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A Self-Healing Elastomer Synthesis for Flexible Wet Electrical Connectors Based on Reversible Supramolecular Interactions †

Kseniia N. Grafskaia [1,*], Vera A. Solovyeva [1] and Pranay Asthana [2]

[1] Aramco Innovations, 117105 Moscow, Russia; vera.solovyeva@aramcoinnovations.com
[2] Saudi Aramco, Dhahran 31311, Saudi Arabia; pranay.asthana@aramco.com
* Correspondence: kseniia.grafskaia@aramcoinnovations.com
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Abstract

This article describes a flexible electric connection that has a liquid-metal-infused conductive core, an insulating shell, and an outside self-healing elastomeric cover. The emphasis is on the chemistry of the elastomer, which incorporates hydrogen bonds into a silicone network to enable self-healing of the outer layer of the electric connector. This elastomer, created by standard polymerization with balanced strength and healing kinetics, recovers structural integrity in air within hours. Electrical studies show that connection remains robust even after repeated damage, emphasizing the multi-bond technique for long-lasting, stretchy electronic components.

Keywords: self-healing elastomer; downhole wet connector; flexible connector

1. Introduction

Reliable power and data transmission in fluid-filled, mechanically active, and corrosive environments, such as offshore oil and gas, marine renewable energy, and deep-sea research, are continuous technical issues [1–3]. Wet-mate connections with inflexible metallic pin-and-socket designs in oil-filled chambers are prone to corrosion, mechanical damage from vibration and misalignment, and irreversible failure upon breach, resulting in costly interventions. A trend toward active, multi-functional material systems has resulted in a promising tri-layer coaxial design with an inner conductive core, an intermediate dielectric layer, and an outer protective layer capable of self-healing (Figure 1). While the inner layers offer electrical conductivity and insulation, the outer layer serves as the first line of defense against abrasion, puncture, and chemical assault. Its intrinsic self-healing mechanism to repair small mechanical damage is crucial for increasing service life in inaccessible subsea or downhole sites where a single breach might result in catastrophic failure.

The intrinsic healing process is based on the reversibility of the embedded bonds. Mechanical damage causes small fractures in the polymer network, exposing new reactive groups. At normal temperatures, hydrogen bonds reassociate almost instantly, resulting in mechanical recovery and sealing.

Previous research has investigated conductive composites [4–6], dielectric elastomers [7,8], and self-healing polymers [9–11], but their integration into a cohesive and functionally graded link remains a key frontier. The current study uses a specifically designed silicone elastomer, with hydrogen bond connections, which can be used directly

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as the outer layer of a flexible electrical connector. The elastomer's adhesion to the intermediate dielectric layer is strong enough to avoid delamination, and its healing capacity increases the connector's operating life in harsh environments.

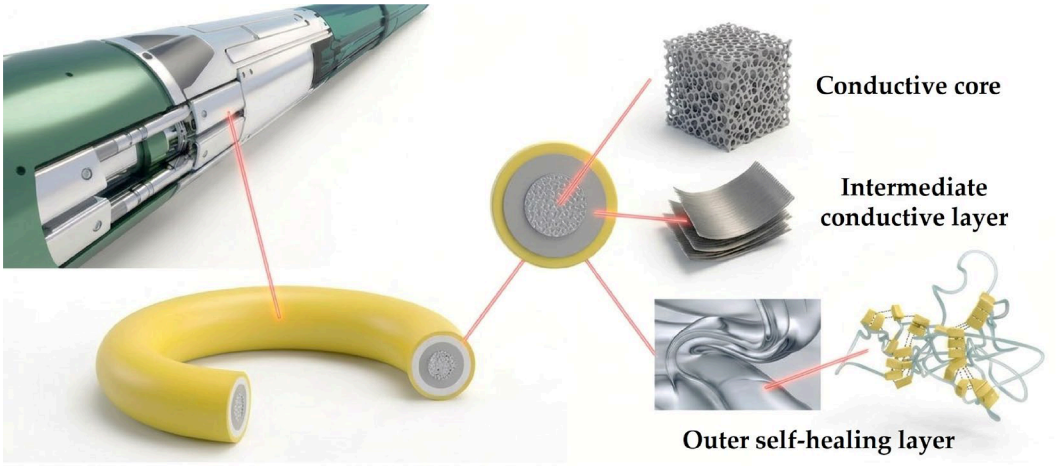


Figure 1. Construction of flexible electrical connector with self-healing outer shell.

