation may produce monotonic increases or decreases in resistance through tunneling-gap evolution and strain-activated contact formation mechanisms. The addition of metallic fillers introduces a fundamentally different transport behavior. Metallic particles, particularly those with spherical morphology and high stiffness, can form direct Ohmic contacts under deformation. As a result, tensile strain may not simply degrade conductivity through gap widening, but can instead enhance conductivity through strain-induced contact formation driven by particle rearrangement, transverse compression, and local stress concentration [44–49]. Such behavior cannot be captured by conventional tunneling-based models and requires alternative descriptions based on contact-percolation mechanisms [50–57]. In particular, a unified framework linking filler composition, mechanical deformation, and electrical transport mechanisms is still lacking.

From an engineering design perspective, stretchable conductive composites require careful selection of material constituents, filler loading, conductive network architecture, and specimen geometry to achieve a desired electrical response under mechanical deformation. In this study, graphite content, copper addition, and silicone matrix composition are treated as key design variables for controlling strain-dependent resistance behavior. The graphite-only composites are designed to evaluate tunneling-dominated resistance changes, while the graphite–copper hybrid composites are designed to examine whether the addition of a metallic filler can promote contact-assisted conductive pathways during deformation. This composition-driven design approach allows the electrical response of the composite to be tailored for different functional requirements, such as resistance-increasing, resistance-decreasing, or tunable electromechanical behavior. Therefore, the present work contributes to engineering design by connecting material formulation, manufacturing method, microstructure, and functional electrical performance in stretchable conductive composite systems. While graphite-filled elastomers and tunneling-percolation models have been studied, direct comparison of graphite-only and graphite–copper hybrid silicone composites under the same tensile deformation condition remains less developed. Thus, the novelty of this work lies in showing that filler composition can be used to design different strain–resistance responses: graphite-only composites exhibit resistance increase associated with tunneling-gap widening, while graphite–copper hybrid composites exhibit resistance decrease associated with strain-assisted formation of low-resistance conductive pathways. Therefore, this study provides a composition-dependent design approach for tuning electromechanical response in stretchable conductive composites. By combining ex-

perimental measurements with tunneling and strain-activated contact-percolation models, we establish a unified framework that explains the transition between resistance-increasing, resistance-decreasing, and non-monotonic behaviors. These results demonstrate that mechanical strain can be used as an active design parameter to tailor electrical response, providing new guidelines for the development of adaptive and multifunctional stretchable conductive materials.

2. Materials and Methods

2.1. Materials

Graphite powder was used as the primary conductive filler in this study due to its high intrinsic electrical conductivity and platelet morphology with approximate length of 40 μm and thickness of 3 μm , which facilitates conductive network formation in elastomeric matrices. A silicone (Smooth-on, 5600 Lower Macungie Road, Macungie, PA, USA) elastomer (part A and Part B) system was selected as the insulating matrix because of its high elasticity, mechanical stability under large deformation, and chemical compatibility with carbon-based fillers. In addition, copper powder (Metal powder USA, 13415 55th Ave NW, Gig Harbor, WA, USA) with an average particle size of approximately 40 μm was incorporated to introduce a highly conductive secondary filler phase. The silicone elastomer components were prepared according to the manufacturer's recommended mixing ratios (1:1) to ensure consistent crosslinking behavior across all samples.

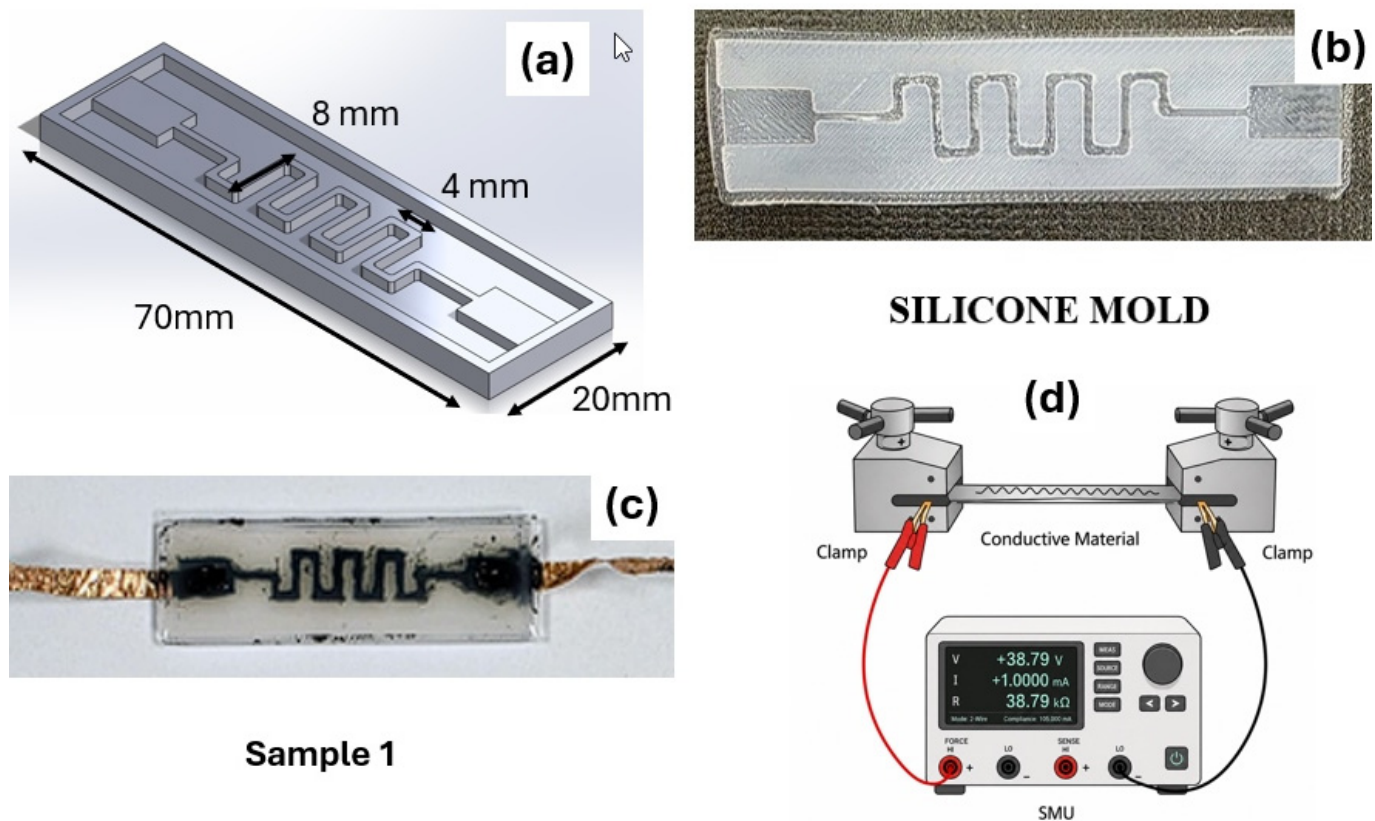
The graphite powder was selected as the primary conductive filler due to its high electrical conductivity and platelet morphology, with an approximate lateral size of 40 μm and thickness of 3 μm . Copper powder with an average particle size of approximately 40 μm was used as a secondary conductive filler to introduce metallic conductive pathways in the hybrid composites. A two-part silicone elastomer system was used as the stretchable insulating matrix. The silicone components were mixed according to the manufacturer-recommended 1:1 ratio to maintain consistent curing and mechanical stability.

