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# Analysis of Dynamic Covalent Crosslinks in Self-Healing Conductive Polymers for Flexible Electronics

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**Abstract**
A new class of conductive, self-healing polymers based on dynamic covalent crosslinking chemistry is presented. The material exhibits autonomous healing at room temperature, recovering up to 95% of its electrical conductivity and tensile strength after damage. Rheological and spectroscopic analyses confirm reversible bond exchange under mild conditions. Demonstrations in stretchable circuit prototypes highlight its potential in wearable electronics, soft robotics, and smart sensing devices.

***Keywords:*** Conductive polymers; Flexible electronics; Self-healing materials; Smart materials
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## 1. Introduction

Flexible and wearable electronics are the most recent developments in technology as they overcome the conventional restrictions of rigid silicon-based devices to submit innovative solutions in advanced health care systems, folded displays, and artificial skins. However, the flexibility of these devices makes them continuously subjected to mechanical stresses, scratches and microscopic fractures, which lead to failure in electronic circuits and hence shortening product lifespan. Therefore, a severe need is highlighted to develop new generation of functional materials not only flexible but have the ability for self-repairing when subjected to damage [1-3]. Researchers have developed self-healing conductive polymers those combine the unique electrical properties of organic semiconductors and mechanical properties of conventional polymers [4,5]. Such combination allows designing electronic systems with ability for self-healing similar to the biological tissues [6,7]. The largest challenge is how to achieve accurate balance between the high electrical conductivity, the mechanical stability, and self-healing ability [8]. Mostly, one of these elements is enhanced on the account of the two others, which requires a deep understanding of the molecular geometry as well as a design of polymer networks allowing the polymer chains to move and rejoin but not losing the charge transport pathways [9-11].

The dynamic covalent bonds (DCBs) represent the cornerstone in the design of self-healing smart materials and composites. They care combining the stability of the conventional covalent bonds with the reversibility of non-covalent bonds (such as hydrogen bonds) [12-14]. In flexible electronics, these bonds show the ability to break and reshape under external inducing conditions such as heat, light or mechanical pressure. This allows the broken polymer chains at the damage interface to reconstruct the crosslinking network and hence restore the structural integrity of the material [15]. Throughout the incorporation of bonds like imine, disulfide, or Diels-Alder interactions into the structures of the conductive polymers, materials can be produced with shape memory and remarkable ability to recover semi-perfect electrical conductivity after complete damage [16]. The most featured characteristic of the dynamic covalent bonds (DCBs) is their high strength when compared to other bonds. This makes the produced material tough enough to withstand the harsh operation conditions of the flexible devices while keeping sufficient dynamics to exchange bonds upon rupture. Such synergy between the structural rigidity and local fluidity gives the dynamic crosslinked polymers a big potential for practical applications such as biosensors and soft mechanical actuators [17-20].

In light of rapid advancement in fabrication techniques, this study focuses on exploring the mechanisms of reaction between dynamic crosslinked polymer with the conductive fillers or self-contained polymer chains such as PEDOT:PSS or polyaniline in order to find practical solutions for the challenges facing the flexible electronics [21-23]. The combination of electrical conductivity with the self-healing properties throughout the dynamic covalent bonds (DCBs) can stimulate new routes to develop

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sustainable and eco-friendly devices those contribute in the reduction of electronic wastes by increasing the device lifespan and decrease the need for frequent replacement due to mechanical damages [24,25]. Furthermore, this study explores the interference between crosslinking density and charge diffusion rate in addition to the optimization of the bonding interface to prevent the increment in electrical resistance at the healing point [26-28]. The final goal of the proposed methodology is represented by establishing a foundation to develop autonomous electronic systems with capability to monitor their health situation and self-repairing to support the reliability of wearable devices in critical medical applications and hence innovating new generation of flexible robotics those interact easily and efficiently with complex environments to achieve an actual leap in multi-functional smart materials technology [29-33].

