



## Dielectric and Thermal Properties of One-Pot Hydrothermal Synthesis of Exfoliated Molybdenum Disulfide Doped In Polyvinylidene Fluoride Nanocomposite



Abdallah A. Shaltout<sup>a</sup>, Nasser Y. Mostafa<sup>b</sup>, Ragab M. Mahani<sup>c\*</sup>, Mohamed M. Ibrahim<sup>d</sup>

<sup>a</sup>Spectroscopy Department, Physics Research Institute, National Research Centre, El Behouth St., 12622 Dokki, Cairo, Egypt

<sup>b</sup>Chemistry Department, Faculty of Science, Suez Canal University, Ismailia, Egypt

<sup>c</sup>Microwave Physics & Dielectrics Dep, Physics Research Institute, National Research Centre, 33 El-Bohouth St., Dokki, Giza 12622, Egypt

<sup>d</sup>Chemistry Department, Faculty of Science, Taif University, 21944 Taif, P.O.Box 11099, Saudi Arabia

*In Loving Memory of Late Professor Doctor "Mohamed Refaat Hussein Mahran"*

### Abstract

Eco-friendly and economically visible processing routes of nanomaterial and nanocomposite is essentially required for the industrial scaling of nanotechnology. The present work aims to prepare ferroelectric polymer nanocomposites with high permittivity to be useful as high dielectric constant (high- $k$ ) materials due to their potential applications in the electronic and electrical industries. Therefore, the semiconducting molybdenum disulfide (MoS<sub>2</sub>) exfoliated by the one-pot hydrothermal method was introduced at different concentrations into a ferroelectric polyvinylidene fluoride (PVDF), forming MoS<sub>2</sub>/PVDF nanocomposite films which were annealed at 160° C for 5 h. At room temperature, the electrical properties of these films were investigated over a wide range of frequencies using the broadband dielectric spectrometer (BDS). In addition, the thermal properties of MoS<sub>2</sub>/PVDF nanocomposite were evaluated. It was found that the samples annealing at 160 °C did not affect the electrical properties but showed electrical stability of all the nanocomposite films. The interfacial polarization and DC conductivity highly affected the samples electrical properties. The composites with high loading of MoS<sub>2</sub> nanosheets (>1wt. %) remarkably enhanced both of permittivity and electrical conductivity.

**Keywords:** Exfoliated MoS<sub>2</sub> nanosheets; PVDF; thermal properties; Permittivity; Conductivity.

### 1. Introduction

The materials of high-dielectric constant (high- $\kappa$ ) materials have attracted much attention due to their potential applications in the electrical and electronic industries. Polymers are often used as dielectric materials because of their ease of processing and flexibility. Besides, most polymers have low permittivity (low- $\kappa$ ), which means that their ability to store electrical energy is limited. To address this issue, two strategies have been suggested. The first involves adding high- $\kappa$  dielectric fillers, such as ferroelectric ceramics, to a polymer matrix to increase its permittivity while maintaining other desirable properties [1, 2]. The second strategy involves incorporating conductive fillers, such as carbon and graphene nanomaterial, into the polymer matrices to create a percolating system. [3, 4]. Recently, the advances in understanding the optical, thermal, and electrical properties of layered nanomaterials have allowed the incorporation of them

in many energy conversions, optical and electrical sensing, catalysis, and energy storage applications. MoS<sub>2</sub>, a transition metal dichalcogenide (TMD), has garnered significant attention due to its unique electrical properties [5-7]. Besides, MoS<sub>2</sub> nanosheets have weak van der Waals forces between the layer structures and thus they can be exfoliated into a few-layer or even single-layer. Generally, a physical method is used for the exfoliation process [8, 9]. MoS<sub>2</sub> nanosheets have magnetized a lot of attention because of their unique semiconducting feature [10], ratable band gap [11, 12], and tunable dielectric constant [10]. Layered nanomaterials have promising features that enable them to tune the electrical properties of polymer-based composites. Recent advances in our understanding of the electrical and thermal properties of these materials have made it possible to incorporate them into different applications, including energy conversion, optical and electrical sensing, and catalysis.

\*Corresponding author e-mail: [rmsoliman66@yahoo.com](mailto:rmsoliman66@yahoo.com); (Ragab Mahani).

