

Article

Liquid-Phase Exfoliated Graphene and Polytetrafluoroethylene for Highly Durable and Reusable Chemical Leak Detection Sensors

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Abstract: Graphene-based chemical sensors hold promise across diverse applications owing to their exceptional sensitivity and selectivity. However, achieving their long-term durability and reusability while preserving high sensitivity remains a significant challenge, particularly in harsh environments where exposure to strong chemicals is inevitable. This paper presents a novel approach to address this challenge by synergistically integrating liquid-phase exfoliated graphene (LPEG) with polytetrafluoroethylene (PTFE) within a single sensing strip. Through a comprehensive experimental investigation, we demonstrate the fabrication of highly durable and reusable chemical leak detection sensors by combining LPEG and PTFE. Furthermore, we explore the sensing mechanism, highlighting the roles of LPEG and PTFE in enhancing sensitivity and selectivity, along with durability and reusability. Performance evaluation reveals the sensors' robustness against mechanical and chemical degradation, coupled with excellent recyclability. This innovative approach holds promise for applications in environmental monitoring, industrial safety, and healthcare, thus advancing the field of graphene-based chemical leak detection sensors.

Keywords: graphene; liquid-phase exfoliation; sensors; graphene/PTFE; acid sensing



Citation: Rubab, N.; Sohn, E.; Kang, W.-S.; Kim, T. Liquid-Phase Exfoliated Graphene and Polytetrafluoroethylene for Highly Durable and Reusable Chemical Leak Detection Sensors. *Chemosensors* **2024**, *12*, 77. <https://doi.org/10.3390/chemosensors12050077>

Received: 29 February 2024

Revised: 5 April 2024

Accepted: 30 April 2024

Published: 7 May 2024



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1. Introduction

Graphene, a two-dimensional carbon allotrope, has emerged as a revolutionary material with remarkable properties, including high electrical conductivity, exceptional mechanical strength, and a large surface area-to-volume ratio [1–4]. These unique characteristics make graphene an attractive candidate for various applications, including electronics, energy storage, and sensing [5–11]. In particular, graphene-based chemical sensors have garnered significant attention due to their potential in the highly sensitive and selective detection of analytes in diverse environments [12–14].

Chemical sensors play a critical role in the industrial, environmental, and healthcare sectors by monitoring the presence of specific chemicals or gases [6,10,15,16]. However, ensuring the long-term durability and reusability of these sensors remains a challenge, especially when exposed to harsh operating conditions. In particular, the detection of chemical leaks in commercial storage settings poses significant challenges due to the corrosive nature of many chemicals and the potential for mechanical breakdown of sensor components.

Despite notable strides in developing graphene-based chemical sensors, previous endeavors have encountered limitations, particularly in achieving sustained performance in rigorous acidic or basic environments. Conventional chemical sensors have typically been fabricated using active sensing materials, such as carbonaceous materials, metal oxides, or polymers, coated onto a substrate to detect specific gases or chemicals. However, these sensor designs often fail when deployed in environments where strong chemicals are present due to several reasons. Firstly, the coatings on these sensors are prone to

degradation when exposed to corrosive chemicals, leading to a loss of sensitivity and selectivity over time. Additionally, the adhesion between the coating and the substrate is often insufficient, resulting in delamination or detachment of the sensing layer. These shortcomings have rendered conventional chemical sensors unsuitable for long-term use in harsh chemical environments, highlighting the need for alternative sensor designs with improved durability and stability.

In this context, this study presents a novel approach to address the challenges associated with developing highly durable and reusable chemical leak detection sensors. By synergistically integrating liquid-phase exfoliated graphene (LPEG) with polytetrafluoroethylene (PTFE) and embedding these LPEG–PTFE sensing components into a sensing strip, we aim to enhance the sensor's performance and robustness against chemical degradation. This combination of material selection and sensor design offers the potential for creating sensors capable of withstanding harsh operating conditions while maintaining sensitivity and selectivity.

Our approach was twofold. Firstly, embedding graphene-based sensing components into a sensing strip instead of simply coating the strip with a graphene sensing layer offers several advantages, particularly when the sensors are exposed to strong chemicals such as strong acids and bases. Embedding the graphene-based sensing part ensures better integration and adhesion between the sensing material and the substrate, enhancing the overall robustness and stability of the sensor. This integration minimizes the risk of delamination or detachment of the sensing layer, which can occur with coated sensors, especially when subjected to aggressive chemical environments. Additionally, embedding the sensing component allows for more precise control over the thickness and uniformity of the graphene layer throughout the sensing strip, ensuring consistent and reliable performance. Moreover, embedding the sensing part into the strip provides better protection against physical damage or abrasion, which can compromise sensor integrity and performance over time. Therefore, the embedding approach offers superior durability, stability, and performance compared to coating, particularly in harsh chemical environments, making it a preferable choice for graphene-based chemical leak detection sensors.