## 2. Experimental Work

The experimental work of this study concentrates on the design and synthesis of a new polymer matrix based on the engineering of the dynamic covalent bonds (DCBs) in order to achieve the optimum balance between the electrical conductivity and ability of self-healing. Figure (1) shows schematically the experimental procedure considered in this work. The procedure was started by the careful selection of monomers to form the reversible crosslinking network, such as imine or boronate esters, which were incorporated to conductive polymer chains or carbon nanofillers to ensure the existence of efficient pathways for charge transport. The crosslinking density was accurately controlled in order the ultra-high levels impede the polymer chains movement that is necessary for healing process, whereas the insufficient levels would cause degradation in the mechanical properties. The produced material was subjected to advanced rheological analysis in order to study its elastic viscosity. The strain and frequency sweep tests showed a behavior similar to that of solids under normal conditions with apparent capability for chain flow when sufficiently subjected to the strain, which confirmed the dynamic bond exchange. The integrity of the produced material was not depending only on the physical contact but also on actual chemical reconstruction of the molecular network at the damage interface.

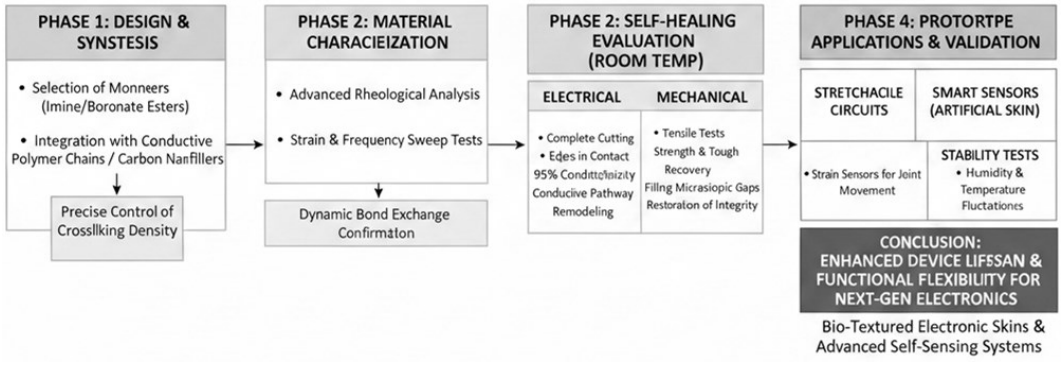


**Fig. (1) Dynamic covalent bond polymer matrix research methodology**

In the next stage, the experiments was focused on the assessment of self-healing from both mechanical and electrical viewpoints under room conditions without any external effect, which represents a qualitative leap in the sustainability of flexible electronics. The polymer samples was completely cut then the cut edges were put in direct contact for different time intervals. The results revealed excellent recovery of the electrical conductivity up to 95% within few hours. This is attributed to the reconstruction of the conductive pathways soon after the polymer networks are rejoined [34,35]. From mechanical viewpoint, the tensile tests revealed similar recovery in strength and toughness, which is ascribed to the ability of dynamic covalent bonds to fill the microscopic gaps and recover the original integrity of the material. The high capability of healing at room temperature makes this material optimum for the practical applications requiring simultaneous response for mechanical failures as this would reduce the risks of operational failure in flexible biomedical devices that are continuously subjected to bending and stretching [36].

The produced material was used to fabricate prototypes of stretchable circuits and smart sensors to confirm its feasibility in the real practical environments. The conductive polymer was used as an electrode in the strain sensors used to monitor the movement of human joints and it showed a linear and stable response after frequent cycles of cutting and healing. An artificial skin made from the

 

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produced material was tested in flexible robots. It showed an ability to keep the electrical data flow in spite of the deep scratches resulted during movement. Furthermore, the stability of the produced material was tested under fluctuated humidity and temperature to guarantee its suitability for wearable electronics. The results of the prototypes proved that integrating dynamic chemistry with conductive polymers not only supports the device lifespan but also provides the feature of functional flexibility, which is not provided by the traditional materials. This work presents a practical evidence for the feasibility of manufacturing new generation of smart electronics capable of surviving and operating in harsh environments, which prepares to develop bio-textured electronic skins and advanced self-sensing systems that would change the current understanding of the interaction between human and machine.