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Nowadays, most of the layered nanomaterial procedures of the MoS<sub>2</sub>/polymer nanocomposite are based on the exfoliation method [13-16]. However, the exfoliation procedure of the MoS<sub>2</sub> is suffering from re-aggregation [17, 18] and is a halt in front of industrial mass use in energy devices [19]. Worthy mentioning that restacking of layered nanomaterial during processing may significantly alter the desired properties for particular applications. The processing of layered materials is sensitive to working conditions and hard to scale up for industrial production. For economical and practical reasons, it is necessary to search for easy and scalable methods for the production of ready exfoliation layered nanomaterial. Hydrothermal synthesis (HS) is an eco-friendly and closed system in which solvent can be recycled. However, the most important technological advantage of HS is that atoms assemble in solution to form nanostructure, which stabilize and arrest at any phase [20-22]. With proper processing techniques, the produced nanostructure can be preserved like exfoliated nanosheets [20].

The electrical conductivity of the layered nanomaterial of MoS<sub>2</sub> was enhanced by adding conductive polymers. As a conductive and ferroelectric polymer, polyvinylidene fluoride (PVDF) has been widely used due to its unique electrical and mechanical properties [23, 24]. So, doping PVDF with the MoS<sub>2</sub> nanosheets synthesized by the hydrothermal method results in forming a ferroelectric polymer composite (MoS<sub>2</sub>/PVDF). This composite has great advantages like low cost, eco-friendly, wide band gap energy, variable dielectric properties, high-energy storage density, and stable mechanical properties [25-27]. The electrical energy storage capacity of composites was found to increase with low MoS<sub>2</sub> content while high loading enhanced the permittivity [26]. The repeatability and the simplicity of the fabrication of large-scale MoS<sub>2</sub>/PVDF nanocomposite using the hydrothermal method makes it more applicable in many applications such as anticorrosion, electrical energy storage, and supercapacitor devices. However, a detailed documentation of the role of MoS<sub>2</sub> nanostructures in the electrical properties of polymer nanocomposites has been lacking. Therefore, in the present article, the ready-to-use exfoliated MoS<sub>2</sub> nanosheets were synthesized by a one-pot hydrothermal method based on wet chemical synthesis, which was then introduced into the PVDF matrix at different concentrations, i.e. from 0.1% up to 20%. A high-resolution transmission electron microscopy (HRTEM) was utilized to describe the MoS<sub>2</sub> nanosheets. Differential scanning calorimetry (DSC), derivative thermogravimetry (DTG) and, thermal gravimetric analysis (TGA) were performed to measure the thermal properties of the MoS<sub>2</sub>/PVDF nanocomposites. At room temperature, the broadband dielectric spectrometer (BDS), and the AC

conductivity were used to demonstrate the dielectric properties of the present MoS<sub>2</sub>/PVDF nanocomposites over a wide range of frequencies.

## 2. Experimental Setup Chemical and Reagents

Ammonium molybdate tetrahydrate with  $\geq 99\%$  purity ((NH<sub>4</sub>)<sub>6</sub>Mo<sub>7</sub>O<sub>24</sub>·4H<sub>2</sub>O) and thiourea were purchased from Sigma-Aldrich. For the experimental work, a Milli-Q ultrapure water was used. The aqueous solutions were prepared by deionized water.

### Synthesis of MoS<sub>2</sub>/PVDF nanocomposite films

Exfoliated nanosheets of MoS<sub>2</sub> were synthesized by hydrothermal processing using a homemade autoclaved setup. 0.004 moles of ammonium molybdate tetrahydrate and 0.06 moles thiourea were dissolved in 140 mL ultrapure water. The resulting solution was transferred to a Teflon-coated stainless steel autoclave and kept at 200°C with stirring speed of 350 RPM for 24 h. After that, the autoclave was allowed to cool down and the final product was filtered and washed with ethanol multiple times. To prepare plain PVDF and different wt. % ratios (x: 0.1, 0.3, 0.5, 1, 3, 5, 10, and 20%) of MoS<sub>2</sub>/PVDF nanocomposite films. Further information about the preparation of the MoS<sub>2</sub>/PVDF nanocomposite films was given elsewhere [27].

### Instrumentation

The High-resolution impedance analyzer spectrometer (Schlumberger Solartron 1260) was used to measure the dielectric properties. The permittivity ( $\epsilon'$ ), the dielectric loss ( $\epsilon''$ ), and the AC conductivity ( $\sigma_{ac}$ ) were concerned. These properties are related to each other and can be calculated from the output parameters, i.e. capacitance  $C(\omega)$ , and loss tangent ( $\tan \delta$ ) as follow:

$$\epsilon'(\omega) = \frac{d}{A} \epsilon_o C \quad (1)$$

$$\epsilon''(\omega) = \epsilon' \tan \delta \quad (2)$$

$$\sigma_{ac}(\omega) = \epsilon_o \omega \epsilon'' \quad (3)$$

The thickness of the sample (in meters) was represented by 'd', while the surface area (in square meters) was represented by 'A'. The permittivity of free space was denoted by  $\epsilon_o$  ( $8.854 \times 10^{-12} \text{ Fm}^{-1}$ ) and the angular frequency was represented by ( $\omega$ ), which is equal to  $2\pi f$  ( $f$  is the applied field frequency). The dielectric properties of MoS<sub>2</sub>/PVDF nanocomposite films were measured at room temperature before and after annealing at 160°C for 5 hours. The measurements were conducted over a wide range of frequencies ( $10^{-1}$ - $10^6$  Hz).