Secondly, our approach utilizes liquid-phase exfoliated graphene as the active sensing material in fabricating graphene-based resistive chemical leak detection sensors, offering distinct advantages. The liquid-phase exfoliation method enables the production of graphene flakes with high quality and consistency, overcoming the scalability limitations and control challenges associated with traditional graphene synthesis methods [17–22]. This approach ensures the uniform dispersion of highly conductive graphene flakes within the sensing material, facilitating enhanced sensitivity and uniform response across the sensor surface. Additionally, liquid-phase exfoliated graphene exhibits superior electrical properties compared to chemically modified graphene, making it particularly well suited for sensor applications where high conductivity or low resistance is essential. Thus, the use of liquid-phase exfoliated graphene enhances the performance, reliability, and scalability of graphene-based chemical leak detection sensors, further emphasizing its significance in sensor fabrication.

Moreover, the incorporation of polytetrafluoroethylene (PTFE) into the sensor design is also crucial for developing durable and reusable chemical leak detection sensors. PTFE, known for its exceptional chemical-resistant properties, enhances the sensor's robustness against chemical degradation. By integrating PTFE with liquid-phase exfoliated graphene, our approach aims to create sensors capable of withstanding harsh operating conditions while maintaining sensitivity and selectivity. This combination ensures long-term durability and reusability, addressing critical challenges in practical sensor implementation, especially in environments exposed to strong acids and bases. This paper explores the fabrication methodology, structural characteristics, sensing mechanism, and performance metrics of the developed sensors. Furthermore, we discuss the implications of this research for expanding the applications of graphene-based sensors and addressing challenges associated with their practical implementation. Overall, this study contributes to advancing the field of chemical

sensing technology, offering promising avenues for the development of highly durable and reusable sensors for diverse industrial, environmental, and healthcare applications.

2. Materials and Methods

Liquid-phase exfoliated graphene flakes were utilized as active materials for the chemical leak detection sensors [19,23]. Graphite powder was sourced from Sigma-Aldrich (Saint Louis, MO, USA) (282863) and Bay Carbon (SP-1), with particle sizes ranging from 1 to 10 μm and 100 to 200 μm , respectively. Dimethylformamide (DMF) and N-Methyl-2-pyrrolidone (NMP), purchased from Sigma-Aldrich, were employed as solvents for graphene production. Both the graphite powder and the solvents were used as received. Isopropanol (IPA) and deionized water were employed to precipitate the liquid-phase exfoliated graphene flakes from the dispersion, to separate the graphene flakes and remove by-products and impurities. Polytetrafluoroethylene (PTFE) with a particle size of 20 μm was purchased from Sigma-Aldrich.

2.1. Liquid-Phase Exfoliation of Graphene

Graphene dispersions were prepared by dispersing 500 mg of graphite powders in 100 mL of organic solvents at a concentration of 5 mg mL^{-1} and adding 500 mg of sodium hydroxide (NaOH) to the dispersion. Liquid-phase exfoliation of graphite was conducted through microfluidization to produce few-layer graphene, as depicted in Figure 1A. This method utilized a high-pressure homogenizer to exert pressure on the fluid containing graphite, pushing it through a microchannel. Exfoliation was driven by cavitation, high shear stress, and particle collision. The liquid-phase exfoliation of graphite into graphene involved the following steps: (i) high-shear mixing of the graphite in the solvent at 6000 rpm for an hour to achieve a homogeneous dispersion of graphite in a solvent; (ii) further processing of the dispersion in a paste mixer at 500 rpm for two minutes, with the process repeated three times at intervals to enhance dispersion homogeneity; and (iii) loading of the dispersion into a high-pressure homogenizer for the shear-controlled exfoliation of graphite into graphene. The graphene synthesis was effectively accomplished at this stage through microfluidization based on shear force-controlled exfoliation. Unexfoliated graphite particles were removed by centrifugation, and the exfoliated graphene in the supernatant was separated for further analysis. Deionized (DI) water and isopropyl alcohol (IPA) were then added to precipitate the liquid-phase exfoliated graphene flakes from the dispersion and remove impurities. The precipitated graphene flakes were collected and freeze-dried.