2.2. Composite Fabrication

A total of seven composite formulations were prepared to systematically investigate the effect of filler loading and filler type on electrical transport phenomena. For each composition, the required amounts of graphite, copper, and silicone elastomer were weighed and mixed using mechanical stirring to ensure uniform dispersion of fillers within the polymer matrix. For composite fabrication, the required amounts of graphite, copper, and silicone elastomer were weighed according to the compositions listed in Table 1. The fillers were gradually added to the silicone mixture and mechanically stirred until a visually homogeneous conductive paste was obtained. The mixture was then degassed for 5 min to reduce trapped air before casting into the molds. The same mixing and degassing procedure was used for all formulations to maintain consistency. The paste was then cast into 3D-printed molds designed for uniaxial tensile deformation and simultaneous electrical resistance measurement. After casting, the specimens were cured for 24 h at 25 °C. No additional post-curing treatment was applied unless otherwise stated. After casting, the specimens were cured overnight at room temperature before testing. Figure 1 shows the CAD design of the mold, the fabricated silicone mold, the manufactured conductive composite specimen, and the experimental setup used for resistance measurement during tensile deformation. The compositions are shown in Table 1. Sample 1 did not show a stable measurable resistance under the selected measurement configuration because the conductive network was insufficiently formed at 40 wt% graphite without copper addition. Therefore, the resistance value is reported as non-conductive under the present measurement range. Sample 7 contains 60 wt% graphite and 40 wt% silicone type A only.

Table 1. Composition of composites and measured resistance.

Sample	Graphite (%wt)	Copper (%wt)	Silicone (A + B) (%wt)	Resistance (k Ω)
1	40	0	60	Non-conductive
2	40	10	50	2259.00
3	50	0	50	227.40
4	50	10	40	217.40
5	50	20	30	135.20
6	60	10	30	2.94
7	60	0	40 (A)	2.21


Figure 1. Setup for measuring resistance: (a) Cad design of mold, (b) fabricated silicone mold, (c) manufactured device with conductive composite, and (d) experimental setup for electrical conductivity measurement under tensile strain.

The composite mixtures were then cast into 3D-printed molds designed to produce specimens suitable for uniaxial tensile testing and electrical measurements as shown in Figure 1. After casting, the samples were cured overnight. All specimens were allowed to equilibrate at room temperature prior to characterizations (Figure 1d).

2.3. Uniaxial Loading and Electrical Resistance Measurement

Uniaxial tensile testing was performed to characterize the strain-dependent electrical response of the composites. Uniaxial tensile testing was performed under ambient laboratory conditions at a temperature of approximately 25 °C and a relative humidity of approximately 55%. Tensile loading was applied at a constant strain rate to ensure quasi-static deformation conditions and minimize rate-dependent viscoelastic effects. Each specimen was subjected to monotonic tensile deformation up to 60% engineering strain at a strain rate of 10 mm/min. Electrical resistance was measured at strain intervals of 10% using a two-probe configuration (Fluke Corporation Everett, Washington, DC, USA). Conductive contacts were securely attached to both ends of the specimen to maintain stable

electrical connection during deformation. Resistance values were recorded as a function of applied tensile strain. Multiple measurements were performed to confirm repeatability and reduce uncertainty associated with contact resistance and sample-to-sample variation. Because the two-probe method may include contact resistance, this limitation was considered during measurements. Two repeated resistance readings were taken during testing, and the measured values lay within approximately 5% of the reported mean value. However, repeated specimens for each formulation were not included in the present study. Future work can include multiple specimens for each composition to quantify sample-to-sample variability and improve reproducibility.

2.4. Analytical Modeling of Strain-Dependent Electrical Transport

2.4.1. Tunneling Model for Graphite–Silicone Composites

For the conductive composites studied here, the high-strain electrical response is interpreted in terms of electron tunneling across insulating silicone barriers separating adjacent graphite platelets. When the conductive network is stretched beyond the regime where direct particle contacts dominate, electrical transport becomes controlled by quantum-mechanical tunneling through nanometric gaps.

The resistance contribution associated with tunneling is described using the Simmons tunneling model, which expresses the tunneling current density across the insulating barrier. For applied voltages, the model leads to an exponential dependence of resistance on the barrier thickness, which can be written in simplified form as [58]:

$$R_{\text{tunnel}} = \frac{h^2 d}{A e^2 \sqrt{2m\phi}} \exp\left(\frac{4\pi d}{h} \sqrt{2m\phi}\right) \quad (1)$$

where R_{tunnel} is the tunneling resistance across the insulating barrier, h is Planck's constant, d is the thickness of the tunneling barrier, A is the effective tunneling contact area, e is the elementary electron charge, m is the electron mass, and ϕ is the barrier height or tunneling potential energy barrier. To account for mechanical deformation, the tunneling gap is assumed to evolve with strain according to a linear relationship:

$$d(\epsilon) = d_0 + k\epsilon \quad (2)$$

where d_0 is the initial effective tunneling gap at zero strain, and k is the strain-dependent gap evolution coefficient that captures the rate at which inter-particle separation increases under tensile loading.