This research examines the composition of the three-layer flexible electrical connector and the synthesis, characterization, and performance of the self-healing elastomer that forms the outer protective layer. The elastomer is a silicone-based network that incorporates hydrogen bonds to enable autonomous repair at ambient temperature without external stimulation. The elastomer is created using normal silicone polymerization, with the stoichiometry carefully controlled to balance crosslink density (for strength) and chain mobility (for healing kinetics).

2. Materials and Methods

2.1. Materials

PDMS prepolymer and curing agent (Sylgard 184) were obtained from Dow Corning (Midland, MI, USA) and used directly. Eutectic gallium-indium (eGaIn), tetrahydrofuran (THF), hydrochloric acid, aqueous potassium carbonate, diethylene-triamine (DTA) were obtained from Sigma-Aldrich (St. Louis, MO, USA) and used directly. Succinic anhydride-terminated PDMS and amine-terminated PDMS of molecular weight (0.9 kg mol^{-1}) were obtained from Gelest (Morrisville, PA, USA) and used directly.

2.2. Material Design Strategy for Flexible Wet Electrical Connector

Silicone elastomers were selected as the primary materials for fabricating the three-layer flexible connector. This choice was driven by their exceptional thermal stability, chemical resistance, and tunable mechanical properties, which collectively enable reliable performance under the high-pressure and high-temperature conditions characteristic for downhole environments. The connector architecture comprises three functional layers: a conductive core, an intermediate insulating layer, and an outer self-healing shell.

2.2.1. Conductive Core Fabrication

The liquid-metal-filled PDMS composites (PDMS-LM) were selected as the optimal core material. The conductive core is constructed from a polydimethylsiloxane (PDMS) sponge infiltrated with liquid metal, providing the necessary electrical conductivity. PDMS sponges were prepared using sugar cubes as a sacrificial template. The PDMS prepolymer and curing agent (Sylgard 184, Dow Corning) were mixed at a 10:1 weight ratio and used to impregnate the sugar templates. Degassing under vacuum for 2 h ensured complete

infiltration of the liquid PDMS into the voids of the sugar template. Alternatively, the PDMS prepolymer could be mixed with sugar grains in a 1:5 ratio to obtain a paste-like mass that could be reshaped into tubes, rings, or plates. Following curing at 65 °C for 3 h, the PDMS surface was wiped clean to expose the sugar, which was then dissolved and washed away in deionized water at 60 °C under stirring. The resulting PDMS sponges were dried at 100 °C for 2 h. This technique enables the fabrication of PDMS sponges with tunable porosity and in tubular geometries suitable for connector applications.

To render the PDMS sponge conductive, liquid metal—specifically eutectic gallium–indium (eGaIn)—was incorporated through vacuum-assisted infiltration. The PDMS sponges were placed in a bath containing the liquid metal for 5 to 60 min under vacuum, allowing the metal to penetrate into the porous network. Liquid metals naturally form a thin gallium oxide (Ga_2O_3) skin upon exposure to air, which prevents droplet coalescence and enhances adhesion to the PDMS siloxane network. The conductive properties of this core were measured and optimized to meet the required conductivity specifications.

2.2.2. Insulating Layer Configuration

An intermediate conductive layer comprising carbon tape was applied between the conductive core and the dielectric layer. This layer serves a dual purpose: it reinforces the connector structure and mitigates signal attenuation, thereby enhancing overall mechanical integrity and electrical performance.