2.2. Fabrication of Graphene–PTFE (LPEG–PTFE) Composite for Chemical Leak Detection Sensors

Liquid-phase exfoliated graphene flakes were mixed with PTFE binders using a powder mixer. The resulting graphene–PTFE (LPEG–PTFE) mixture was then loaded into a circular mold and sandwiched between two layers of PTFE powders with a stacking order of PTFE/LPEG–PTFE/PTFE. The stacked layers in the mold were then heated and pressed into a disc shape using a hot press, as presented in Figure 1C. The circular disc of PTFE/LPEG–PTFE/PTFE was sliced into a thin LPEG–PTFE-based sensor strip using a skiving machine, as depicted in Figure 1C. The fabricated LPEG–PTFE-based sensor strips were 15 m in length, as shown in Figure S4A.

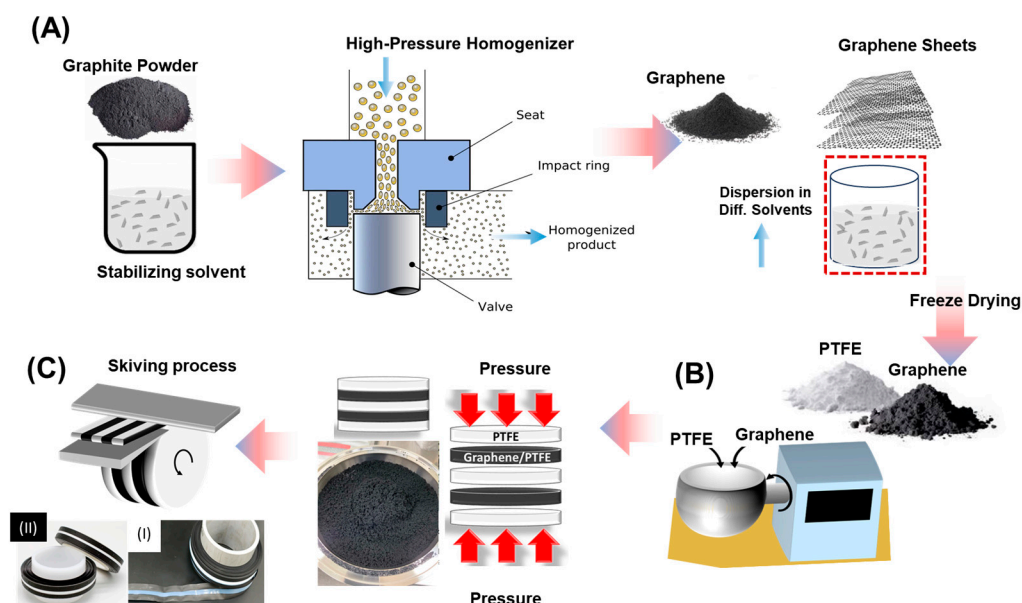


Figure 1. Concept and structure of the chemical leak detection sensors. (A) Schematic representation of liquid-phase exfoliation of graphite into few-layer graphene through microfluidization using a high-pressure homogenizer. (B) Freeze-dried liquid-phase exfoliated graphene flakes were mixed with PTFE binders and hot-pressed to form a disc of PTFE/LPEG-PTFE/PTFE. (C) PTFE/LPEG-PTFE/PTFE discs were sliced into a LPEG-PTFE-based sensor strip using a skiving machine. The actual image of the Gr-PTFE-based sensor strip is shown.

3. Results

Characterization of the Liquid-Phase Exfoliated Graphene Flakes

Liquid-phase exfoliated graphene was produced through microfluidization using a high-pressure homogenizer from the dispersion of graphite in organic solvent. Prior to the liquid exfoliation of graphite in the organic solvent, sodium hydroxide (NaOH) was added to improve the peeling efficiency of graphite and the yield of graphene. The added NaOH appeared to intercalate into the graphite, increasing the interplanar distance of the graphitic layers and weakening the interplanar interaction. This resulted in a significant improvement in the production yield of the graphene flakes [24]. After the liquid-phase exfoliation of graphite, the produced few-layer graphene was collected by the sedimentation of graphene from the dispersion. The gradual sedimentation of graphene was possible when solvents such as DI water and IPA were added, both of which had surface tension and surface energy far different from that of graphene and NMP. The precipitated graphene flakes were then further separated from the solvent by centrifugation and freeze-dried. Finally, the graphene flakes were obtained in the form of powder and used for the fabrication of the sensor and analysis.