Substituting Equation (2) into Equation (1) yields a strain-dependent resistance expression that can be fitted directly to experimental resistance–strain data in the tunneling-dominated regime. The extracted parameters d_0 and k provide physically meaningful measures of nanoscale interfacial separation and its mechanical sensitivity.

2.4.2. Strain-Activated Contact-Percolation Model for Hybrid Composites

For the hybrid graphite–copper composite (Sample 5), the experimental resistance decreases monotonically with increasing strain, indicating a transport mechanism fundamentally different from tunneling-dominated behavior. In this system, the presence of mechanically rigid, spherical copper particles enables strain-induced formation of direct metallic contacts, leading to deformation-enhanced conductivity. To capture this behavior, a strain-activated contact-percolation model is employed, in which tensile deformation progressively activates low-resistance conductive pathways through contact formation. The resistance is expressed as:

$$R(\epsilon) = R_c + (R_0 - R_c) \exp(-\beta\epsilon) \quad (3)$$

where $R(\epsilon)$ is the electrical resistance at strain ϵ , R_0 is the resistance in the undeformed state, dominated by tunneling and limited contact pathways, R_c is the asymptotic resistance corresponding to a contact-dominated conductive network at high strain, and β is a strain-activation coefficient that quantifies the rate at which metallic contacts form during deformation. At zero strain ($\epsilon = 0$), Equation (3) reduces to $R(0) = R_0$, consistent with a network in which conductive pathways are limited by particle separation and tunneling barriers. As strain increases, the exponential term decays, reflecting the progressive formation of Cu–Cu and Cu–graphite contacts driven by particle rearrangement, transverse compression, and local stress concentration. In the limit of large strain, the resistance approaches R_c , representing a mechanically stabilized, contact-dominated conduction network. This model captures the essential physics of strain-activated percolation without assuming uniform contact formation or static percolation thresholds, and it is applied over the full strain range for the hybrid composite.

3. Results and Discussion

3.1. Response of Graphite–Silicone Composites with Composition

Table 1 summarizes the effect of filler composition on the initial electrical resistance of graphite–silicone and graphite–copper–silicone composites, whereas Figure 2 graphically illustrates the influence of graphite and copper compositions on the initial electrical conductivity of the fabricated composites. Figure 2a shows the variation in electrical resistance with increasing copper concentration while the graphite composition is fixed at 50 wt%, and Figure 2b represents the effect of graphite addition in graphite–copper–silicone systems while the copper concentration is fixed at 10 wt%.

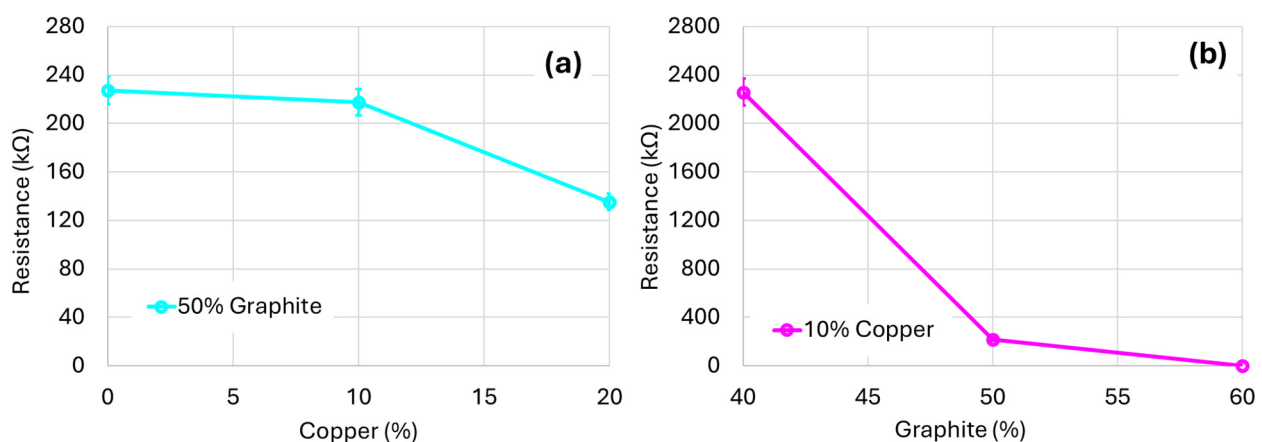


Figure 2. Effect of compositions on electrical conductivity: (a) Graphite composition variation, and (b) copper composition variation.

Figure 2 presents the measured resistance trends as a function of graphite and copper content, while Figure 3 shows the corresponding SEM microstructures of the selected composites. As shown in Figure 2a, when the graphite content is fixed at 50 wt%, increasing the copper concentration progressively decreases the electrical resistance. The graphite-only composite as shown in Figure 3b with 50 wt% graphite and 0 wt% copper (Figure 3b) shows a resistance of approximately 227.4 kΩ. With the addition of 10 wt% copper, the resistance slightly decreases to 217.4 kΩ, and a further addition of 20 wt% copper reduces the resistance to 135.2 kΩ. This trend indicates that copper particles contribute to the formation of additional conductive pathways within the graphite–silicone network. The highly conductive copper particles reduce the effective transport distance between neighboring

graphite flakes and promote Cu–graphite and Cu–Cu conductive connections, thereby improving overall electrical conductivity.

