ANTAL KERPELY DOCTORAL SCHOOL OF MATERIALS SCIENCE & TECHNOLOGY

Development of Polyurethane-based Nanocomposites to be Applied as Flexible Pressure Sensors

A Ph.D. dissertation submitted to Antal Kerpely Doctoral School of Materials Science & Technology for the degree of Doctor of Philosophy in the subject of Materials Science and Technology

by

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1. Introduction

Flexible pressure sensor (FPS) are devices that are designed to measure pressure by detecting deformation flexibly [1]. Conventional pressure sensors, in contrast, are rigid sensors and are not capable to adjust to complex shapes [2]. Conventional sensors are usually heavier than FPS and might also need to be installed in a certain way. Therefore, FPS are appropriate for a wide range of applications for which conventional sensors are not [3]. In general, FPS has several benefits over conventional sensors, such as the ability to measure irregular shapes, lightweight, long lifetime, and good pressure sensitivity. On the other hand, conventional sensors may be suitable for limited applications. FPS is promising as it can be used in many advanced applications such as health monitoring [4], intelligent robots [5], and aerospace devices [6].

The sensing mechanism of FPS can be piezoresistive [7], capacitive [8] and piezoelectric [9] . The response of the sensing mechanism differs depending on the type of materials that make up the sensor and the sensor's structure. The operating principle of the piezoresistive sensor is based on the change in electrical resistance in response to the application of pressure. When an external force acts on a piezoresistive sensor and causes it to change shape, the distance between the conductive fillers decreases, resulting in a decrease in electrical resistance. On the other hand, the capacitive sensor's working principle depends on the change in capacitance when pressure is applied to it [10]. A capacitive pressure sensor consists of two parallel conducting plates separated by the dielectric material. The FPS piezoelectricity type takes advantage of this effect by measuring the change in voltage as a function of the applied pressure. Piezoelectricity means that materials generate electrical voltage when pressure is applied to them. When pressure is applied, the piezoelectric sensor deforms and produces polarization phenomena. At the same time, negative and positive charges accumulate on their opposing surfaces and convert the mechanical pressure into an electrical signal [11]. All these types of electrical signals are measurable and calculated as a function of pressure. However, the piezoresistive type is the best choice because, first, piezoresistive sensors are manufactured cost-effectively. Second, due to its high-pressure sensitivity, it is able to detect any pressure fluctuations [12]. Third, the piezoresistive sensor has high mechanical strength which makes it withstand mechanical stress without breaking [13].

2. Thesis objective

The goal is to develop a piezoresistive FPS for e-skin applications using nanocomposite technology. The piezoresistive type is the best sensing mechanism because it is low-cost, it has high-pressure sensitivity and can detect minute pressure variations. Nanocomposites have been selected for the fabrication of FPS because they are very sensitive, low cost, and very durable. Nanocomposites consist of a matrix and a nanofiller. Since hybrid nanofillers are less expensive and more pressure sensitive than pure CNT-based pressure sensors, N-BCNT and CB were selected as nanofillers. In turn, the introduction of a hybrid nanofiller improves the dispersion quality, leading to an increase in effective conduction paths. Silicone rubber was chosen as the polymer matrix due to its high flexibility and excellent chemical and electrical properties. To ensure the functionality of FPS, it is important to obtain a homogeneous dispersion of nanofillers in the polymer matrix. However, it can be difficult to disperse nanofillers uniformly in polymers with high viscosity, such as silicone rubber, which could limit the performance of the sensors. To solve this problem, we have developed a novel approach based on the dispersion of nanofillers on the skeleton of PU foam by dip coating and subsequent impregnation of the coated PU with SR. It is worth mentioning that we optimized PU foam to obtain the best pressure sensitivity by mixing commonly used isocyanate and polyol with different isocyanate indices (1.0:0.8, 1.0:1.0, 1.0:1.1) to obtain different pore volumes. The results show that the sample with a larger pore volume of PU foam leads to higher pressure sensitivity because the PU scaffold is smaller. Consequently, more nanofillers are interconnected, resulting in more conductive paths and a larger effective conductive area in the PU scaffold. The current research provides the shortest electron transport channels in silicone rubber. In addition, the use of a hybrid N-BCNT:CB nanofiller has a synergistic effect on pressure sensor performance that cannot be achieved with either material alone.