The morphologies of the liquid-phase exfoliated graphene flakes were characterized by TEM and SEM. Figure 2 shows the TEM and SEM images of the graphene flakes precipitated and separated from the graphene dispersion by adding non-solvents such as DI water and IPA to the dispersion. The TEM images show few-layer graphenes with lateral sizes typically of a few micrometers, as represented in Figure 2A,B [25]. In some regions, we observed wrinkled graphene sheets and few-layer graphene sheets with their edges folded slightly. No structural defects, such as vacancies, were found. However, the SEM image of the graphene flakes precipitated by adding IPA to the dispersion showed a considerable number of particulate impurities on the graphene surfaces, as shown in Figure 2D. These impurities appeared to be sodium hydroxide (NaOH), or sodium salts solidified during the freeze-drying process, indicating that IPA was ineffective in completely removing these impurities and achieving high-quality graphene.

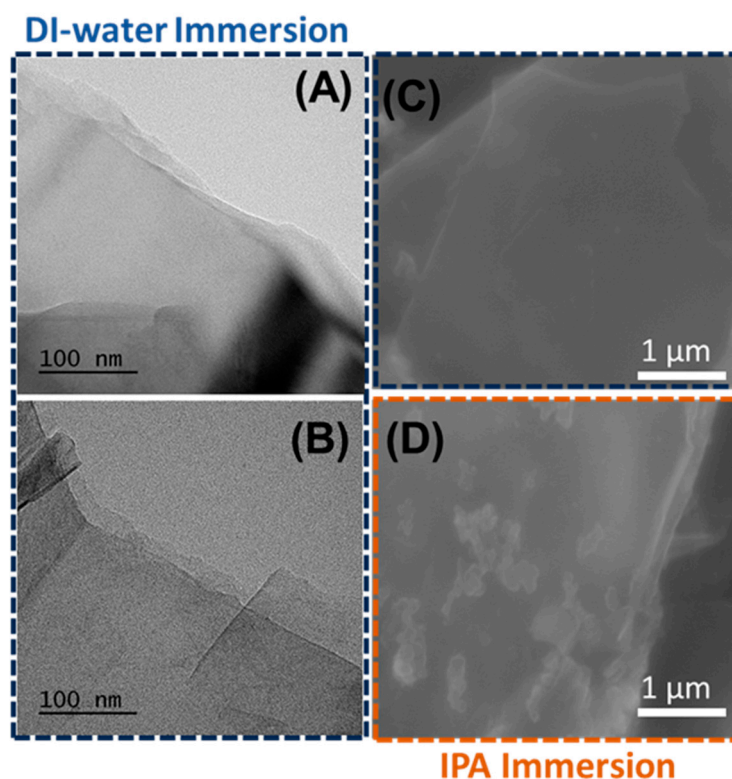


Figure 2. The morphologies of liquid-phase exfoliated graphene flakes. TEM images of (A) DI water-added graphene and (B) IPA-added graphene. SEM images of (C) DI water-added graphene (D) IPA-added graphene.

In contrast, the graphene flakes precipitated in DI water exhibited an evident and identical shape. TEM images of the graphene displayed a partially folded mono-layer or few-layer structure, indicating excellent peeling effects. This finding suggests that adding DI water to graphene dispersion yields individual graphene layers with minimal aggregation or grouping. SEM analysis also revealed that the graphene flakes precipitated in DI water were devoid of particulate impurities. This observation indicated that adding DI water to the graphene dispersion effectively cleansed the graphene flakes and removed impurities.

We conducted a comparison and analysis of the powder conductivity of graphene flakes synthesized under various conditions. Notably, the results highlighted the impact of the solvents added to the graphene dispersion on the powder conductivity of the graphene flakes. Figure 3 shows that the electrical conductivity of the graphene produced by adding DI water was as high as 1150 S/m at 20 MPa, which surpassed the conductivity values of graphene flakes with IPA addition. The results indicated that DI water-added graphene flakes were devoid of impurities, facilitating close contact between each graphene layer and resulting in low contact resistance. The powder conductivity of the graphene flakes was measured using two electrodes and a four-probe method. The powder conductivity was measured using the two-electrode method and plotted against the packing density of the powered graphene flakes, as in Figure 3. Using the four-probe method, the power conductivity of the graphene flakes was measured to be 1150 S/m at a pressure of 20 MPa.