2.2.3. Self-Healing Outer Layer

The outer shell consists of a self-healing elastomer—a modified silicone-based material that functions as an insulating dielectric layer. The self-healing mechanism is based on reversible hydrogen bonding within a hybrid crosslinked network architecture. This design addresses the fundamental trade-off between mechanical robustness and autonomous repair capability. Hydrogen bonds constitute non-covalent supramolecular interactions that provide rapid reversible crosslinking through multi-strength hydrogen-bonding motifs. These bonds reattach almost instantaneously after rupture, enabling immediate initial healing.

The self-healing elastomer was synthesized using PDMS as the elastomeric backbone, selected for its exceptionally low glass-transition temperature (−123 °C), which ensures adequate chain mobility for efficient healing, combined with high stretchability and superior thermal and chemical stability. To maximize hydrogen-bonding density, a tetra-functional amine-terminated crosslinker (TDA-PDMS) was used as cross-linker.

The self-healing outer layer was subsequently deposited atop the dielectric coating via dip-coating in the elastomer precursor formulation. This three-layer construction—comprising a conductive liquid-metal-filled PDMS sponge core, an intermediate carbon tape-reinforced insulating layer, and a self-healing silicone-based outer shell—provides the necessary combination of conductivity, insulation, mechanical reinforcement, and autonomous repair capability required for reliable downhole connector operation.

2.3. Conductive Core Characterization

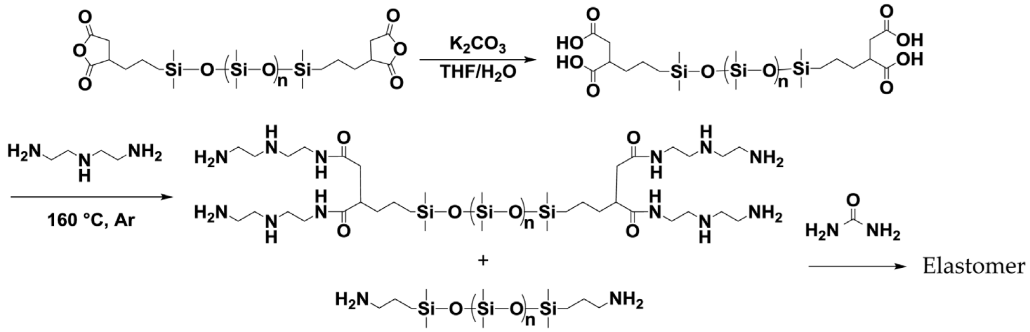
Electrical characterization of the conductive core was performed using a Rigol DS1054 oscilloscope (four-channel, 50 MHz, 1 GSa/s, RIGOL Technologies Co., Ltd., Suzhou, China), a Uni-T UTG962E function/arbitrary waveform generator (60 MHz, 200 MSa/s, Uni-Trend Technology Co., Ltd., Dongguan, Guangdong Province, China), and a Uni-T UT61E multimeter (AC/DC voltage up to 750 V, current up to 10 A, resistance up to 220 MΩ, Uni-Trend Technology Co., Ltd., Dongguan, Guangdong Province, China). Resistivity measurements were conducted at 25 °C and ambient humidity (approximately

33%) using Dupont 2.54 series pin connectors (DuPont de Nemours, Inc., Wilmington, DE, USA).

2.4. Self-Healing Elastomer Synthesis and Characterization

2.4.1. Synthesis

The synthesis of the self-healing elastomer was based on a previously described approach [12]. The tetra-functional amine-terminated crosslinker (TDA-PDMS) was first synthesized (Scheme 1). Succinic anhydride-terminated PDMS (9.0 g, 12.9 mmol) was dissolved in tetrahydrofuran and reacted with aqueous potassium carbonate for 12 h. Following pH adjustment to 7 with hydrochloric acid, the resulting tetra-acid terminated PDMS precipitate was collected, washed, and dried. This intermediate (5.0 g, 7.14 mmol) was subsequently coupled with diethylenetriamine (7.37 g, 71.4 mmol) at 120 °C for 2 h, followed by 160 °C for 24 h under an inert atmosphere, yielding the four-arm amine-terminated crosslinker after dialysis purification.