3. Preparation of N-BCNT: CB-PU/SR flexible pressure sensors for e-skin application.

Manufacturing high-performance FPS for e-skin applications requires optimum manufacturing technology. To obtain high performance of FPS, a novel technology was used by dispersion of nanofiller on PU foam by dip coating to obtain electrically conductive structure, then impregnation is silicon rubber. This technology provides a shortcut for electron transport inside silicone rubber. In addition, the best nanofiller mixing was found to be a 7:3 N-BCNT: CB mixing ratio, because the use of a hybrid nanofiller revealed improved pressure sensitivity compared to the use of N-BCNT alone. Furthermore, the selection of PU foam was according to a suitable mixing ratio of polyol: isocyanate (1:0.8), as it was found to result in the best sensing performance. The e-skin fabrication process was conducted in several steps, as outlined in Figure 1. The PU foam was cast using a topline casting machine, and the nanofiller (N-BCNT: CB) was dispersed in patosolv solvent via ultrasonic treatment for 8 minutes. The resulting nanofiller suspension was then utilized to impregnate the PU foam, which was dried at 105°C to evaporate the solvent, resulting in the formation of a stable N-BCNT: CB/PU system. The PU samples were then impregnated with SR and subjected to vacuum treatment to fill the pores of the PU foam. This resulted in the formation of the N-BCNT: CB/PU-SR pressure sensor, which was then cut and shaped for use in e-skin applications.

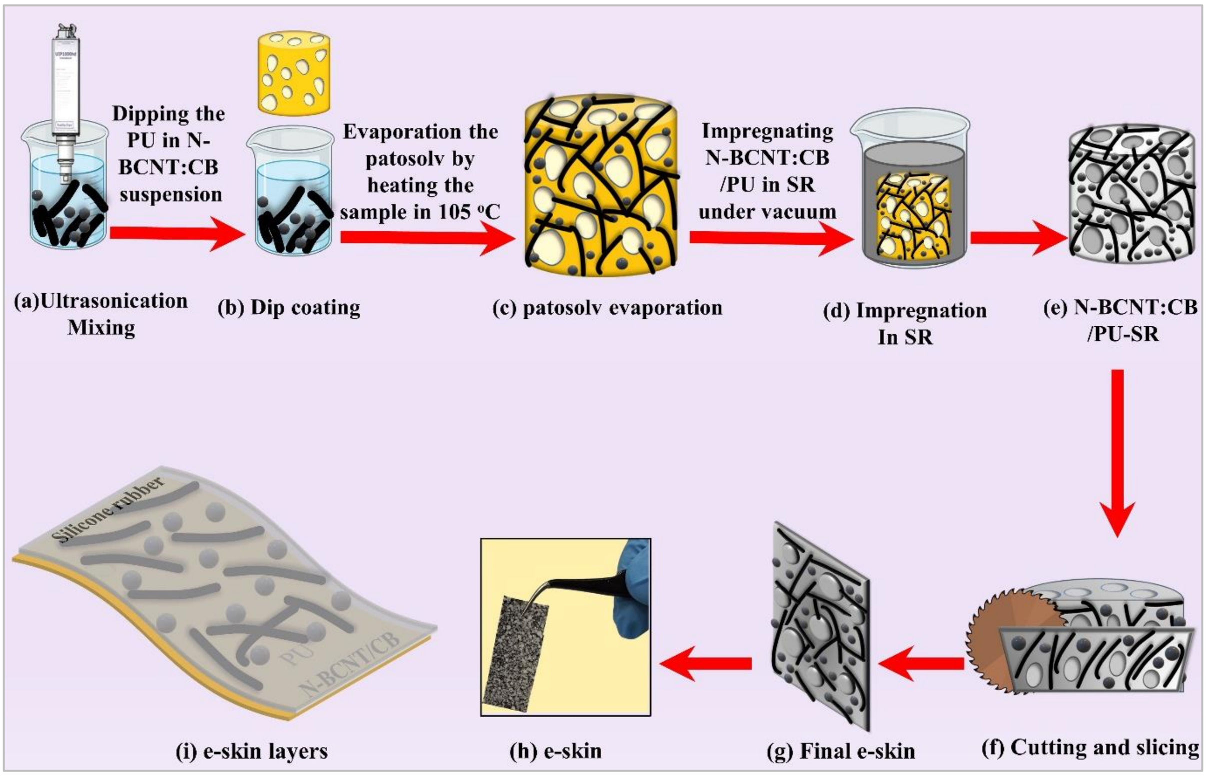

Figure 1. Preparation steps for the manufacture of e-skin. The process includes mixing a nanofiller with ultrasonication in patosolv (a), dip coating of PU in the nanofiller suspension (b), heating to evaporate the patosolv (c), impregnating the sample in SR to obtain N-BCNT: CB/PU-SR (d-e), cutting and slicing (f), resulting in the final e-skin sample (g) and the e-skin (h), e-skin layers (i).