The differential scanning calorimetry (DSC), derivative thermogravimetry (DTG), and the thermal gravimetric analysis (TGA) were used to measure the thermal properties of the MoS<sub>2</sub>/PVDF nanocomposite films at different concentrations of MoS<sub>2</sub>, using Shimadzu Simultaneous DTG-60H thermal analyzer, Japan. All thermal measurements were conducted under a nitrogen atmosphere at a flow rate of 30

mL/min. The measurements ranged from 20 to 1000°C with a heating rate of 10°C/min.

### 3. Results and Discussions

#### Morphology of the MoS<sub>2</sub>/PVDF nanocomposite

The morphology of the synthesized nanocomposites was further characterized using HRTEM to elucidate the microstructural properties, morphology, and dispersion of exfoliated nanosheets of MoS<sub>2</sub> in the PVDF. The morphology of the as-prepared films shows MoS<sub>2</sub> nanosheets assemblies that are poorly stacked few-layer MoS<sub>2</sub> with interlamellar spacing. Fig. 1 displays exfoliated nanosheets of 3 wt% MoS<sub>2</sub> doped in PVDF prepared by Hydrothermal processing. The surface contains combinations of the MoS<sub>2</sub> nanosheets distributed and retained homogeneously in the composites, Figure 1(A). The

exfoliated nanosheets of MoS<sub>2</sub> originated from the superstructures during the hydrothermal process of the MoS<sub>2</sub>/PVDF nanocomposites. The density of the exfoliated nanosheets depends on the weight percent of the doped MoS<sub>2</sub> in PVDF and it increases as the concentration of MoS<sub>2</sub> increases. The electron diffraction pattern of MoS<sub>2</sub>/PVDF film shows diffused rings caused by short-range ordering of MoS<sub>2</sub>, with few diffraction spots indicating the formation of an ordered structure in two dimensions (2D), mainly the hexagonal structure of the MoS<sub>2</sub>. The electron diffraction pattern of MoS<sub>2</sub> supports the hybrid crystal structure of MoS<sub>2</sub>/PVDF nanocomposites. The dispersion of the exfoliated nanosheets of MoS<sub>2</sub> through the PVDF matrix has an important role in the thermal and dielectric properties of their composites.

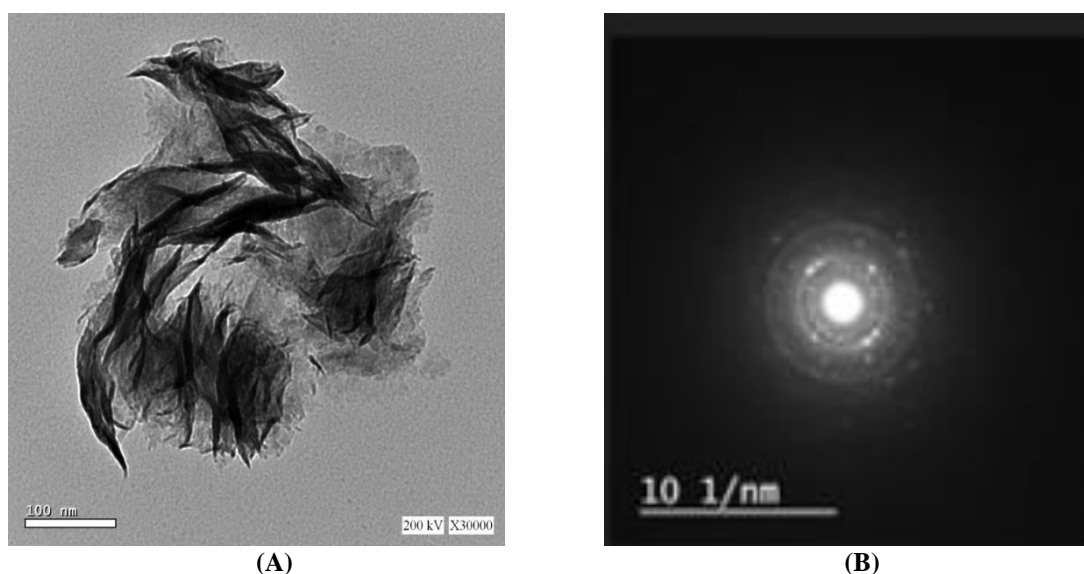


Figure 1. HRSEM image of MoS<sub>2</sub>/PVDF nanocomposite film (A) and the corresponding electron diffraction pattern (B).