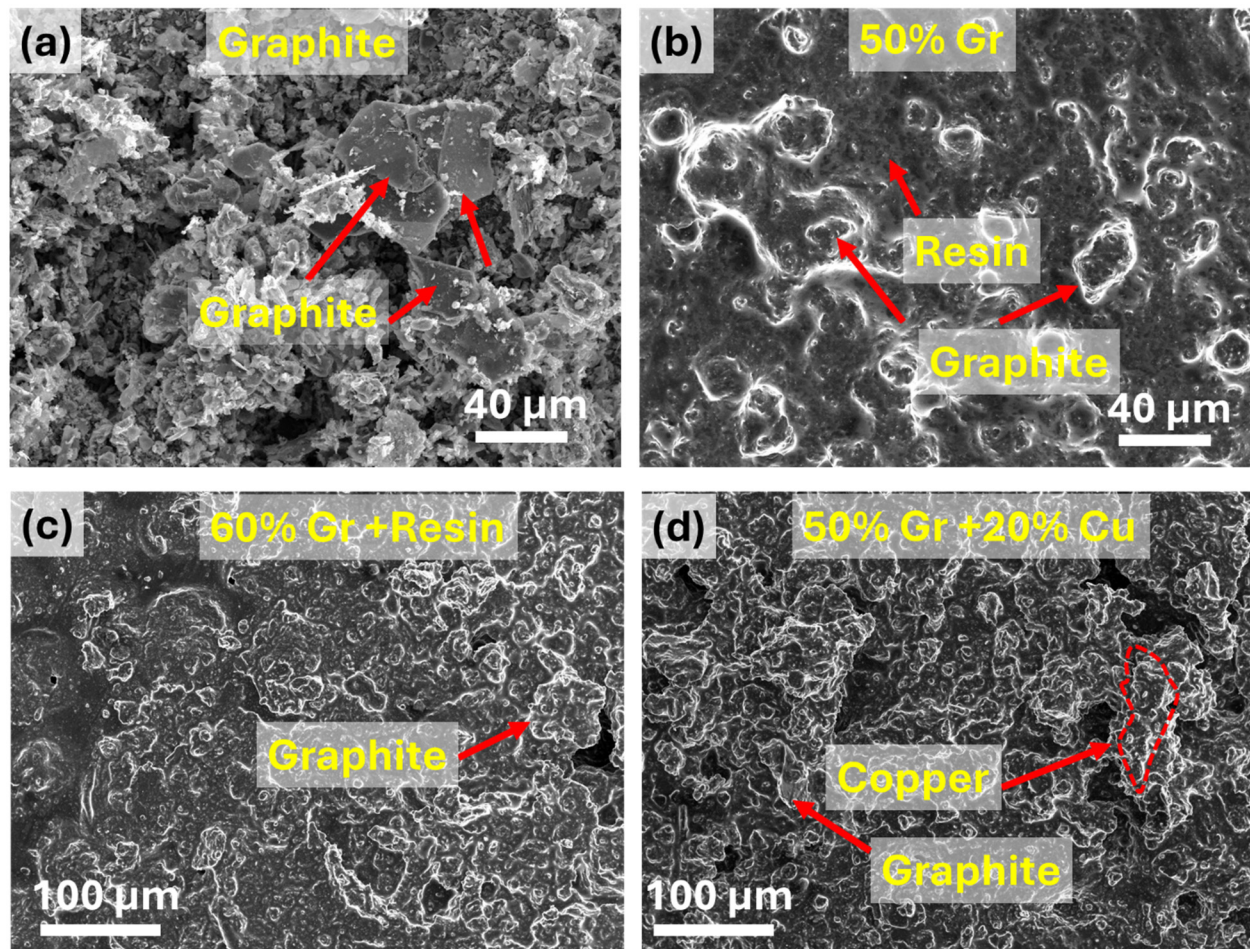


Figure 3. Scanning electron microscope (SEM): (a) Graphite; (b) silicone—50% graphite composite; (c) silicone—60% graphite composite; and (d) silicone—50% graphite + 20% copper composites.

Figure 2b shows the influence of graphite content when the copper concentration is fixed at 10 wt%. At 40 wt% graphite and 10 wt% copper, the composite exhibits a high resistance of approximately 2259 k Ω , indicating that the conductive network remains weakly connected and close to or below the effective percolation threshold. When the graphite content increases to 50 wt%, the resistance decreases significantly to 217.4 k Ω , suggesting the onset of a more continuous conductive network. A further increase in graphite content to 60 wt% leads to a dramatic reduction in resistance to approximately 2.943 k Ω (Figure 3c), confirming the formation of a dense and well-percolated conductive framework. Overall, Figure 2 demonstrates that both graphite and copper contents influence the initial electrical resistance, but their roles are different. Graphite primarily governs the formation of the percolated conductive network, while copper acts as a secondary conductive modifier that enhances electrical transport when sufficient graphite (above 50 wt%) connectivity exists. These results provide a baseline for understanding the strain-dependent electrical response discussed in the following sections.

3.2. Strain-Dependent Electrical Response of Graphite–Silicone Composites

Figure 4 represents the strain-dependent electrical resistance of the composites at applied tensile strains of 10% to 60% at a strain step of 10%. Figure 4a–c illustrate dis-

tinct transport behaviors corresponding to different filler compositions and conductive network structures.