4. Piezoresistive and electrical properties of the pressure sensor

The final N-BCNT: CB/PU-SR composite has been tested as a pressure sensor and the relationship between normalized resistance and applied pressure was determined (Figure 2A). The normalized resistance increased as the pressure increased, and vice versa. This phenomenon was attributed to the decrease in distance between nanofillers under applied pressure. Accordingly, the sensors demonstrated an increase in electrical conductivity. To measure the sensitivity of the pressure sensors the following formula is applied: $S = (\Delta R/R_0)/\Delta P$ [50]. Here, ΔR is the resistance change, R_0 represents the initial resistance, and ΔP is the difference in applied pressure. In the case if the developed system, it was revealed that within the 0-120 kPa range, the pressure sensitivity was 0.4 kPa^{-1}, while in the 180-1000 kPa range, it was 0.07 kPa^{-1}. This suggests that the sensors can effectively detect small changes in pressure across a wide range. Given this, the composite holds the potential to be applicable in various circumstances. The excellent sensitivity of the prepared N-BCNT: CB/PU-SR composite can be associated with the synergistic effect of N-BCNT and CB, which can improve the overall performance of the pressure sensor. N-BCNTs are well-known for their superior electrical conductivity[14] and good mechanical properties, making them appropriate for utilization in pressure sensors. CB is good conductive material and has a high surface area,[15][16] which could also promote the pressure sensitivity of the sensor. When N-BCNT and CB are combined as nanofillers in a sensor, they could display a synergistic effect, causing improved pressure sensitivity.[17][18] This may be due to the combined electrical and mechanical properties of the two materials that can enhance the sensitivity of the sensor to pressure changes. In addition, the high surface area of CB may further increase the sensitivity of the sensor by providing more points of contact with the polymer matrix, allowing a more sensitive response to pressure changes. Overall, the synergistic effect of N-BCNT and CB could contribute to the good pressure sensitivity of the sensor. The hysteresis loop of the sensor was tested within a pressure range of 0-1000 kPa, using normalized resistance as a system of measurement (Figure 2B). The hysteresis loop of the sensor showed a minimum difference between the increase and decrease in applied pressure, possibly owing to the viscoelastic properties of the SR polymer matrix. Using SR in conjunction with PU caused a remarkably reduced hysteresis loop, increasing the effectiveness of the pressure sensor and thus, enable accurate measurement of pressure changes within the specified range.