#### Thermal Analysis of the MoS<sub>2</sub>/PVDF nanocomposites

The thermal decomposition of the MoS<sub>2</sub> nanocomposite doped in the PVDF nanocomposites was studied at different loading ratios of MoS<sub>2</sub> nanoparticles (5, 10, and 20 wt%). Figure 2 shows the derivative mass loss (DTG) and the differential scanning calorimetry (DSC) curves at a temperature range from room temperature up to 700°C with a heating rate of 10°C/min under a nitrogen atmosphere. The pure PVDF undergoes thermal decomposition in a single step process, and its maximum decomposition temperature is around 472.5°C. The addition of MoS<sub>2</sub> resulted in a gradual increase in the thermal stability of PVDF. For example, when the MoS<sub>2</sub> content in the film was 10 wt%, the maximum decomposition temperature was found to be 362°C, which is higher than that of pure PVDF (350°C), leading to an enhancement in the

thermal stability of PVDF. This increase in thermal stability is attributed to the hindered spread of volatile decomposing products within the nanocomposites. The amalgamation of MoS<sub>2</sub> nanoparticles leads to the PVDF polymer's huge molecular chains becoming more anarchism and tangledly arranged, which hinders the movement of the polymer pieces at the interfaces between the PVDF and MoS<sub>2</sub> surface [28-30]. This, in turn, increases the thermal stability of PVDF.

#### The dielectric study

The dielectric properties of MoS<sub>2</sub>/PVDF nanocomposite films with different concentrations of MoS<sub>2</sub> (0, 0.1, 0.3, 0.5, 1, 3, 5, 10, and 20 wt. %) before and after annealing at 160°C for 5 hours, were investigated as a function of frequency at room temperature, as shown in Figure 3. The study examined the permittivity ( $\epsilon'$ ), dielectric loss ( $\epsilon''$ ), and

AC conductivity ( $\sigma_{ac}$ ). The study focused on the electrical properties of the treated and untreated composite films loaded with 0.3 and 3 wt % MoS<sub>2</sub> (Figure 3, right). In general,  $\epsilon'$  for all nanocomposite films were much higher values in the low-frequency region ( $\sim 10^4$ ) due to the contribution of all polarization components (space charge, dipolar, ionic, and electronic) [31]. At higher frequencies,  $\epsilon'$  decreased and became almost constant, showing much lower values ( $\sim 10$ ). Such a decrease in  $\epsilon'$  at high frequencies is attributed to that some types of polarization components became less effective, i.e. total polarization decreases.

Similar behavior is noticed for  $\epsilon''$  with the exception that it increases at frequencies higher than  $10^5$  Hz. At low frequencies,  $\epsilon''$  shows high values which lead to imperfect charge transport, similar to DC conductivity ( $\sigma_{dc}$ ) [32].  $\sigma_{dc}$  mainly comes from the long-range movements and alignments of free-charge carriers in response to the electric field, resulting in a net flow of charge through the materials.  $\sigma_{dc}$  effect

appears as plateau-like behavior, particularly for the composites loaded with 1 wt. % MoS<sub>2</sub> or higher. Above 1 wt. % of MoS<sub>2</sub>, the effect shifts to higher frequencies, indicating that a percolating of the MoS<sub>2</sub> network throughout the composites has been enhanced. Basically, the electrical conductivity of MoS<sub>2</sub>/PVDF nanocomposites can be enhanced upon loading with MoS<sub>2</sub> nanosheets. Above a critical frequency called hopping frequency ( $\omega_p$ ), the conductivity rapidly increases as the frequency increases, obeying the universal power law  $A\omega^s$  [32, 33].

For being MoS<sub>2</sub>/PVDF nanocomposites heterogeneous (have different phases), the accumulation of the free charge carriers (electrons, holes, ions) at these interfaces may also dominate the dielectric properties. This phenomenon is well known as interfacial polarization or Maxwell-Wagner-Sillars (MWS) polarization. As a result, the high values seen in  $\epsilon''$  could be due to the superposition of  $\sigma_{ac}$  and MWS effects.