Scheme 1. Synthesis of TDA-PDMS and self-healing elastomer.

For elastomer formation, an amine-terminated PDMS with a molecular weight of 0.9 kg mol^{-1}, TDA-PDMS, and urea were combined in a fixed molar ratio of 5:1:15. The mixture was heated under argon flow at a controlled rate of 20 °C per 30 min until reaching 150 °C, and then maintained at this temperature for 48 h. The product was washed with methanol, dried under vacuum, and processed by melt pressing at 160 °C into homogeneous films.

2.4.2. Characterization

^{1}H NMR spectra were recorded on a Bruker spectrometer (400 MHz) (Bruker Corporation, Billerica, MA, USA) using DMSO-d6. IR spectra were recorded on a Nicolet iS50 FT-IR Spectrometer (Thermo Scientific, Madison, WI, USA) over a scanning range of 4000–550 cm^{-1} in Attenuated Total Reflectance (ATR) mode. The thermal properties of the products were characterized through thermogravimetric analysis on a TGA 5500 (TA Instrument, New Castle, DE, USA). The temperature was initially ramped to 100 °C and held for 10 min, followed by an increase to a target temperature of 800 °C at a heating rate of 20 °C/min. The experiments were carried out in a nitrogen atmosphere at a flow rate of 15 mL/min. Samples weighing approximately 50 ± 1 mg were placed in an open platinum pan for analysis.

3. Results and Discussion

3.1. Flexible Wet Electrical Connector Configuration

The integration of the three distinct functional layers resulted in the successful fabrication of the flexible electrical connector. The inner conductive core, consisting of a PDMS sponge template infiltrated with eGaIn liquid metal, provided a continuous and stable conductive network. The intimate contact between the liquid metal and the

PDMS scaffold—facilitated by the gallium oxide (Ga_2O_3) skin that enhances wetting and adhesion—ensured reliable electrical performance, with resistivity values confirmed to meet the required conductivity specifications during electrical characterization.

Surrounding this core, the intermediate carbon tape layer was incorporated to serve a dual critical function: it acts as a mechanical reinforcement for the connector assembly while simultaneously functioning as an interfacial buffer that effectively mitigates signal attenuation between the conductive core and the outer insulating shell. This intermediate layer thus ensures stable signal transmission even under mechanical deformation.

The outer shell, composed of the self-healing crosslinked PDMS elastomer, acts as a dielectric insulating barrier that protects the inner conductive elements from environmental degradation and electrical shorting. Notably, the reversible hydrogen-bonding network within this outer layer enables autonomous repair of surface damage: upon rupture, the multi-strength hydrogen-bonding motifs reattach almost instantaneously, allowing the elastomer to recover its structural integrity and insulating properties. This self-healing capability is particularly advantageous for extending the operational lifespan of the connector under cyclic mechanical loading, thermal cycling, and potential abrasive conditions encountered downhole.

3.2. Self-Healing Elastomer Characterization

Figure 2 presents the comparative IR spectra of anhydride-terminated PDMS, tetra-acid-terminated PDMS and tetra-diethylenetriamine-terminated PDMS. With the normalized Fourier-transform infrared (FT-IR) absorption band corresponding to the C–Si bond at 1257 cm^{-1} [13], the significantly decreased intensity of the absorption bands lying at 3511–3120 cm^{-1} (N–H stretching), 1740–1625 cm^{-1} (C–O stretching), 1550 cm^{-1} (asymmetric stretching vibration of the deprotonated carboxylate group O-C=O^-) [14] and 1561 cm^{-1} (associated N–H bending) manifests a decreased intensity of the urea groups [15].