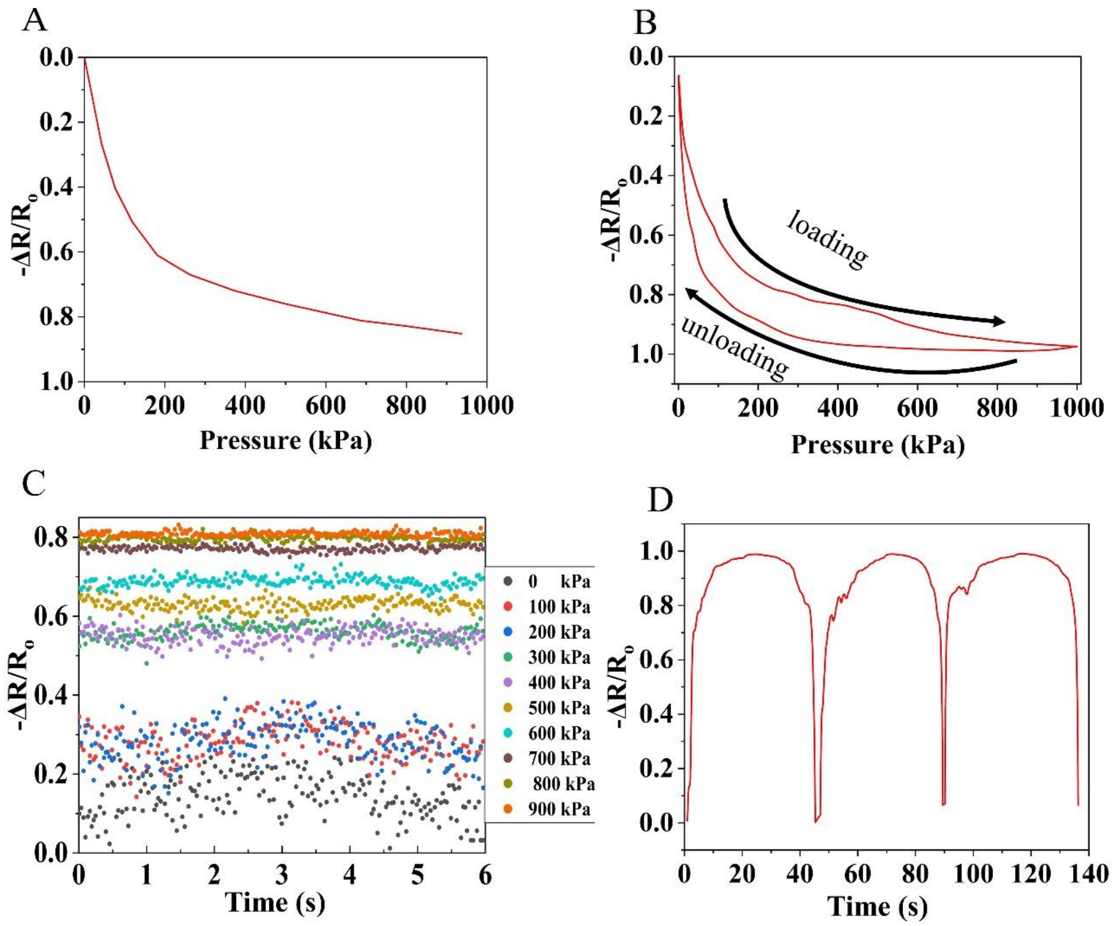

Figure 2. Normalized resistance *vs* time curve of the developed N-BCNT:CB/PU-SR composite for different pressure levels (A), loading and unloading of applying pressure *vs* normalized resistance (B), normalized resistance *vs* time (C), and (D) cyclic pressure for e-skin.

The effect of pressure on the signal stability of the developed FPS was also examined by utilizing normalized pressure as a function of time (Figure 2C). The investigation indicated an inversely proportional relationship between pressure applied and noise level. Hence, the noise level is greater at low pressures and reduced at high pressures. This behavior has two possible reasons. First, the signal noise is greatly dependent on the pressure sensitivity of the pressure sensor. At low pressure, sensitivity is high, which causes a high output signal and a high signal noise. On the other hand, at high pressure, the sensitivity is low, causing low output signal and a lowered signal noise. The second explanation for the inverse relationship between pressure and signal noise is correlated to the distances between the nanofiller in the polymer matrix and the causing electrical current flow. In contrast, at high pressure, the distances between the nanofiller in the polymer matrix are decreased, causing the formation of new conductive networks and a greater current flow. This causes lower signal noise ratio. Briefly, the inverse relationship between pressure and signal noise in nanocomposites is caused by the pressure sensitivity of the sensor and the distances between the nanofiller in the polymer matrix.

A cyclic load test was performed on the N-BCNT:CB/PU-SR composite prepared for e-skin applications, to evaluate the stability and performance of the sensor under repeated loading and unloading conditions (Figure 2D). It was shown that the normalized resistance of the sensor returned to a certain extent after the applied pressure was removed, suggesting its repeatability and stability. The combination of N-BCNT and CB as nanofillers in the sensor improved its electrical conductivity. The sensor showed good reproducibility and recoverability, possibly linked to the strong interaction at the interface between N-BCNT and CB in the pressure sensor.