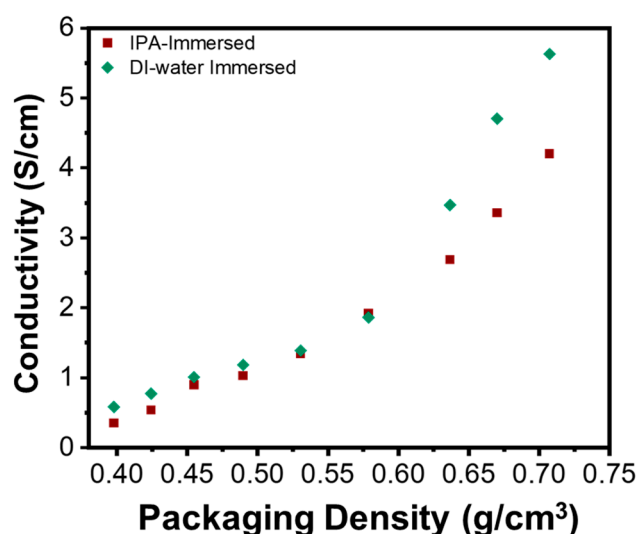


Figure 3. Comparison of powder conductivity of DI water-added and IPA-added graphene flakes.

The quality of graphene was evaluated through Raman spectroscopy, as depicted in Figure 4. The liquid-exfoliated graphene flakes displayed a sharp G peak at 1570 cm^{-1} and a D peak at 1355 cm^{-1} , along with a 2D peak at 2700 cm^{-1} . In the Raman spectra of DI water- and IPA-added graphene samples, a noticeable difference was observed in the ratio of the D peak, which signifies structural defects, and the G peak, indicative of graphite structure. Specifically, the intensity ratio between the D peak and the G peak (I_D/I_G) for DI water- and IPA-added graphene flakes was measured at 0.57 and 0.83, respectively. The higher I_D/I_G ratio for the IPA-added graphene samples suggests a more significant presence of defects, resulting in reduced graphene quality. These findings suggest that the quality of the graphene can be controlled by adding different solvents for the sedimentation of liquid-phase exfoliated graphene from the dispersion. Notably, DI water-added graphene flakes exhibited higher powder conductivity and a lower value of I_D/I_G , indicating superior quality.

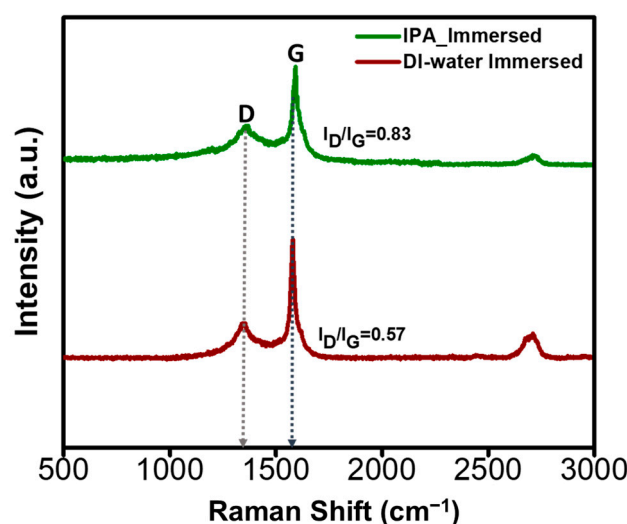


Figure 4. Raman spectra of liquid-phase exfoliated graphene flakes obtained by adding water and IPA to the graphene dispersion.

To fabricate the sensing strip, liquid-phase exfoliated graphene flakes were blended with PTFE binders to create a graphene-PTFE (LPEG-PTFE) composite, which was then encased between two layers of PTFE within a circular mold and compressed into a PTFE/LPEG-PTFE/PTFE disc. Using a skiving machine, the disc was sliced into a thin

strip measuring 15 m in length, composed of PTFE/LPEG–PTFE/PTFE. This fabrication method offers scalability, facilitating the continuous production of sensor strips. Within the sensor strip, the sensing component (LPEG–PTFE) is seamlessly integrated and embedded into the PTFE substrate, resulting in a single-layer structure. This design enhances durability, enabling multiple uses even in harsh environments. In contrast, conventional sensors typically involve the coating or printing of graphene layers onto the substrate, resulting in a multi-layered structure that is susceptible to delamination upon exposure to strong chemicals.