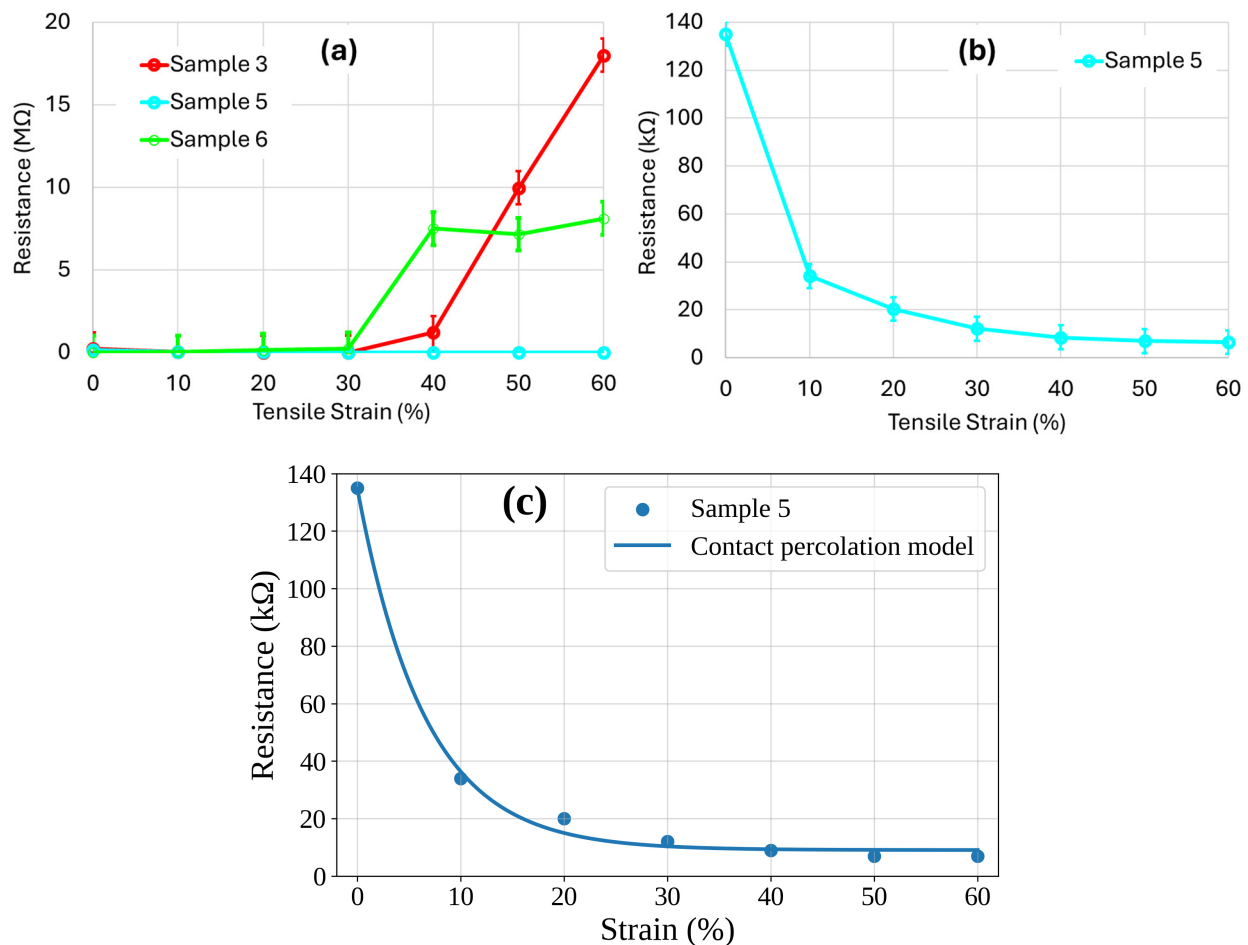


Figure 4. (a,b) Effect of tensile strain on electrical resistance, and (c) contact-percolation model data fitting.

Samples 3, 5, and 6 were selected for detailed strain-dependent analysis because they represent three distinct conductive network conditions. Sample 3 represents a graphite-only composite near the conductive transition regime, Sample 6 represents a higher graphite-loading composite with a denser conductive network, and Sample 5 represents a hybrid graphite–copper composite that exhibits a contrasting resistance-decreasing response under tensile deformation. Therefore, these samples provide representative cases for comparing tunneling-dominated and contact-assisted transport mechanisms. In Figure 4a, the strain-dependent electrical resistance responses are shown for Samples 3 and 6. Both samples exhibit a monotonic increase in electrical resistance with increasing tensile strain. At lower strain levels, the resistance increases gradually, while at higher strain levels (30–60%) the increase becomes more pronounced. This behavior is attributed to progressive widening of the inter-particle and tunneling gaps between neighboring graphite platelets during tensile deformation. As axial strain increases, the conductive pathways become increasingly separated, resulting in larger tunneling barrier thickness and consequently higher electrical resistance. The electrical transport behavior is therefore dominated by tunneling-controlled conduction throughout the entire strain range. Figure 4b shows the strain-dependent resistance behavior of Sample 5 containing 50 wt% graphite and 20 wt% copper particles. Unlike Samples 3 and 6, this sample exhibits a monotonic decrease in resistance with increasing strain. At low strain levels, electrical transport is primarily governed by tunneling between graphite particles, resulting in relatively high resistance. However, as tensile

deformation increases, the elastomer matrix undergoes axial stretching accompanied by Poisson-ratio-driven transverse contraction. This transverse contraction promotes rearrangement and local proximity enhancement of the rigid spherical copper particles and neighboring graphite platelets. As strain increases from 10% to 60%, the tunneling gap between conductive fillers decreases, significantly increasing the probability of direct Cu–Cu, Cu–graphite, and graphite–graphite contact formation. These contacts act as low-resistance conductive pathways, progressively reducing the overall resistance. At higher strain levels, contact-assisted conduction dominates over tunneling, resulting in deformation-enhanced conductivity. This behavior reflects a strain-activated contact-percolation mechanism, which fundamentally differs from the tunneling-dominated response observed in all other samples.

3.3. Quantitative Analysis Using the Simmons Tunneling Model

To quantify the strain-dependent electrical behavior, the resistance–strain responses of the composites were analyzed using models corresponding to their dominant transport mechanisms. For graphite–silicone composites, the increase in resistance with strain is governed by electron tunneling across elastomeric barriers separating adjacent graphite platelets. In the tunneling-dominated regime, resistance exhibits an exponential dependence on inter-particle separation, which is well described by the Simmons tunneling model as described by Equations (1) and (2). For fitting Equation (1), the tunneling barrier height was assumed to be $\phi = 0.5$ eV, corresponding to 8.01×10^{-20} J, based on representative values reported for carbon-filler/polymer conductive composites [59]. Using the strain-dependent tunneling gap described in Equation (2), the high-strain tunneling-dominated region was fitted by treating d_0 , k , and A as fitting parameters. The fitted values were $d_0 = 0.100$ nm, $k = 1.621$ nm per unit strain, and $A = 4.94 \times 10^{-18}$ m². The fitted model showed reasonable agreement with the experimental high-strain–resistance response, with a log-scale R^2 value of 0.908.

In Sample 5, a fundamentally different behavior is observed. The resistance decreases monotonically with increasing strain, indicating that tunneling alone cannot describe the transport mechanism. Instead, electrical conduction is governed by a combination of tunneling and strain-activated contact formation. This behavior can be represented by a parallel transport model:

$$R_{\text{total}}^{-1} = R_{\text{tunnel}}^{-1} + R_{\text{contact}}^{-1} \quad (4)$$

To capture the strain-dependent evolution of contact formation, a phenomenological contact-percolation model is employed using Equation (3):

$$R(\epsilon) = R_c + (R_0 - R_c)\exp(-\beta\epsilon) \quad (5)$$

R_0 is the initial tunneling-dominated resistance, R_c is the asymptotic resistance associated with a contact-dominated network, and β is the strain-activation coefficient. The fitted parameters indicate rapid formation of conductive pathways, confirming strong coupling between mechanical deformation and electrical transport. The calculated parameters for Sample 5 are listed in Table 2. To provide clearer physical interpretation of the fitting parameters, the key model parameters are summarized in Table 2. In the tunneling model, the parameter k represents the strain sensitivity of tunneling resistance. Physically, k describes how rapidly the effective tunneling gap increases with applied tensile strain. A higher k value indicates stronger sensitivity of electrical resistance to deformation, which is associated with faster separation of conductive graphite platelets and increased tunneling barrier thickness.