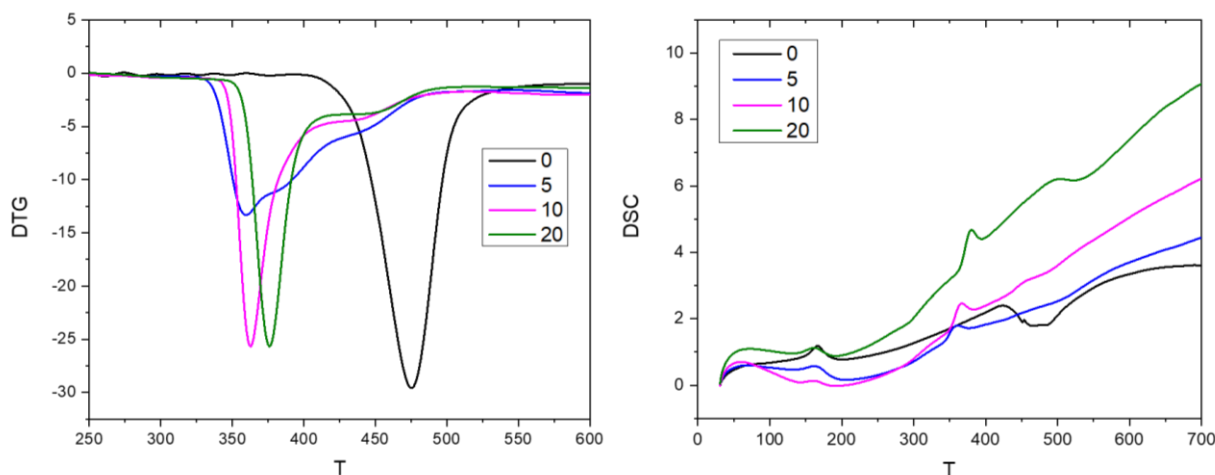


Figure 2. DTG-DSC thermograms of PVDF at different loading ratio of MoS<sub>2</sub> nanoparticles (5.0, 10.0, and 20.0 wt%).

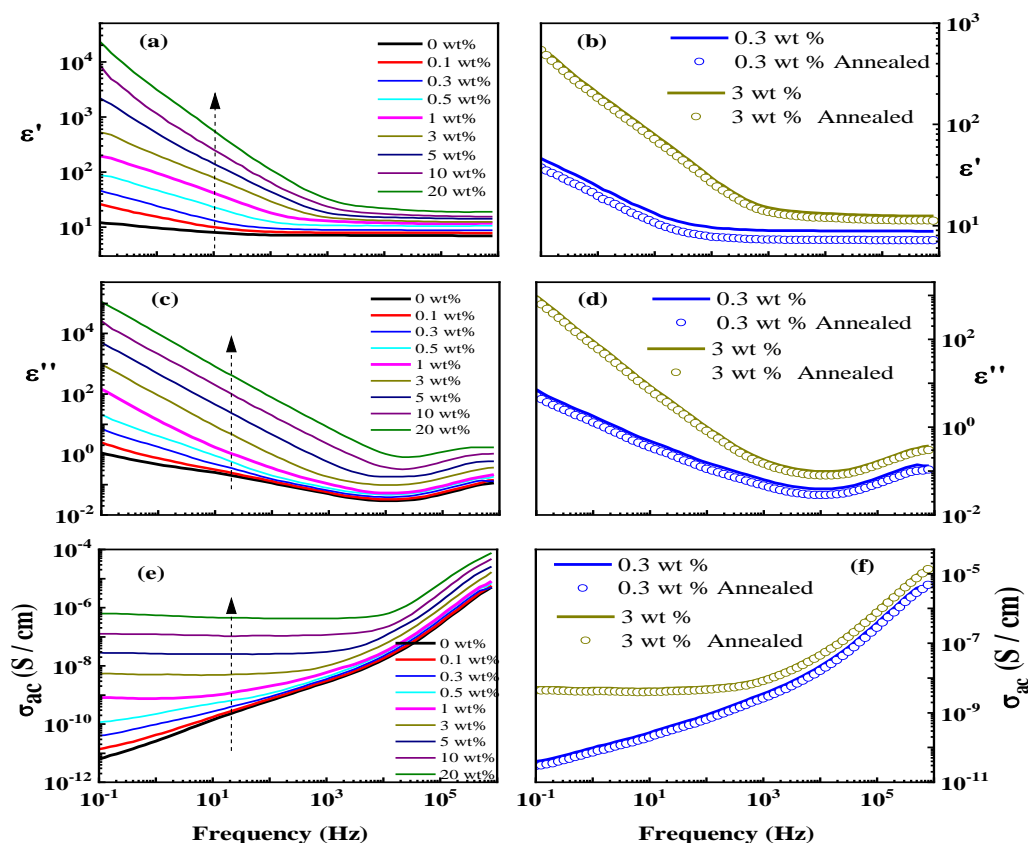


Figure 3. The frequency dependence of permittivity ( $\epsilon'$ ), dielectric loss ( $\epsilon''$ ), and AC conductivity ( $\sigma_{ac}$ ) for all  $\text{MoS}_2/\text{PVDF}$  nano-composite films before annealing (a, c, and e) and after annealing for only 0.3 wt.%  $\text{MoS}_2/\text{PVDF}$  and 3 wt.%  $\text{MoS}_2/\text{PVDF}$  at  $160^\circ\text{C}$  for 5 h (b, d and f)