## 3. Results and Discussion

Figure (2a) shows the variation of electrical conductivity of the self-healing material with time after the occurrence of the damage. This result indicates the ability of the physical system to recover its electrical function gradually with high efficiency. During the first stage, the electrical conductivity is very low as the conductive pathways are interrupted due to cutting or mechanical damage, therefore, the network of electron or hole transport within the polymer matrix is disrupted. Over time, the conductivity increases gradually due to the activated self-healing mechanisms, including re-convergence of the polymer chains, diffusion of the conductive particles, and reconstruction of the internal conductive networks. It was observed that the increase in the electrical conductivity was not linear but rather abruptly varied after a specific time interval and this behavior is attributed to the percolation threshold reached by the system as the conductive pathways are connected sufficiently to allow the effective electrical transport. The comparison between the two charts shows apparent differences in the recovery rate, which indicates the effect of differences in the structures and natures of the dynamic bonds responsible to the healing. As 90% of the initial conductivity was restored, the system can be reasonably described as efficient to support the properties of these materials in flexible electronics and smart systems that require fast and reliable functional recovery [37-39].

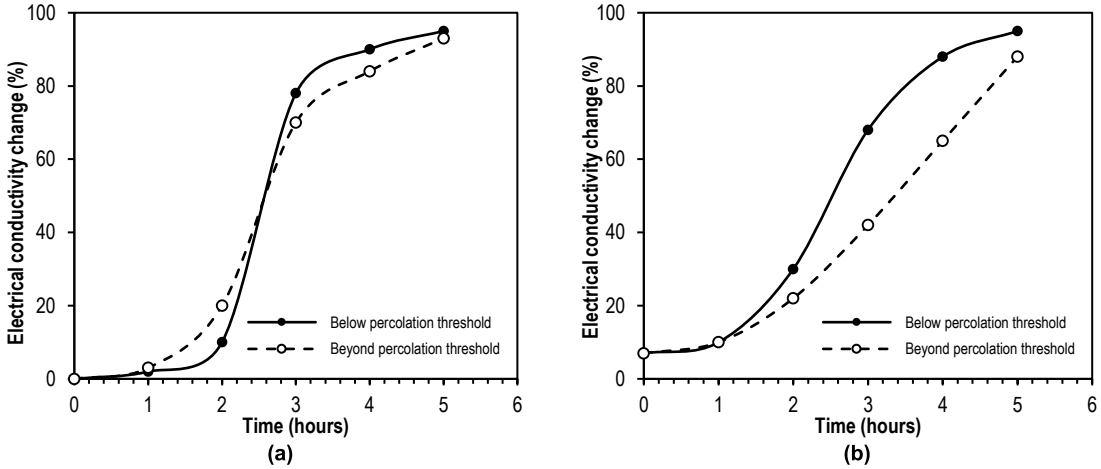


**Fig. (2) (a) Evolution of the electrical conductivity of the self-healing material as a function of time after damage occurs, and (b) Percentage of electrical conductivity recovery as a function of time**

Figure (2b) explains the percentage of electrical conductivity recovery as a function of time to introduce the comprehensive effectiveness of healing process. The recovery process includes three distinct phases: an initial phase (slow), a growth phase (accelerated), and a stability phase (saturation). In the first phase, the recovery efficiency is limited due to the slow kinetics of polymer chains or initial rearrangement of the conductive particles within the matrix. Then, in the second phase, a sharp increase in the recovery rate is observed due to the cooperative interaction between the different healing mechanisms, such as forming hydrogen or reversible ionic bonds, or physically recombining damaged regions. In the final phase, the maximum value of 95% is achieved when the system reaches a quasi-stable state as most conductive pathways are reconstructed. The main difference between the two charts is observed in the speed at which the material reaches its final state, which imposes different healing dynamics or efficient network structure. These results indicate that the electrical healing is not restricted on the surface or specific areas but rather extended into the internal structure of the material,

 

which represents a crucial factor to ensure the long-term stability of the electrical performance under frequent operation conditions [40].