Table 2. Fitted parameters for the strain-activated contact-percolation model (Sample 5).

Parameter	Numerical Value	Physical Interpretation
R_0	$1.35 \times 10^5 \Omega$	Initial tunneling-dominated resistance
R_c	$8.9 \times 10^3 \Omega$	Asymptotic contact-dominated resistance
β	15.1	Strain-activation rate of metallic contact formation

For the hybrid graphite–copper composite, the contact-percolation parameter β describes the rate of strain-activated conductive pathway formation. In this model, β is defined as a positive coefficient in the expression $R(\epsilon) = R_c + (R_0 - R_c)\exp(-\beta\epsilon)$. The fitted value of $\beta = 15.1$ indicates rapid resistance reduction with increasing tensile strain, which is consistent with progressive formation of low-resistance Cu–Cu and Cu–graphite conductive pathways caused by transverse contraction, particle rearrangement, and reduced filler separation during tensile deformation.

3.4. Interpretation of Strain-Dependent Transport Behavior

The experimental and analytical results reveal that the electrical transport behavior of the composites is strongly dependent on filler composition and conductive network architecture. In graphite–silicone composites (Samples 3 and 6), electrical transport is primarily governed by electron tunneling between neighboring graphite platelets. Under tensile deformation, progressive widening of inter-particle spacing increases the tunneling barrier thickness, resulting in a monotonic increase in resistance with increasing strain. In contrast, the hybrid graphite–copper composite (Sample 5) exhibits a different transport response characterized by decreasing resistance under tensile loading. The addition of copper particles introduces additional conductive pathways and promotes contact formation between neighboring conductive fillers during deformation. As strain increases, transverse contraction and local particle rearrangement enhance conductive interactions, leading to progressively lower resistance and deformation-enhanced conductivity.

These observations indicate that the electromechanical response can be tailored through appropriate filler selection and conductive network design, providing a pathway for developing stretchable conductive materials with tunable electrical functionality.

4. Computational Modeling and Transport Pathways

To provide mechanistic insight into the strain-dependent electrical behavior observed experimentally, representative volume element (RVE)-based computational simulations were performed for two model systems corresponding to the tested composites: Model 1 (Sample 3: graphite–silicone composite) as shown in Figure 5a and Model 2 (Sample 5: graphite–copper–silicone composite) as shown in Figure 5b. The simulations were designed to capture the evolution of filler spacing, local deformation fields, conductive pathway interactions, and anisotropic structural changes under uniaxial tensile loading up to 60% engineering strain.

The RVE for Model 1 consists of graphite flakes with a length of 40 μm and thickness of 3 μm embedded within a silicone elastomer matrix, with an initial inter-flake spacing of approximately 75 μm . The graphite fillers were represented as platelet-like inclusions to reflect their anisotropic morphology and mechanical interaction within the silicone matrix. For Model 2, the same graphite flake configuration was retained, while a spherical copper particle with a diameter of 40 μm was introduced at the center of the RVE. This addition resulted in an initial gap of approximately 17.5 μm between the copper particle and adjacent graphite flakes, representing a hybrid conductive network combining tunneling and potential contact-assisted conductive pathways. In both models, the silicone matrix was treated as a hyperelastic continuum, while graphite and copper fillers were assumed

to behave elastically relative to the matrix, enabling realistic stress transfer and mechanical constraint effects.