The morphology of the developed N-BCNT:CB/PU-SR composite-based e-skin sensor and its contribution to the performance were evaluated by using SEM (Figure 3, A–E). The images illustrate the e-skin as comprising a PU coating with a nanofiller, which surrounds a flexible SR. The nanofiller coating on the PU provides the pressure sensor with electrical conductivity, while the SR is responsible for the durability and flexibility, enabling it to resist a variety of conditions and environments without damaging or breaking. The SEM images with different magnifications also show a uniform dispersion of the nanofiller through the PU (Figure 3, C-E). This homogenous structure improves the sensor's performance by allowing more effective electrical conductivity and enhanced data transmission. Additionally, the synergistic effect of the 7:3 N-BCNT:CB combination was also visualized (Figure 3E). The combination of CB and N-BCNT further enhances the sensor's conductivity, enabling it to correctly measure and transmit pressure data. The above-mentioned synergistic effect also improves the sensor's reliability and durability, which enhances the overall performance of the sensor.

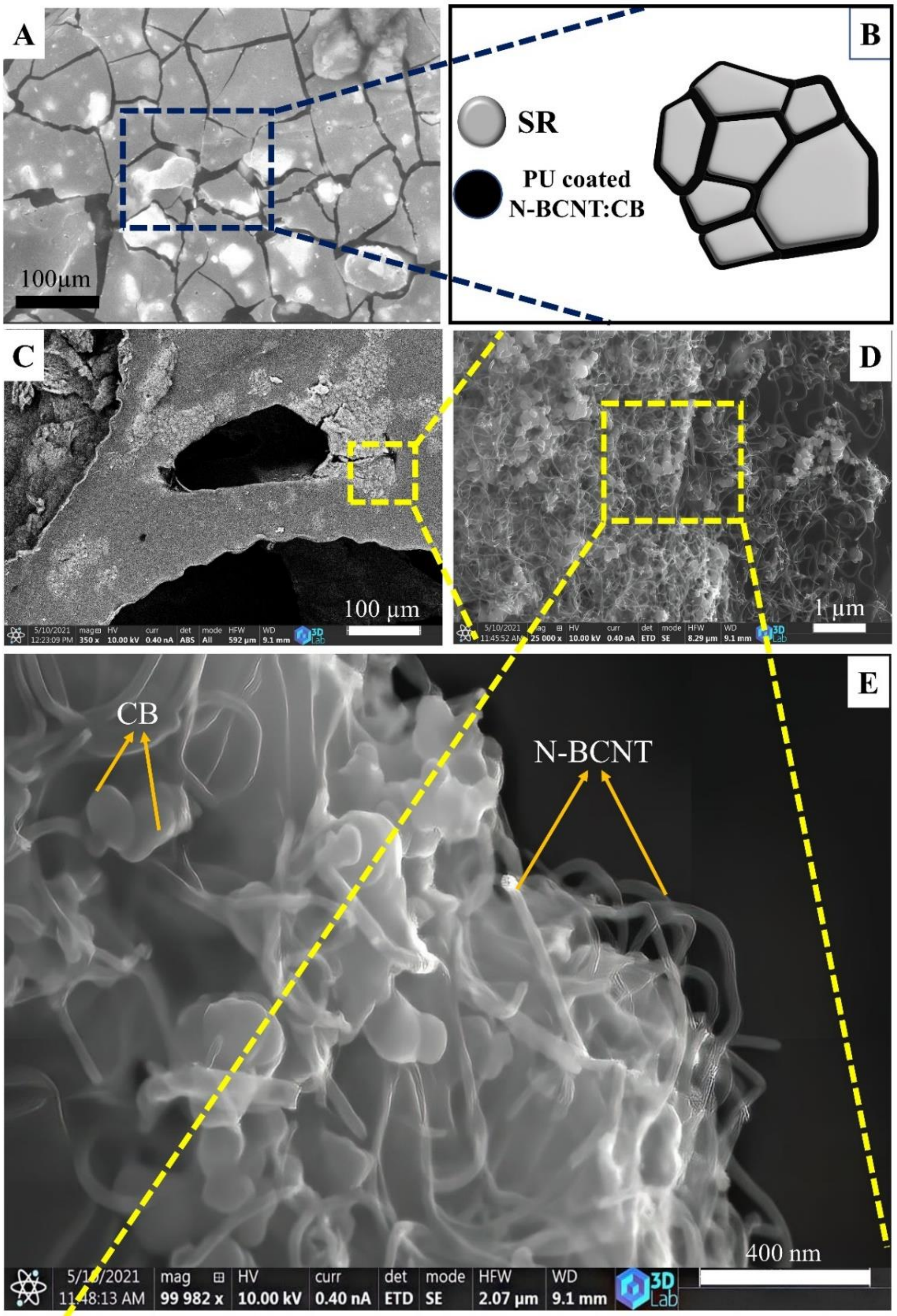

Figure 3 Morphology of the developed N-BCNT:CB/PU-SR composite-based e-skin (A). Schematic represented of the incorporation of SR surrounding the N-BCNT:CB coated PU (B). SEM images of the N-BCNT:CB/PU (C-D), and showcase of the synergistic effect between N-BCNT and CB (E).