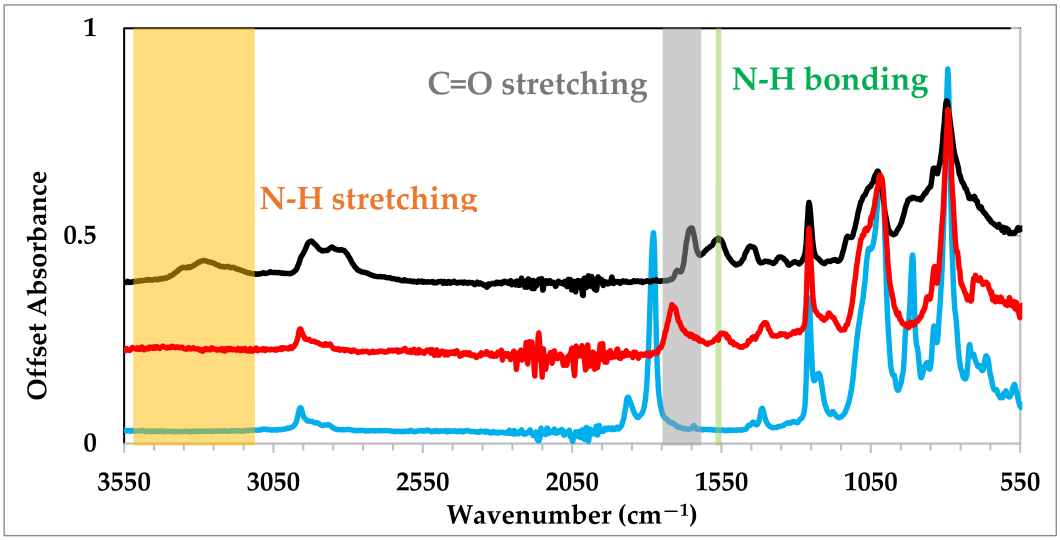


Figure 2. Comparative IR spectra of anhydride-terminated PDMS (blue curve), tetra-acid-terminated PDMS (red curve) and tetra-diethylenetriamine-terminated PDMS (black curve).

The successful synthesis of anhydride-terminated, tetra-acid-terminated, and tetra-diethylenetriamine-terminated PDMS was confirmed by ^1H NMR spectroscopy (Figure 3). The ring-opening of the succinic anhydride end-groups was evidenced by the complete disappearance of the characteristic anhydride methylene multiplets at δ 2.9–2.5 ppm (labeled e, f) and the concurrent emergence of a prominent, sharp signal at δ 2.3 ppm (labeled e + f), which is assigned to the four equivalent methylene protons of the newly formed succinic

acid moieties, accompanied by an upfield shift in the propyl linker protons to δ 1.7 (d) and 1.3 (c) ppm. Upon subsequent coupling with diethylenetriamine, complete amidation was confirmed by the disappearance of the distinct carboxylic acid peak at δ 2.3 ppm and the emergence of new, broad, and highly overlapping signals in the δ 2.5–3.5 ppm region, characteristic of the methylene protons adjacent to the amide nitrogen (δ 3.2–3.4 ppm). Crucially, the PDMS backbone signals (labeled b at ~0.5 ppm and a + a′ at ~0.0 ppm) remained consistent and unchanged across all three spectra, confirming the structural integrity of the polymer chain throughout the entire transformation process.