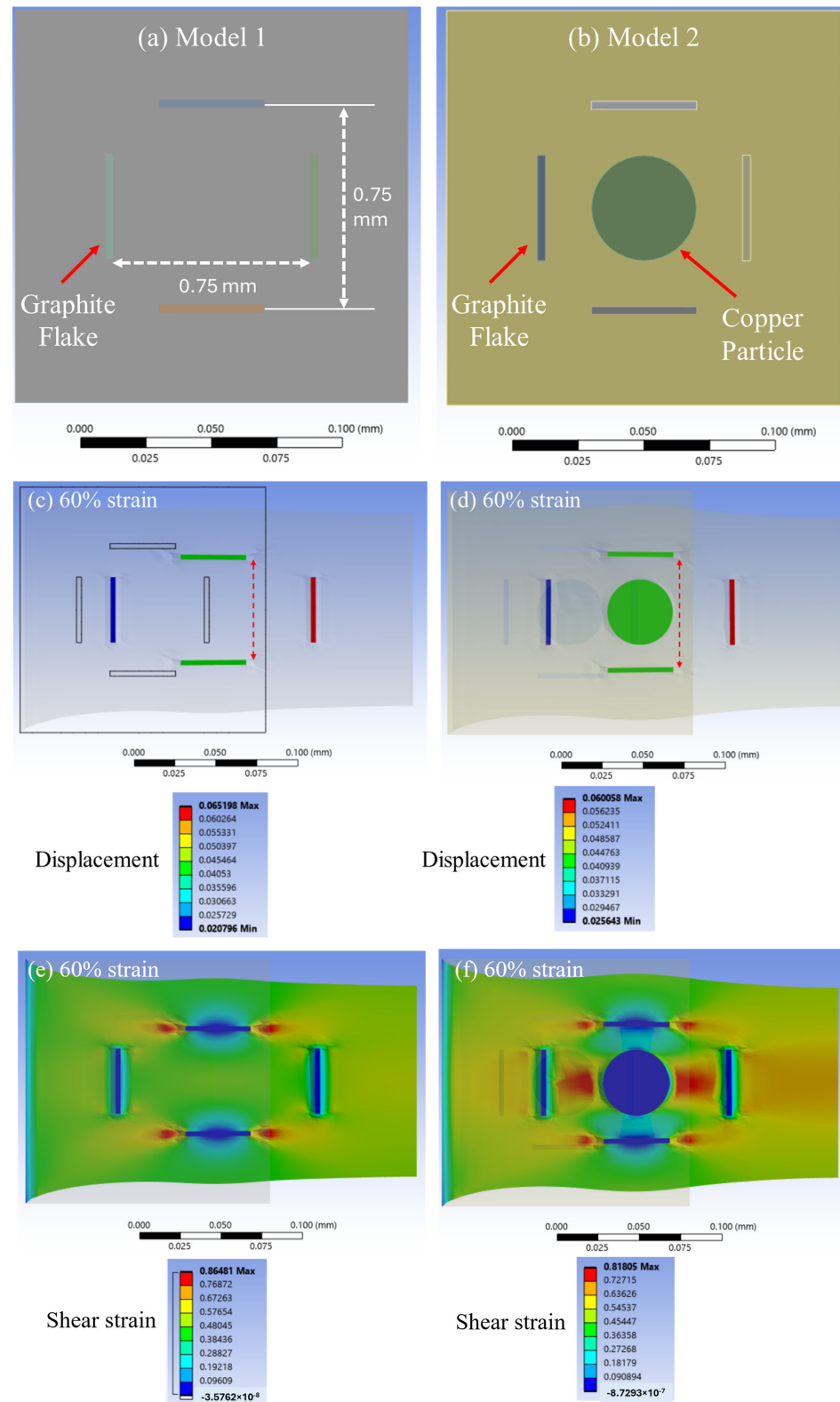


Figure 5. Computational model of composite RVE: (a) Model 1: Graphite-only; (b) Model 2: Graphite and copper; (c) tensile loading of Model 1 and Model 2; (d) displacement at 60% strain; (e,f) shear strain mapping at 60% strain.

The RVE model parameters were selected based on the experimentally used filler characteristics and the observed strain-dependent resistance behavior. The graphite fillers were modeled as platelet-like inclusions with an approximate length of 40 μm and thickness of 3 μm , consistent with the graphite powder used in the experiments. For the hybrid composite model, a copper particle with an approximate diameter of 40 μm was introduced into the RVE to represent the metallic secondary filler phase. The initial spacing between conductive fillers was selected to represent the relative filler separation expected in graphite–silicone and graphite–copper–silicone composites. Graphite was represented using platelet-like inclusions, while copper was represented as a spherical particle. The selected RVE spacing values were intended to represent local filler separation in simplified graphite-only and graphite–copper configurations. However, the RVE model does not statistically reproduce the full random filler distribution, flake orientation distribution, or copper particle dispersion observed in the real composites. Therefore, the computational results are used to support qualitative mechanistic interpretation of strain-induced gap evolution rather than to provide a fully validated quantitative prediction of the composite microstructure. It should be noted that the RVE model is a simplified mechanistic representation and does not fully reconstruct the random three-dimensional filler distribution in the actual composite. Therefore, the RVE results are used to support the interpretation of strain-induced gap widening, gap reduction, and conductive pathway formation rather than to provide a complete quantitative reconstruction of the experimental microstructure. Future work will include statistically reconstructed microstructures from higher-resolution microscopy to further improve quantitative model accuracy.

Uniaxial tensile deformation was applied along the axial (loading) direction up to 60% engineering strain. For Model 1 (Sample 3), the simulations reveal (Figure 5c,e) a pronounced anisotropic evolution of graphite flake spacing under applied strain. At 60% tensile strain, the inter-flake distance in the transverse direction decreases by approximately 16%, while the spacing in the axial loading direction increases by approximately 60%. This opposing behavior arises from the Poisson effect, where transverse contraction occurs simultaneously with axial elongation. Although radial contraction locally reduces the distance between neighboring graphite flakes, the dominant effect along the loading direction is progressive separation of conductive pathways and widening of tunneling gaps. As electrical transport primarily occurs along the tensile loading direction, this axial gap expansion progressively disrupts conductive pathways and increases tunneling barrier thickness. Consequently, the effective tunneling distance between graphite flakes increases continuously during tensile deformation, resulting in a monotonic resistance increase consistent with tunneling-dominated electrical transport behavior as observed in the experiments.

In contrast, Model 2 (Sample 5) exhibits fundamentally different microstructural evolution due to the presence of copper particles. The simulations (Figure 5d,f) show that the graphite–graphite spacing remains similar to that observed in Model 1 at 60% strain. However, the gap between the copper particle and adjacent graphite flakes decreases by approximately 25% under the same deformation condition. This reduction in separation is driven by Poisson-ratio-induced transverse contraction and localized stress concentration around the copper particle, which promotes particle rearrangement and enhanced proximity between conductive fillers. Additionally, tensile deformation facilitates alignment and interaction between copper particles and neighboring graphite platelets, enabling a progressive tunneling-gap reduction and formation of preferential conductive pathways. As strain increases, the probability of direct Cu–Cu, Cu–graphite, and localized graphite–graphite contact formation increases significantly, resulting in low-resistance conductive pathways that progressively dominate electrical transport behavior. These computational

results provide direct mechanistic support for the experimentally observed electromechanical responses. In Model 1 (Sample 3), the dominant effect of axial gap expansion increases tunneling distance and produces a monotonic resistance increase under tensile deformation. In contrast, Model 2 (Sample 5) demonstrates that the presence of copper particles introduces strain-induced transverse contraction effects, tunneling-gap reduction, and conductive pathway alignment, which collectively enhance conductive interactions and reduce effective transport distance. This behavior promotes strain-activated formation of low-resistance conductive pathways through Cu–Cu and Cu–graphite contact formation, resulting in a monotonic resistance decrease and deformation-enhanced conductivity under tensile loading. The present study focuses primarily on the strain-dependent electrical response of graphite–silicone and graphite–copper–silicone composites under monotonic tensile deformation. Full mechanical characterization, including stress–strain behavior, modulus, elongation at break, and failure analysis, was not the primary scope of this work. It is acknowledged as a limitation because filler addition can affect the stretchability and mechanical robustness of elastomeric composites. Future work will include coupled mechanical and electrical characterization to correlate modulus, failure strain, resistance evolution, and electromechanical stability. The current work is limited to monotonic tensile deformation up to 60% strain. Although the results provide useful insight into composition-dependent strain–resistance behavior, cyclic durability, resistance recovery, and hysteresis were not evaluated in this study. Future work will include cyclic loading–unloading experiments to assess repeatability, resistance recovery, hysteresis, and long-term electromechanical stability under repeated deformation.