5. Application of e-skin

Figure 4 demonstrate the relative change in resistance over time for a flexible pressure sensor designed to detect finger, wrist, elbow, and knee movements as well as twisting and stretching. The data demonstrate the sensor's excellent sensitivity and responsiveness to different movements, as evidenced by the distinct waveforms on each curve for each motion. The pressure sensor's high sensitivity is achieved through the combination of N-BCNT and CB, with N-BCNT providing electrical conductivity while CB fills any gaps between them. This working relationship enables the sensor to detect and respond to various movements. Additionally, the good dispersion of the nanofiller and the high flexibility of the SR contribute to the sensor's sensitivity. The efficient transfer of movements within the SR facilitates efficient detection, further increasing its response. On the other hand, the high flexibility of the SR enables it to detect movements even when deformed. These findings indicate that pressure sensors have potential applications beyond e-skin applications where accuracy in detection and response to different movements is essential.

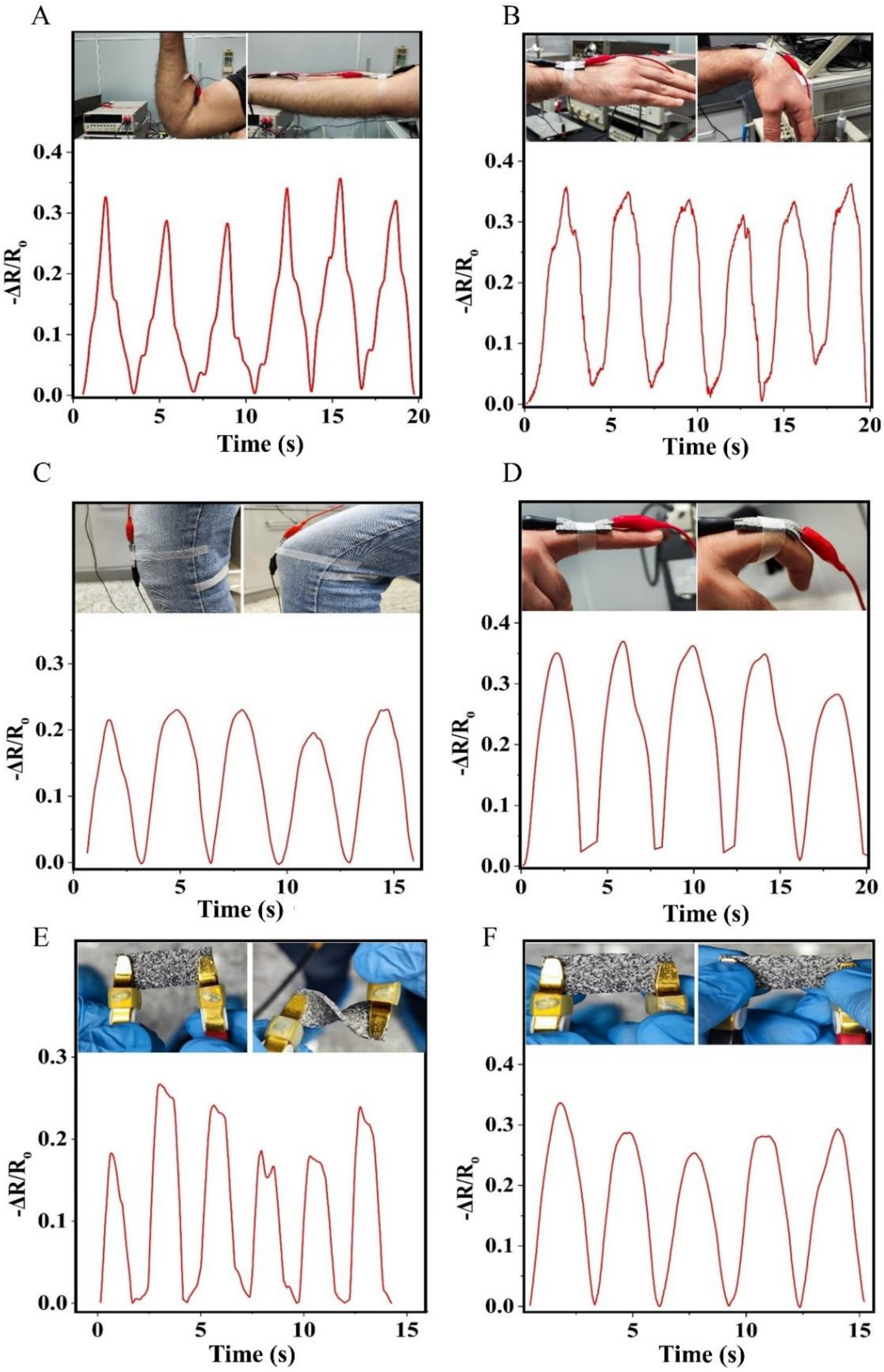

Figure 4 The N-BCNT:CB/PU-SR composite-based e-skin tested to detect various movements: (A) elbow, (B) wrist, (C) index finger, and (D) knee, as well as (E) twists and (F) stretching movements.

6. Conclusion

Flexible pressure sensors have a wide range of applications in areas such as e-skin and wearable technology. Nanocomposites consisting of nanofillers combined with a polymer matrix have shown tremendous potential and are more cost-effective than other conventional methods. Fabrication of flexible pressure sensors using nanocomposite technology primarily involves converting insulating polymers into conductive ones by mixing them with conductive nanofillers to sense the change in electrical resistance in response to applied pressure. To efficiently utilize the properties of nanofillers in FPS, it is critical to create a continuous conductive network pathway within the polymer matrix and achieve an electrical conductivity percolation threshold. In conventional approaches such as direct mixing and mixing in solution, the nanofiller loading must be increased to reach the electrical threshold. However, when the nanofiller loading is high, bundles are more likely to occur, this bundling causes an increase in the viscosity of the polymer matrix, which affects sensor performance. Also, the use of high-power ultrasonication and shear mixing to achieve uniform dispersion of nanofillers in the polymer matrix. However, this approach can lead to thermal