The impact of  $\text{MoS}_2$  on the permittivity of  $\text{PVDF}/\text{MoS}_2$  nanocomposites was studied in detail. Figure 4 illustrates the relationship between  $\text{MoS}_2$  loading and  $\epsilon'$  at 1 Hz. In the figure,  $\epsilon'$  may exhibit percolation-like electrical characteristics. It increases slowly with increasing  $\text{MoS}_2$  loading until it reaches a critical concentration (1 wt.%), after which it significantly increases at higher concentrations. This may be correlated to the decrease in the optical band gap, regardless of whether the samples were annealed

or not. Moreover, upon loading with 20 wt. %  $\text{MoS}_2$ ,  $\epsilon'$  showed much higher values ( $\sim 3125$ ), which is six orders of magnitude higher than that of the pure  $\text{PVDF}$ . Additionally, the values are ten times higher than those recently reported for the same composites [26]. These findings suggest that  $\text{MoS}_2$  nanosheets can significantly enhance the permittivity and the electrical conductivity of  $\text{PVDF}/\text{MoS}_2$  nanocomposites, making them useful as High-dielectric constant (high- $\kappa$ ) materials.

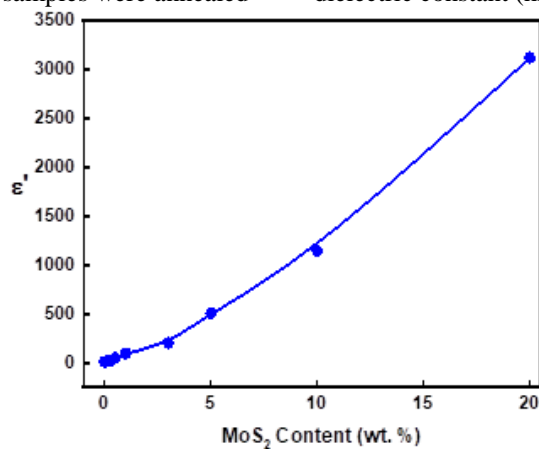


Figure 4.  $\text{MoS}_2$  content dependence of the permittivity ( $\epsilon'$ ), and AC conductivity ( $\sigma_{ac}$ ) at 1Hz for  $\text{PVDF}/\text{MoS}_2$  nano-composite films before annealing.

#### 4. Conclusion

A simple casting method using the hydrothermal approach was used to prepare different concentrations (0.1, 0.3, 0.5, 1, 3, 5, 10, and 20%) of MoS<sub>2</sub> nanosheets. These nanosheets were then doped into a PVDF polymer matrix, forming MoS<sub>2</sub>/PVDF nanocomposite films. MoS<sub>2</sub> significantly enhanced the electrical properties of these films. When the concentration of MoS<sub>2</sub> nanosheets was particularly high (20 wt %), the permittivity of MoS<sub>2</sub>/PVDF composite film reached values much higher (~3125 at 1 Hz) than that of pure PVDF (~10 at 1 Hz). This research has the potential to pave the way for constructing MoS<sub>2</sub>-based composites for high-dielectric constant materials and electrical energy storage applications.

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#### Ethics approval

Not applicable.

#### \* Consent to participate

The authors agree to the journal's policy

#### \* Consent for publication

The authors agree to the journal's policy

#### \* Availability of data and materials

Data will be available from the corresponding author upon reasonable request.

#### Competing interests

The authors declare that they have no competing interests.

#### Authors' contributions

Abdallah A. Shaltout suggested, wrote and reviewed the research article.

Nasser Y. Mostafa prepared the samples and shared in writing the manuscript

Ragab Mahani wrote and analyzed the dielectric data and shared in in writing the manuscript

Mohamed M. Ibrahim wrote the thermal analysis data and shared in writing the main manuscript.

#### 5. References

- [1] J. Hu, S. Zhang, B. Tang, 2D filler-reinforced polymer nanocomposite dielectrics for high-k dielectric and energy storage applications, *Energy Storage Materials*, 34 (2021) 260-281.
- [2] B. Xie, T. Wang, J. Cai, Q. Zheng, Z. Liu, K. Guo, P. Mao, H. Zhang, S. Jiang, High energy density of ferroelectric polymer nanocomposites

utilizing PZT@SiO<sub>2</sub> nanocubes with morphotropic phase boundary, *Chemical Engineering Journal*, 434 (2022) 134659.