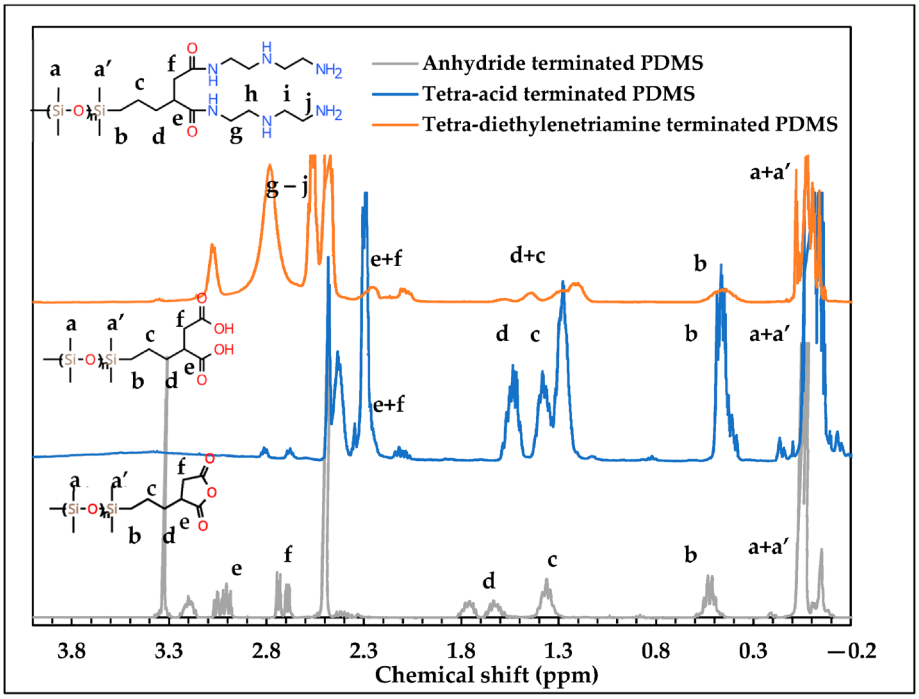


Figure 3. Comparative ^{1}H NMR spectra of anhydride-terminated PDMS, tetra acid-terminated PDMS and tetra-diethylenetriamine-terminated PDMS.

The TGA data for the synthesized tetra-diethylenetriamine (TDA) terminated PDMS, initial aminopropyl (AP) terminated PDMS and crosslinked self-healing elastomer reveal notable differences in thermal stability and decomposition behavior (see Figure 4 and Table 1) based on weight loss data at different temperature ranges. The self-healing elastomer exhibits no significant weight loss up to 450 °C (Figure 4) and higher thermal stability than the initial AP terminated PDMS and TDA terminated PDMS in temperature range between 100 °C and 400 °C, as indicated by the lower weight loss of self-healing elastomer in comparison with the AP terminated PDMS and TDA terminated PDMS. This suggests that the incorporation of the TDA terminated PDMS into the AP terminated PDMS leads to slower structure degradation. Complete decomposition of the self-healing elastomer occurs at 600 °C, which is lower than the TDA terminated PDMS, but close to the decomposition temperature of the AP terminated PDMS. Thus, it confirms that the self-healing elastomer contains far less TDA terminated PDMS than AP terminated PDMS. The different thermal behavior of the self-healing elastomer compared with that of the raw materials (TDA terminated and AP terminated PDMS) should be attributed to the effect of chemical/physical crosslinking which limits the mobility of polymer chains and the growth of a crystalline structure [13].