degradation and structural damage, which affects the durability of the sensor. This is because the high power can cause the polymer chains to break in the center point or at the structurally weakest points. An alternative approach is to disperse nanofillers on a foam skeleton using an immersion process. This structure is then impregnated and combined with a polymer to produce the final sensor system. In this approach, the nanofiller spans the entire volume of the polymer and provides electrical conductivity in the macroscopic dimension of the sensor. It also minimizes the negative impact on the sensor's performance since no further processing is required. Using this approach, a flexible pressure sensor was fabricated for use in e-skin applications, with the lowest added weight percentage (0.4) compared to previous research.

New Scientific Results

Thesis 1

Fabrication of a novel flexible pressure sensor for e-skin applications with very low nanofiller loading. Moreover, compared to other works, our pressure sensor satisfies the electrical conductivity at nano-filler loading as low as (0.4 wt.%).

Thesis 2

Improve the dispersibility of the nanofiller through a unique technique for the first time by dispersing it on the PU foam skeleton to create a continuous conductive network, which is then impregnated in silicone rubber. This approach is like an implanted conductor network in the insulating silicone rubber. Each of these materials has a specific role: PU serves as the supporting skeleton, the nanofiller provides the electrical conductivity to measure the change in resistance when pressure is applied, and the silicone rubber serves as the matrix and provides the flexibility and elasticity of the nanocomposite.

Thesis 3

A novel investigation of the effect of the pore volume of PU foam by designing three different types of PU prepared as the supporting skeleton for the nanofiller, using isocyanate indices of 1.0:0.8, 1.0:1.0, and 1.0:1.1 to obtain different pore volumes of PU foam. Our work revealed that the best isocyanate index was 1.0:0.8 because the synthesized PU foam has a higher pore volume than the others. Regarding the pressure sensitivity of the pressure sensor, the results show that the PU foam with high pore volume has the highest pressure sensitivity when used as a supporting skeleton for the nanofiller. Since a high total pore volume leads to a lower PU foam scaffold, more nanofillers are consequently interconnected, resulting in more conductive paths and a larger effective conductive area in the PU scaffold. In addition, a large pore volume provides suitable spatial pores for elastic deformation of flexible pressure sensors.

