# MATERIAL CHARACTERIZATION OF POLYURETHANE DIELECTRIC ELASTOMER

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# MATERIAL CHARACTERIZATION OF POLYURETHANE DIELECTRIC ELASTOMER

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(Mechanical Engineering)

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# ABSTRACT

Polyurethane (PU) is a promising dielectric elastomer (DE), however increasing its dielectric constant is essential for PU to become a viable DE. Incorporating graphene oxide (GO) as a filler has the potential but may increase dielectric loss and conductivity. Zirconium silicate ($ZrSiO_4$) is an alternative, but excessive use potentially decreasing the actuation strain. This study delves into the impact of polydopamine (PDA) modification on GO and $ZrSiO_4$, and their influence on the dielectric and mechanical properties of PU composites. Nanoparticles were modified with PDA, then mixed with PU matrix at various weight percentage through melt-mixing process. The modified nanoparticles (GO-PDA and $ZrSiO_4$-PDA) significantly enhances the dielectric constant of PU composites compared to unmodified nanoparticles. PU with GO-PDA shows reduction in dielectric loss and conductivity with 70% and 50% decrement compared to unmodified GO due to insulative barriers in GO-PDA. However, $ZrSiO_4$ modification results in lattice contraction, leading to an increase in dielectric loss (up to 100%) and conductivity (up to 354%) for composites with $ZrSiO_4$-PDA. A hybrid composite incorporating 2.5 wt.% of GO-PDA and 30 wt.% of $ZrSiO_4$ was synthesized, demonstrating elevated dielectric constant up to 25, albeit with elevated losses (up to 0.48) and conductivity (up to $5 \times 10^{-7}$ S/m), which may stem from a reaction between PDA and $ZrSiO_4$, affecting the insulating properties of GO-PDA and causing lattice contraction of $ZrSiO_4$. This study comprehensively explores the effects of PDA modification on GO and $ZrSiO_4$ in PU-based elastomers, shedding light on electrical, mechanical properties, and morphology. It informs potential strategies for developing DE for generators or actuators.

# ABSTRAK

Poliuretana (PU) adalah sejenis elastomer dielektrik (DE) yang berpontensi, namun peningkatan pemalar dielektriknya adalah penting bagi PU untuk menjadi DE yang boleh diaplikasikan. Penyertaan grafin oksida (GO) sebagai pengisi mempunyai potensi tetapi boleh meningkatkan kehilangan dielektrik dan kekonduksian. Zirkonium Silikat ($ZrSiO_4$) merupakan alternatif, tetapi penggunaan berlebihan boleh mengurangkan regangan aktuasi. Kajian ini mengkaji kesan modifikasi (PDA) terhadap GO dan $ZrSiO_4$, serta kesannya terhadap sifat dielektrik dan mekanikal komposit PU. Nanopartikel dimodifikasi dengan PDA, kemudian dicampur dengan matriks PU pada peratus berat yang berbeza melalui proses pencampuran lebur. Nanopartikel yang diubahsuai (GO-PDA dan $ZrSiO_4$-PDA) secara signifikan meningkatkan pemalar dielektrik komposit PU berbanding nanopartikel yang tanpa modifikasi. PU dengan GO-PDA menunjukkan penurunan kehilangan dielektrik dan kekonduksian sebanyak 70% dan 50% berbanding GO yang tanpa modifikasi disebabkan penghalang insulatif dalam GO-PDA. Walau bagaimanapun, pengubahsuaian $ZrSiO_4$ mengakibatkan penyusutan kekisi, menyebabkan peningkatan kehilangan dieletrik (hingga 100%) dan kekonduksian (hingga 354%) bagi komposit dengan $ZrSiO_4$-PDA. Satu komposit hibrid yang mengandungi 2.5 wt.% GO-PDA dan 30 wt.% $ZrSiO_4$ telah disintesis, dengan pemalar dielektrik yang tinggi sehingga 25, namun disertai kehilangan yang tinggi (hingga 0.48) dan kekonduksian (hingga 5 X 10-7 S/m), yang mungkin disebabkan oleh tindak balas antara PDA dan $ZrSiO_4$ yang mempengaruhi sifat insulatif GO-PDA dan menyebabkan penyusutan kesisi $ZrSiO_4$. Kajian ini secara menyeluruh meneroka kesan pengubahsuaian PDA terhadap GO dan $ZrSiO_4$ dalam elastomer PU, memberikan penerangan tentang sifat-sifat elektrik, mekanikal, dan

morfologi. Ia memberikan pandangan terhadap strategi yang berpotensi ke arah membangunkan DE sebagai generator atau aktuator.