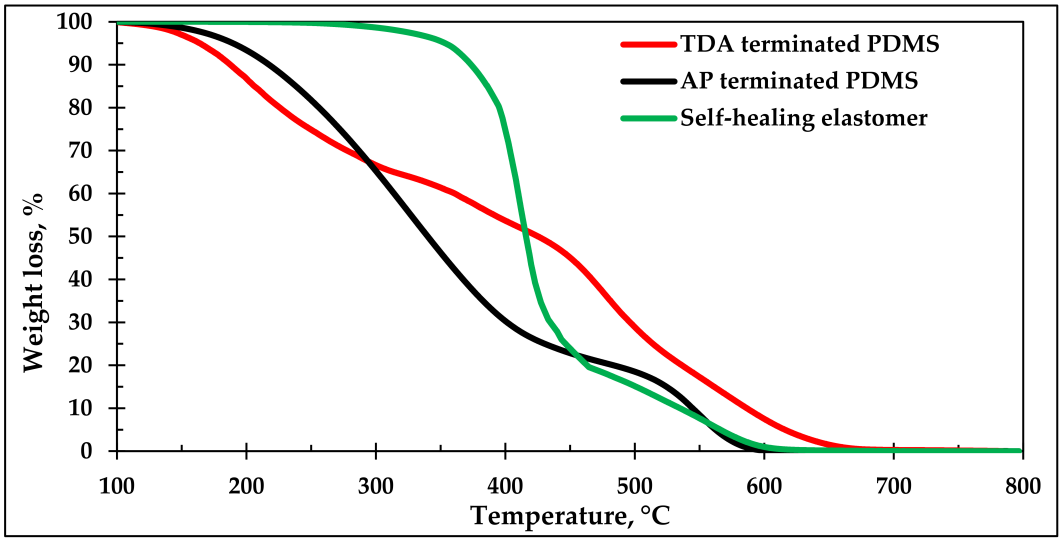


Figure 4. TGA curves of TDA terminated PDMS (red), AP terminated PDMS (black) and the self-healing elastomer after crosslinking (green).

Table 1. Weight loss of TDA terminated PDMS, AP terminated PDMS and the self-healing elastomer after crosslinking in different temperature ranges.

Sample	Weight Losses, %		
	100–250 °C	250–400 °C	400–800 °C
TDA terminated PDMS	25.13	21.42	53.45
Aminopropyl terminated PDMS	18.25	51.45	30.30
Self-healing elastomer (after crosslinking)	0.25	22.38	77.37

3.3. Flexible Electrical Connector Integration and Electrical Performance

The self-healing outer layer was successfully integrated with a PDMS-LM conductive core (a gallium–indium-tin alloy in a PDMS sponge matrix) and a dielectric silicone-based middle layer to produce a functional three-layer nonmetallic connector (Figure 1).

The resistivity of PDMS-LM tubes with diameters of 1.0, 0.8, and 0.6 cm was evaluated both through the entire sample length and at half-length positions, consistently yielding values of 0.2 Ω. Samples with a three-layer configuration were tested for resistivity across the surface and thickness, confirming continuous dielectric coverage and a conductive core resistivity of 0.2 Ω. Pierce testing was used to evaluate electrical connections through the multilayer structure without precise pin alignment. Signal integrity was tested by sending sine and step waveforms (amplitude 3.3 V, offset 1.65 V) across the whole connection assembly and comparing input and output signals to ensure minimum signal loss.

The electrical characterization indicated that the conductive core achieved contact resistance $\leq$ 0.1 Ω, meeting specified performance limits. Signal integrity evaluations revealed solid electrical connections with no signal loss across the whole multilayer structure, even after many pierce tests. The extra intermediate conductive layer effectively mitigated the detrimental effects on signal transmission that would otherwise result from the silicone-based insulation layer. The whole connector maintained stable electrical connections after multiple uses and damage events, with microscopy and mechanical tests indicating good structural healing after mechanical damage.

4. Conclusions

By strategically integrating different PDMS-based materials and their tailored chemical modifications—specifically, the liquid-metal-infused conductive sponge core (eGaIn in PDMS), the intermediate carbon tape for mechanical reinforcement and signal stabilization, and the modified crosslinked PDMS elastomer forming the self-healing dielectric outer layer—we have successfully obtained a flexible electrical connector architecture that is inherently suitable for demanding downhole applications. Electrical measurements confirm that the elastomer used as the outer sheath maintains stable insulation and does not compromise the connector's current-carrying capacity, even after repeated damage-and-heal cycles. Moreover, the self-healing outer layer effectively repairs punctures and abrasions, preventing fluid infiltration and thereby safeguarding the inner functional layers against harsh wellbore environments.

Looking forward, future research will focus on improving healing kinetics to achieve sub-hour recovery at typical subsea temperatures (4–20 °C), optimizing the elastomer's barrier properties against aggressive wellbore fluids, and scaling the synthesis for commercial manufacturing. Nevertheless, the present findings offer a robust molecular strategy for durable, flexible electronic components capable of autonomously repairing mechanical damage. This self-healing elastomer system thus represents a critical enabler for the next generation of intelligent wet-mate connections, offering enhanced reliability, reduced operational downtime, and lower lifetime costs in challenging subsea and downhole applications.

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