Thesis 4

Using cutting- edge technology, I evaluate the pore volume from PU is based for the first time on micro-CT, which offers higher accuracy than conventional methods such as mercury intrusion porosimeter, gas adsorption, and SEM. Micro-CT is the best choice because scanning the sample produces a 3D image that enables highly accurate measurements for the analysis of pore size and polyurethane distribution. Micro-CT technology offers a highly accurate method to precisely measure the pore volume within polyurethane foam. Its excellent technology in visualizing each individual pore, a unique advantage over traditional techniques. With its detailed imaging, it preserves the foam's structure without causing damage. Thus, Micro-CT is considered a valuable tool, to in-depth exploration of the foam's pore structure and providing critical insights into pressure sensor behavior and performance. In addition, micro-CT provides a complete understanding of the pore size and distribution of polyurethane foams, which allowed us to accurately investigate the relationship between pore size and pressure sensitivity.

Thesis 5

Novel design of a flexible pressure sensor by impregnation with silicone rubber to fill all pores of the PU foam. In the absence of silicone rubber, the nanofiller could be unintentionally removed from the nanofiller/ PU system when pressure is applied, as only van der Waals forces hold them together. Furthermore, impregnation with silicone rubber can mitigate the toxicity of CNTs in several ways. First, it can create a barrier that prevents CNTs from entering the environment. Second, it can interact with the surface of the CNTs and change their surface chemistry, making them less toxic. I believe that my work makes an important contribution to CNT research by proposing a method to reduce the toxicity of CNTs. This is an important topic as it could enable the use of CNTs in a wider range of applications, including those where humans may be at risk. While my work proposes a technique to reduce CNT toxicity, further research is needed to fully understand the long-term health effects of exposure to CNT-silicone rubber composites.

Thesis 6

Optimization of nanofiller using different mixing ratios (5:5, 6:4, 7:3, 8:2, 9:1) of N-BCNT: CB on the behavior of the flexible pressure sensor investigated. The results show that the optimal mixing ratio is 7:3 N-BCNT: CB, which is even better than N-BCNT alone. This mixing ratio is optimal for pressure sensitivity due to the synergistic effect of the nanofillers. When the ratio of N-BCNT to CB goes beyond 7:3, it becomes more difficult for the N-BCNT to align and expand, and the relative amount of CB also decreases, resulting in minimal contact between the fillers. On the other hand, the reduction of the N-BCNT fraction decreases the ability of electrical charge transport over long distances along the aligned and extended nanotubes.

List of publications

1) **Mohammed Nabeel**, Mousa, M., Viskolcz, B., Fiser, B., & Vanyorek, L. Recent Advances in Flexible Foam Pressure Sensors: Manufacturing, Characterization, and Applications – a Review. *Polymer Reviews*, 2023, 1–41. https://doi.org/10.1080/15583724.2023.2262558 . **Q1 (IF: 14.5)**

2) **Mohammed Nabeel**, László Kuzsella, Béla Viskolcz, Mariann Kollar, Béla Fiser, László Vanyorek, 2023. Synergistic Effect of Carbon Nanotubes and Carbon Black as Nanofillers of Silicone Rubber Pressure Sensors. Arabian Journal of Chemistry 104594. https://doi.org/10.1016/j.arabjc.2023.104594. **Q1 (IF: 6.21)**
3) **Mohammed Nabeel**, Miklós Varga, László Kuzsela, Béla Fiser, Béla Viskolcz, László Vanyorek, 2022. The Effect of Pore, Volume on the Behavior of Polyurethane-Foam-Based Pressure Sensors. Polymers (Basel) 14, 3652. doi.org/10.3390/polym14173652. **Q1 (IF: 4.9)**
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5) **Mohammed Nabeel**, Ali J. Addie, Béla Viskolcz, Mariann Kollar, Béla Fiser, László Vanyorek, Development of Flexible Pressure Sensors Based on Polyurethane-Silicone Rubber-Nanocomposite Systems, Arabian Journal of Chemistry. **Under review Q1 (IF: 6.21).**

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