- [3] X. Zhang, P. Samorì, Graphene/Polymer Nanocomposites for Supercapacitors, *ChemNanoMat*, 3 (2017) 362-372.
- [4] J. Gopal, M. Muthu, I. Sivanesan, A Comprehensive Compilation of Graphene/Fullerene Polymer Nanocomposites for Electrochemical Energy Storage, *Polymers*, 15 (2023) 701.
- [5] X. Li, L. Wu, L. Hao, Y. Fu, Chapter 5 - Emerging 2D Nanomaterials for Supercapacitor Applications, in: K.Y. Cheong, G. Impellizzeri, M.A. Fraga (Eds.) *Emerging Materials for Energy Conversion and Storage*, Elsevier, 2018, pp. 155-183.
- [6] J. Luxa, O. Jankovský, D. Sedmidubský, R. Medlín, M. Maryško, M. Pumera, Z. Sofer, Origin of exotic ferromagnetic behavior in exfoliated layered transition metal dichalcogenides MoS<sub>2</sub> and WS<sub>2</sub>, *Nanoscale*, 8 (2016) 1960-1967.
- [7] P. Tummala, A. Lamperti, M. Alia, E. Kozma, L.G. Nobili, A. Molle, Application-Oriented Growth of a Molybdenum Disulfide (MoS<sub>2</sub>) Single Layer by Means of Parametrically Optimized Chemical Vapor Deposition, *Materials Today: Proceedings*, 13 (2020) 2786.
- [8] Y. Arao, J.D. Tanks, M. Kubouchi, A. Ito, A. Hosoi, H. Kawada, Direct exfoliation of layered materials in low-boiling point solvents using weak acid salts, *Carbon*, 142 (2019) 261-268.
- [9] D. Sun, D. Huang, H. Wang, G.-L. Xu, X. Zhang, R. Zhang, Y. Tang, D. Abd Ei-Hady, W. Alshitari, A. Saad Al-Bogami, K. Amine, M. Shao, 1T MoS<sub>2</sub> nanosheets with extraordinary sodium storage properties via thermal-driven ion intercalation assisted exfoliation of bulky MoS<sub>2</sub>, *Nano Energy*, 61 (2019) 361-369.
- [10] E.J. Santos, E. Kaxiras, Electrically driven tuning of the dielectric constant in MoS<sub>2</sub> layers, *ACS nano*, 7 (2013) 10741-10746.
- [11] Y. Yan, B. Xia, X. Ge, Z. Liu, J.-Y. Wang, X. Wang, Ultrathin MoS<sub>2</sub> Nanoplates with Rich Active Sites as Highly Efficient Catalyst for Hydrogen Evolution, *ACS Applied Materials & Interfaces*, 5 (2013) 12794-12798.
- [12] W. Wu, L. Wang, Y. Li, F. Zhang, L. Lin, S. Niu, D. Chenet, X. Zhang, Y. Hao, T.F. Heinz, J. Hone, Z.L. Wang, Piezoelectricity of single-atomic-layer MoS<sub>2</sub> for energy conversion and piezotronics, *Nature*, 514 (2014) 470-474.
- [13] E.J. Aiub, D. Steinberg, E.A. Thoroh de Souza, L.A.M. Saito, 200-fs mode-locked Erbium-doped fiber laser by using mechanically exfoliated MoS<sub>2</sub> saturable absorber onto D-shaped optical fiber, *Opt Express*, 25 (2017) 10546-10552.