Figure 5 illustrates the reliability of LPEG–PTFE sensors under repeated exposure to various chemicals. Sections of LPEG–PTFE sensors were submerged in three different chemicals—sulfuric acid (H_2SO_4), hydrofluoric acid (HF), and sodium hydroxide (NaOH)—and were then taken out to measure their resistances. This process was repeated three times and results were recorded. Remarkably, the sensors maintained stability and exhibited consistency, demonstrating their robustness in harsh environments. The stability of the sensors is further demonstrated in Table S2, which details the resilience of the sensors. The bar graph in Figure 5 displays the resistance of the sensors before and after the immersion test. Notably, there was no significant difference in resistance after exposures, with only a negligible resistance change observed, typically only a few ohms. This consistency indicates the durability of the sensor strip even after exposure to challenging conditions.

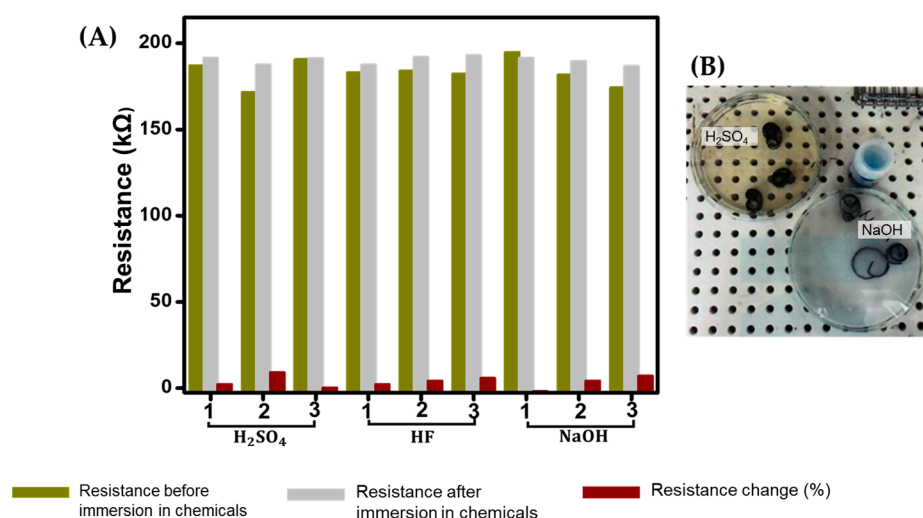


Figure 5. (A) Resistance measurements of LPEG–PTFE sensors before and after immersion in different chemicals: sulfuric acid, hydrofluoric acid, and sodium hydroxide. (B) Photograph of LPEG/PTFE Sensors after the immersion test.

Following the successful synthesis and stability testing of the LPEG–PTFE sensor, the sensitivity of the sensor was further assessed through a series of tests involving various chemicals and concentrations, as depicted in Figure 6. For sensitivity evaluation, sensor lengths of 1 m were utilized, divided into three sections (start, middle, and end), to conduct tests with different chemicals: water, 10% sulfuric acid (H_2SO_4), and 5% sodium hydroxide (NaOH). Resistance values were measured using a resistance meter, as illustrated in Figure 6A. Each test upon exposure to different chemicals lasted approximately 30 s. The “ON” point denotes the exposure of sensors to the chemicals, while “ R_{Drop} ” signifies the point at which resistance immediately dropped to the minimum upon contact with the chemicals. Subsequently, when the chemicals on the sensor strip were removed, the resistance of the sensors recovered back to original value within 5 to 25 s and stabilized afterwards, as depicted in Figure 6B. These results provide compelling evidence of the durability and reusability of the sensors, making them suitable for industrial and commercial applications. Chemical leak detection sensors with durability and reusability have the potential to revolutionize various industries by offering continuous, reliable, and real-time

monitoring of harsh environments. The integration of such sensors into industrial settings can significantly enhance maintenance practices, monitoring capabilities, and safety protocols.