Unified Interpretation of Transport Mechanisms

The combined experimental, analytical, and computational simulation results establish a unified framework for strain-dependent electrical transport in elastomeric conductive composites. In graphite-only systems (Samples 3 and 6), the electrical response is governed primarily by tunneling-controlled transport through mechanically constrained conductive pathways. During tensile deformation, axial stretching progressively increases the inter-particle and tunneling gaps between neighboring graphite platelets, resulting in a monotonic resistance increase throughout the applied strain range. The increase in tunneling barrier thickness suppresses electron transport efficiency and produces deformation-sensitive electrical behavior dominated by tunneling mechanisms.

In contrast, the hybrid graphite–copper composite (Sample 5) exhibits a fundamentally different transport mechanism. Under tensile loading, the elastomer matrix undergoes axial stretching accompanied by Poisson-ratio-driven transverse contraction. This transverse contraction promotes local rearrangement and proximity enhancement between rigid spherical copper particles and neighboring graphite platelets. As strain increases, tunneling gaps between conductive fillers decrease significantly, enabling strain-activated Cu–Cu, Cu–graphite, and localized graphite–graphite contact formation. These low-resistance conductive pathways progressively dominate the electrical transport behavior, resulting in monotonic resistance decrease and deformation-enhanced conductivity. While copper oxidation may be possible and could affect contact resistance between copper particles and neighboring conductive fillers, the copper powder in this study was used as received, and no separate oxidation analysis or surface reduction treatment was performed. The proposed Cu–Cu and Cu–graphite contact formation mechanism should be interpreted as a supported mechanistic explanation rather than direct experimental observation of in situ contact formation. The resistance–strain measurements, SEM-observed filler morphology, and RVE-based simulations collectively support the interpretation that tensile deformation can reduce local filler spacing in the hybrid composite and promote low-resistance conductive

pathway formation. Future work will focus on in situ microscopy and simultaneous electrical mapping during deformation to directly observe conductive pathway evolution.

Although the contact-percolation interpretation is consistent with the observed resistance decrease in the hybrid graphite–copper composite, alternative factors may also contribute to the measured response. These include changes in local filler redistribution, microstructural compaction, and local cracking. Additional four-probe measurements, in situ microscopy, and cyclic electromechanical testing can be used to separate these effects more rigorously.

The computational simulations strongly support these experimentally observed mechanisms by demonstrating axial gap widening in graphite-only systems and transverse gap reduction near copper particles in hybrid composites. These results demonstrate that mechanical strain is not merely a perturbation that degrades electrical performance, but rather an active parameter that can be used to tailor and control conductive transport pathways. By carefully designing filler composition, morphology, and conductive network architecture, it is possible to engineer composites exhibiting either resistance-increasing or resistance-decreasing behavior under mechanical deformation, enabling adaptive and tunable electromechanical functionality for next-generation soft electronics and wearable systems.

To clarify the scope of the present model, the tunneling and contact-percolation descriptions are used to interpret the strain-dependent electrical response at room temperature. Temperature-dependent resistivity measurements would provide additional insight into thermally activated transport mechanisms, including possible hopping or thermally assisted tunneling contributions. Therefore, future work will include temperature-dependent electrical characterization to further distinguish tunneling, hopping, and direct-contact transport mechanisms. Within the scope of this study, the room-temperature resistance–strain measurements, SEM observations, and RVE-based simulations provide supporting evidence for the proposed strain-dependent transport interpretation.

5. Conclusions

In conclusion, this work provides a comprehensive understanding of strain-dependent electrical transport mechanisms in graphite–silicone and hybrid graphite–copper–silicone stretchable conductive composites through combined experimental characterization, analytical modeling, and computational simulations.

1. Graphite-only composites (Samples 3 and 6) exhibited tunneling-dominated electrical transport behavior, where increasing graphite loading from 50 wt% to 60 wt% reduced the initial resistance from approximately 227.40 k Ω to 2.21 k Ω . Under tensile deformation, both systems showed a monotonic resistance increase due to progressive widening of inter-particle and tunneling gaps between neighboring graphite platelets, resulting in increased tunneling barrier thickness and disruption of conductive pathways.
2. The hybrid graphite–copper composite (Sample 5) exhibited fundamentally different electromechanical behavior, where the addition of 20 wt% copper reduced the initial resistance to approximately 135.20 k Ω and further produced a monotonic resistance decrease under tensile loading. This behavior was governed by Poisson-ratio-driven transverse contraction, tunneling-gap reduction, and strain-activated formation of Cu–Cu and Cu–graphite conductive pathways, resulting in deformation-enhanced conductivity.
3. The Simmons tunneling model captured the tunneling-dominated resistance increase observed in graphite-only composites, where tensile deformation increased the effective graphite–graphite tunneling gap and reduced conductive pathway continuity. For the hybrid graphite–copper composite, the strain-activated contact-percolation model quan-

tatively described the monotonic resistance decrease under tensile deformation. The fitted parameters, including $R_0 = 1.35 \times 10^5 \Omega$, $R_c = 8.9 \times 10^3 \Omega$, and $\beta = 15.1$, indicate rapid formation of low-resistance conductive pathways with increasing strain. These values support the interpretation that copper particles promote deformation-enhanced conductivity through Cu–Cu and Cu–graphite contact formation.

4. Representative volume element (RVE)-based computational simulations provided direct mechanistic support for the experimentally observed transport mechanisms by revealing axial tunneling-gap expansion in graphite-only systems and an approximately 25% transverse gap reduction near copper particles in hybrid composites during tensile deformation.

The key scientific contribution of this work is the demonstration that filler composition can be used as an effective design parameter to control strain-dependent electrical transport in stretchable conductive composites. Graphite-only silicone composites exhibit a tunneling-dominated resistance increase under tensile deformation due to progressive widening of graphite–graphite tunneling gaps. In contrast, hybrid graphite–copper–silicone composites exhibit resistance decrease under strain due to Poisson-ratio-driven transverse contraction, copper-assisted conductive pathway formation, and strain-activated contact percolation. These findings show that the strain–resistance response of stretchable conductive composites is not fixed, but can be intentionally designed through the selection and combination of conductive fillers. By tailoring graphite and copper contents, it is possible to engineer composites with resistance-increasing, resistance-decreasing, or tunable electromechanical behavior. This provides a useful design approach for adaptive soft electronics, wearable sensors, stretchable conductors, and multifunctional electromechanical systems.

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