Figure (3a) shows the stress-strain curves under cyclic loading to compare the mechanical response of the material in the initial cycle before damage, in the consequent cycle after the damage, and the self-healing cycle after one full hour. These curves show apparent hysteresis loops as a result of the viscoelastic nature of the material as the elastic behavior is combined with the resulted energy loss due to the internal friction and rearrangement of polymer chains. A reasonable decrease is observed in the maximum stress that the material can withstand after the cut at the same level of strain due to the disruption of the stress-carrying network. However, after healing, the material shows reasonable recovery of stress values that indicates effective reconstruction of the mechanical bonds within the system. The high convergence between the two curves refers to the recovery of the most mechanical structure despite that the response after healing does not exactly match the initial cycle. As well, the relative preservation of the hysteresis loop area after healing indicates the ability of the material to absorb energy and damping, which is a very necessary property in the applications of cyclic loading and mechanical shocks. The dotted line represents the cutting and healing moments as the mechanical recovery occurs within a relatively short time that enhances the feasibility of the material in practical applications [41-43].

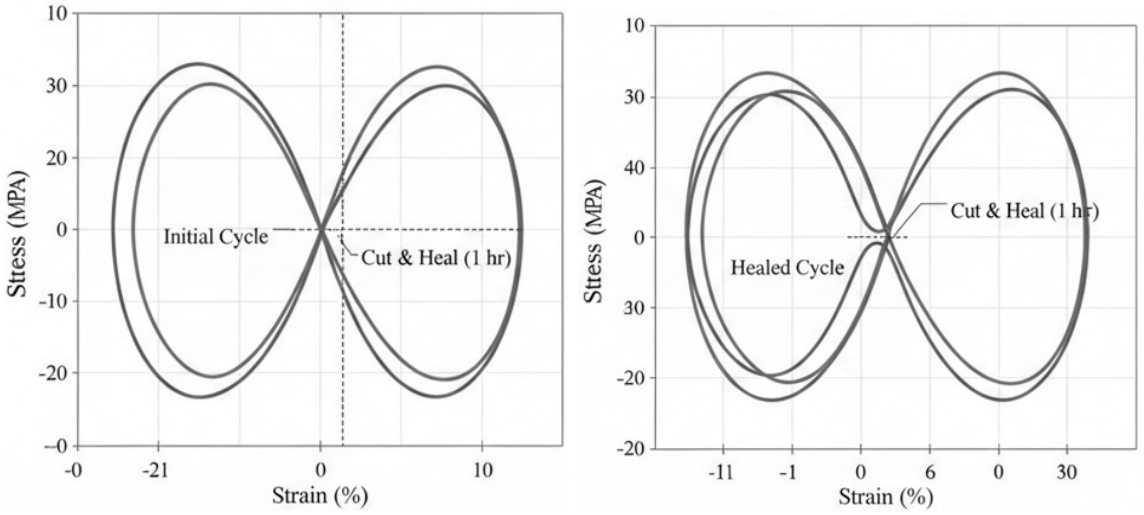


**Fig. (3) (a) The stress-strain relationship under cyclic loading, and (b) the tensile behavior of the material under cyclic loading**

Figure (3b) explains the behavior of material's tensile under cyclic loading with a comparison to the original cycle and self-healing cycle. The results show that the material recovers a large amount of ultimate tensile strength after healing as the recorded values are closer to those of the original undamaged state. Despite the small decrease in the ultimate stress or the limited variation in the shape of the curve, the overall response refers to effective self-healing mechanism to reconstruct the stress-carrying network. Furthermore, the elongation is not reasonably affected at the ultimate stress, which may be interpreted as the material's elasticity and deformability are not highly degraded. The small residual variations can be explained as microscopic defects or molecular re-entanglement in some regions, however, keeping a stable cyclic behavior and hysteresis loop shape confirms the ability of the material to withstand cyclic loading without losing the mechanical performance rapidly. These results highlight the significant potential of these materials in practical applications such as flexible electronics, soft robotics, and bioengineering, as the combination of electrical and mechanical healing properties is required to extend the service life and enhance the operational reliability [44-46].

## 4. Conclusions

In concluding remarks, the incorporation of dynamic covalent bonds into conductive polymer networks can be considered as a fundamental strategy to develop a new generation of flexible and sustained electronics. The prototypes were successfully employed in stretchable circuits, artificial skins, and biosensors as they confirm their ability to withstand harsh operation conditions in addition to the

 

reduction of electronic wastes due to enhancing the service life of the devices. Finally, this work can be a fundamental to develop self-driven electronic systems those are able to monitor their own integrity and repair their defects automatically. This widely allows the innovations in the soft robotic care as well as the advanced interaction between human and machine in order to achieve much more reliable and efficient techniques.

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