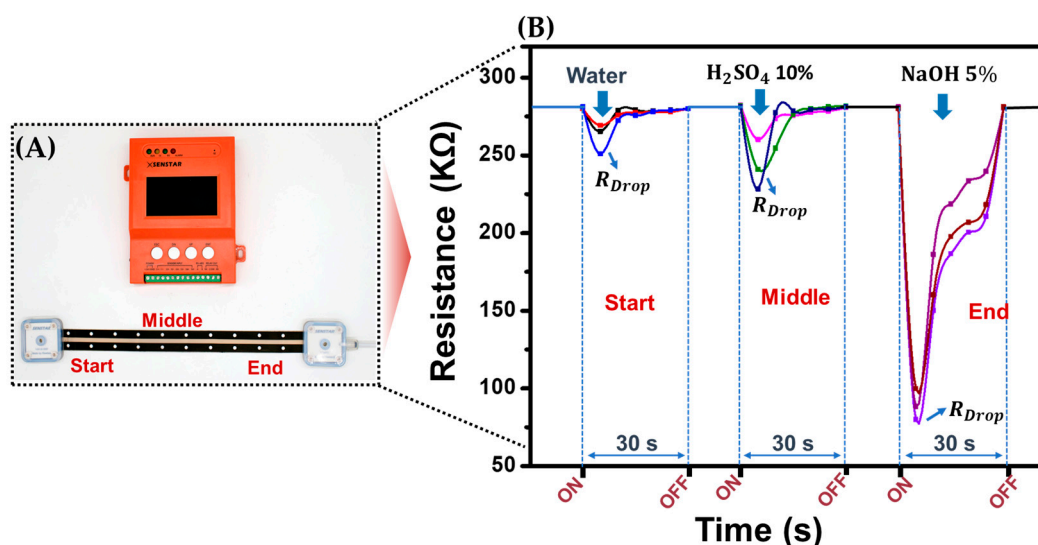


Figure 6. Response of LPEG–PTFE sensors towards different chemicals. (A) Photograph of LPEG–PTFE sensors. (B) Response of the LPEG–PTFE sensors upon contact with water, 10% H₂SO₄, and 5% NaOH solution. The resistance change of the sensors was plotted as a function of time.

The sensing mechanism of the LPEG–PTFE sensor involves a combination of electronic and physical processes. Within the LPEG–PTFE sensor strip, two parallel LPEG–PTFE sensing lines, distinguished by their black color, are separated by an insulating PTFE line. These sensing lines are electrically connected at one end and open at the other, with resistances measured along a length of 1 m. When chemicals with ionic conductivities, such as water, sulfuric acid, and sodium hydroxide, are introduced and placed between the two sensing lines, they establish a conductive path between the sensing lines, leading to a short circuit and a subsequent decrease in resistance due to the reduced current path. The extent of resistance change depends on the ionic conductivity of the chemicals, increasing in the order of water, 10% sulfuric acid, and 5% NaOH. Additionally, the graphene component of the sensor may contribute to this response by undergoing changes in its electronic state upon contact with the chemicals, further influencing the overall resistance of the sensor. Consequently, the LPEG–PTFE sensor exhibits a sensitive response to the presence of conductive liquids, with the magnitude of the response varying depending on the conductivity of the chemicals. This versatile sensing platform offers reliability and applicability across industrial, environmental, and healthcare settings.

Figure S7 presents the results of stability and sensitivity testing conducted on the sensors across various chemical environments. Notably, a sensitivity value of 225.9 ohms was observed for 10% NaOH, indicating the highest sensitivity among the tested conditions (Figure S7A). Additionally, Figure S5 offers a comprehensive analysis of sensor performance in different environments, including water, acetone, NaOH, and H₂SO₄. This emphasizes the sensors' capability of detecting acidic and basic chemicals and demonstrates their consistent output performance, thus highlighting their reliability under diverse chemical conditions. Furthermore, Tables S2 and S3 provide detailed results from testing LPEG–PTFE sensors with strong acids such as hydrofluoric acid (HF) and sulfuric acid (H₂SO₄) at various concentrations. These tables underscore the sensors' robustness in withstanding harsh chemical environments while maintaining reliable sensing capabilities.

4. Discussion

In this study, we introduce an innovative approach aimed at addressing the challenges inherent in developing highly durable and reusable chemical leak detection sensors. Our strategy encompasses two key elements to overcome the limitations typically encountered with graphene-based chemical leak detection sensors. Firstly, we implement a novel method of embedding graphene-based sensing components into a sensing strip, departing from the conventional approach of simply coating the strip. This embedding technique offers several advantages, particularly in environments with strong acids and bases. By ensuring better integration and adhesion between the sensing material and the substrate, this approach enhances overall robustness and stability. Secondly, we leverage the use of liquid-phase exfoliated graphene as an active sensing material, ensuring the uniform dispersion of highly conductive graphene flakes within the sensing material. This results in enhanced sensitivity and uniform response across the sensor surface. Additionally, liquid-phase exfoliated graphene exhibits superior electrical properties compared to chemically modified graphene, rendering it particularly suitable for applications where low resistance is essential.