- [14] Y. Hou, Y. Hu, S. Qiu, L. Liu, W. Xing, W. Hu, Bi<sub>2</sub>Se<sub>3</sub> decorated recyclable liquid-exfoliated MoS<sub>2</sub> nanosheets: Towards suppress smoke emission and improve mechanical properties of epoxy resin, *Journal of Hazardous Materials*, 364 (2019) 720-732.
- [15] C. Jung, H.I. Yang, W. Choi, Effect of Ultraviolet-Ozone Treatment on MoS<sub>2</sub> Monolayers: Comparison of Chemical-Vapor-Deposited Polycrystalline Thin Films and Mechanically Exfoliated Single Crystal Flakes, *Nanoscale Research Letters*, 14 (2019) 278.
- [16] W. Li, Y. Zhang, X. Long, J. Cao, X. Xin, X. Guan, J. Peng, X. Zheng, Gas Sensors Based on Mechanically Exfoliated MoS<sub>2</sub> Nanosheets for Room-Temperature NO<sub>2</sub> Detection, *Sensors (Basel)*, 19 (2019) 2123.
- [17] T.-W. Lee, C.-C. Chen, C. Chen, Chemical Stability and Transformation of Molybdenum Disulfide Nanosheets in Environmental Media, *Environmental Science & Technology*, 53 (2019) 6282-6291.
- [18] X. Yang, Z. Chen, J. Fang, Q. Yang, W. Zhao, X. Qian, C. Liu, D. Zhou, S. Tao, X. Liu, Efficient exfoliation to MoS<sub>2</sub> nanosheets by salt-assisted refluxing and ultrasonication with photocatalytic application, *Materials Letters*, 255 (2019) 126596.
- [19] C. Zhao, R. Wang, Y. Zhang, L. Chen, T. Li, X. Deng, P. Zhang, X. Lu, Electrostatic force-driven anchoring of Ni(OH)<sub>2</sub> nanocrystallites on single-layer MoS<sub>2</sub> for high-performance asymmetric hybrid supercapacitors, *Electrochimica Acta*, 320 (2019) 134591.
- [20] N.Y. Mostafa, A.A. Shaltout, H. Omar, S.A. Abo-El-Enein, Hydrothermal synthesis and characterization of aluminium and sulfate substituted 1.1nm tobermorites, *Journal of Alloys and Compounds*, 467 (2009) 332-337.
- [21] N.Y. Mostafa, M.M. Hessien, A.A. Shaltout, Hydrothermal synthesis and characterizations of Ti substituted Mn-ferrites, *Journal of Alloys and Compounds*, 529 (2012) 29-33.
- [22] G.A.M. Mersal, N.Y. Mostafa, A.-E.H. Omar, Hydrothermal synthesis and processing of hydrogen titanate nanotubes for nicotine electrochemical sensing, *Materials Research Express*, 4 (2017) 085031.
- [23] E.M. Abdelrazek, R. Holze, Structural, optical and some physical properties of PVDF films filled with LiBr/MnCl<sub>2</sub> mixed fillers, *Physica B: Condensed Matter*, 406 (2011) 766-770.
- [24] M. Aatur Rahman, G.-S. Chung, Synthesis of PVDF-graphene nanocomposites and their properties, *Journal of Alloys and Compounds*, 581 (2013) 724-730.
- [25] K. Cai, X. Han, Y. Zhao, R. Zong, F. Zeng, D. Guo, A Green Route to a Low Cost Anisotropic MoS<sub>2</sub>/Poly(Vinylidene Fluoride) Nanocomposite with Ultrahigh Electroactive Phase and Improved Electrical and Mechanical Properties, *ACS Sustainable Chemistry & Engineering*, 6 (2018) 5043-5052.
- [26] Q. Jia, X. Huang, G. Wang, J. Diao, P. Jiang, MoS<sub>2</sub> Nanosheet Superstructures Based Polymer Composites for High-Dielectric and Electrical Energy Storage Applications, *The Journal of Physical Chemistry C*, 120 (2016) 10206-10214.
- [27] A.A. Shaltout, N.Y. Mostafa, R.M. Mahani, S.I. Ahmed, M.A. Allam, E. Alzahrani, Z.K. Heiba, H.H. Wahba, Investigation of structural and optical properties of molybdenum disulfide flakes/polyvinylidene fluoride nanocomposites, *Journal of Materials Research and Technology*, 9 (2020) 14350-14359.
- [28] X.-K. Chen, Y.-J. Zeng, K.-Q. Chen, Thermal Transport in Two-Dimensional Heterostructures, *Frontiers in Materials*, 7 (2020).
- [29] R. Abinaya, J. Archana, S. Harish, M. Navaneethan, C. Muthamizhchelvan, S. Ponnusamy, H. Udono, R. Sugahara, Y. Hayakawa, M. Shimomura, Interface driven energy-filtering and phonon scattering of polyaniline incorporated ultrathin layered molybdenum disulphide nanosheets for promising thermoelectric performance, *Journal of Colloid and Interface Science*, 584 (2021) 295-309.
- [30] M.R. Gharib-Zahedi, A. Koochaki, M. Alaghemandi, Tuning the polymer thermal conductivity through structural modification induced by MoS<sub>2</sub> bilayers, *Soft Matter*, 18 (2022) 6927-6933.
- [31] F. Kremer, A. Schönhal, *Broadband Dielectric Spectroscopy*, 1 ed., Springer-Verlag Berlin Heidelberg, 2003.
- [32] A.K. Jonscher, *Dielectric Relaxation in Solids*, Chelsea Dielectrics Press, 1983.
- [33] A.K. Jonscher, The 'universal' dielectric response, *Nature*, 267 (1977) 673-679.