Furthermore, the integration of polytetrafluoroethylene (PTFE) into the sensor design plays a pivotal role in developing durable and reusable chemical leak detection sensors. With its excellent chemical-resistant properties, PTFE enhances the sensor's robustness against potential failure. By combining PTFE with liquid-phase exfoliated graphene, our approach demonstrates the creation of sensors capable of withstanding harsh operating conditions while maintaining sensitivity and selectivity. This synergistic combination ensures long-term durability and reusability, effectively addressing critical challenges in practical sensor implementation, especially in environments exposed to strong acids and bases.

Overall, our study contributes significantly to the advancement of chemical sensing technology by offering a comprehensive approach to developing highly durable and reusable sensors for a wide range of industrial, environmental, and healthcare applications. These advancements hold great promise for improving monitoring capabilities, enhancing safety protocols, and enabling real-time detection of chemical leakages in various settings. Looking ahead, future directions for graphene-PTFE-based chemical leak detection sensors include further enhancing sensitivity by optimizing the structure and properties of composite sensors based on graphene-PTFE, thereby addressing critical needs in various industrial and commercial applications requiring reliable and efficient detection of chemical leakages.

5. Conclusions

In conclusion, this study has demonstrated the successful fabrication and characterization of graphene-based chemical leak detection sensors with polytetrafluoroethylene (PTFE) integration, offering promising avenues for enhancing sensor durability and sensitivity. Through the synergistic combination of liquid-phase exfoliated graphene (LPEG) with PTFE binders, sensors with superior stability and sensitivity were achieved, as evidenced by the comprehensive testing conducted in various chemical environments. The sensors exhibited remarkable performance in detecting and responding to conductive liquids, including strong acids and bases, showcasing their versatility and reliability for real-world applications. Moreover, the scalability of the fabrication process ensures the feasibility of mass production, further enhancing the practicality and accessibility of these sensors. Overall, the developed graphene-based chemical leak detection sensors hold significant potential for revolutionizing the industrial, environmental, and healthcare sectors by offering continuous, reliable, and real-time monitoring capabilities in harsh chemical environments. Future research directions may focus on optimizing sensor design and exploring additional functionalities to address specific application requirements, thereby further advancing the field of chemical sensing technology.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/chemosensors12050077/s1>, Figure S1: Fabrication of LPEG-PTFE composite and sensor strips. Figure S2: Photographs of LPEG-PTFE sensor and system. Figure S3: Sheet resistance measurements of the LPEG-PTFE sensors. Figure S4: Resistance of the LPEG-PTFE sensors upon contact with water, 10% H₂SO₄, and 5% NaOH solution. Table S1. Electrical conductivity analysis results. Table S2: Corrosion Resistance Test of the LPEG-PTFE sensor. Table S3: Sheet Resistance measurements of LPEG-PTFE sensor strip. Table S4: Dust resistance test of the LPEG-PTFE sensors. Table S5: Thermal Shock Test of the LPEG-PTFE sensors.

Author Contributions: Conceptualization, W.-S.K. and T.K.; Methodology, E.S. and W.-S.K.; Validation, N.R.; Investigation, N.R. and E.S.; Data curation, E.S. and W.-S.K.; Writing—original draft, N.R.; Writing—review & editing, T.K.; Supervision, W.-S.K. and T.K.; project administration, W.-S.K. and T.K.; funding acquisition, W.-S.K. and T.K.. All authors have read and agreed to the published version of the manuscript.

Funding: This work was supported by the National Research Foundation of Korea (NRF) grant funded by the Korean government (MSIT) (No. 2022R1A2C1093082). This work was supported by Nano-Convergence Foundation (www.nanotech2020.org) funded by the Ministry of Science and ICT (MSIT, Korea) & the Ministry of Trade, Industry and Energy (MOTIE, Korea). [Project Name: Development of non-printing integrated toxic chemical detection sensor based on graphene/Project Number: R202100800]. This work was supported by the Gachon University research fund of 2019 (GCU-2019-0832).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Data is contained within the article or supplementary material.

Acknowledgments: We thank Mitesh G. Mapari at Gachon University for measuring the powder conductivity of liquid-exfoliated graphene.

Conflicts of Interest: Authors Eunbee Sohn and Won Seok Kang were employed by company FLOWTECH Co., Ltd. The remaining authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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