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The Examination Committee has met on **11th September 2023** to conduct the final examination of **Muhammad Naiem Naqiuddin bin Zaharin** on his degree thesis entitled **‘Material Characterization of Polyurethane Dielectric Elastomer”.**

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# LIST OF ABBREVIATIONS

| | |
|---|---|
| DE | Dielectric elastomer |
| GO | Graphene oxide |
| $ZrSiO_4$ | Zirconium silicate |
| PU | Polyurethane |
| PDA | Polydopamine |
| DEG | Dielectric elastomer generator |
| DEA | Dielectric elastomer actuator |
| WEC | Wave-energy converter |
| OWC | Oscillating water column |
| Ag | Silver |
| PPMTC | Poly(propylene-monothiocarbonate) |
| Si | Silicone |
| SWCNT | Single-walled carbon nanotube |
| MWCNT | Multi-walled carbon nanotube |
| SR | Silicone Rubber |
| $BaTiO_3$ | Barium titanate |
| $SrTiO_3$ | Strontium titanate |
| CCTO | Copper titanate |
| PZT | Zirconate titanate |
| HDPE | High-density polyethylene |
| TiC | Titanium carbide |

| | |
|---|---|
| $SiO_2$ | Silicon dioxide |
| FESEM | Field emission scanning electron microscopy |
| EDX | Energy dispersive X-ray |
| FTIR | Fourier-transform infrared spectroscopy |
| UTM | Universal Testing Machine |
| Wt% | Weight percentage |
| Pa | Pascal |

# LIST OF SYMBOLS

| | |
|---|---|
| $\varepsilon_O$ | Vacuum permittivity |
| $\varepsilon_r$ | Relative permittivity |
| $U$ | Internal storage |
| $C$ | Capacitance |
| $V$ | Voltage |
| $A$ | Surface area of elastomer |
| $Z$ | Thickness of elastomer |
| $W_E$ | Harvested energy |
| $V_b$ | Bias voltage |
| $\sigma$ | Maxwell pressure |
| $s_z$ | Actuation strain |
| $\beta$ | Electromechanical sensitivity |
| $Y$ | Elastic modulus |
| $s$ | AC conductivity |
| $f$ | Frequency |

# CHAPTER 1

# INTRODUCTION

## 1.0 Introduction

Dielectric elastomer (DE) has been known since the early days of electricity when James Maxwell identified the impact of dielectric materials known as Maxwell stress in his work on the foundation of electromagnetic theory [1]. DE first came to the attention of academics in the 1990s, when various research articles and findings for prospective uses were published. This might be due to DEs' unique ability to transform electrical energy into mechanical energy and vice versa, making them adaptable enough to be used in a variety of sectors. There are various suitable polymers that can act as a dielectric elastomer such as acrylic [2]–[5], silicone rubber [6]–[8], poly (vinylidene fluoride) (PVDF) [9], [10] natural rubber [11], and polyurethane.

Polyurethane has emerged as a potential material to be used as a dielectric elastomer due to its high dielectric constant and huge force outputs, allowing them to be actuated at lower electric fields. However, improvement is still required to make polyurethane a viable dielectric elastomer. Increasing the dielectric permittivity of polyurethane elastomer leads to a rise in capacitance, which aids in the development of better DE. A common method is to create elastomer composites by introducing filler into the system. Conductive filler such as metal or carbon filler and high dielectric filler such as ceramic is proven to be able to increase the polymer dielectric permittivity but at a cost.

In this study, graphene oxide (GO) and zirconium silicate ($ZrSiO_4$) were incorporated into the PU matrix to study the effect of those fillers on the dielectric and mechanical properties of the elastomer. These nanofillers were also modified with polydopamine (PDA) and incorporated with PU matrix to study the effect of the modification towards the elastomer dielectric and mechanical attributes. Last but not least, a hybrid composite of GO and $ZrSiO_4$ was made based on the characterization result of the single-filler composite. Dielectric characterization was done through dielectric analyzer and mechanical characterization was done using tensile test. Other characterization such as morphology, spectral and elemental were also been carried out using FESEM, FTIR and EDX analysis, respectively.

## 1.1 Problem Statement

Polyurethane (PU) possesses several favorable properties, including a high dielectric permittivity, low dielectric loss, and impressive stretchability. However, its suitability as a DE is limited by certain drawbacks, such as a high elastic modulus and low breakdown strength. The high elastic modulus hampers electromechanical sensitivity, thereby impacting the DE's actuation capabilities. Additionally, the low breakdown strength restricts the maximum voltage that can be applied to the elastomer before it conducts electricity. One potential solution to address these challenges is to enhance the elastomer's dielectric permittivity. By increasing the dielectric permittivity, the elastomer's actuation functionality can be achieved with minimal voltage, effectively overcoming the limitation of low breakdown strength. Furthermore, elevating the dielectric permittivity of the material augments its capacitive properties, allowing it to store more energy and perform exceptionally well as a generator.

Graphene oxide (GO) is an excellent filler for this endeavor. It has ultra-high electrical conductivity and a large specific surface area thanks to the unique two-dimensional (2-D) structure of GO, which aids in boosting the elastomer's interfacial polarization and dielectric permittivity [12]–[14]. However, the addition of conductive fillers such as graphene increases the dielectric loss and conductivity of the polymer, resulting in decreased breakdown strength and system efficiency [15]–[17].

Due to its high dielectric constant, zirconium silicate ($ZrSiO_4$) is also a viable option filler for DE composite [18]. Aside from that, its insulative qualities demonstrated low leakage current densities and maximum capacitance densities, which aids in the development of DE with high dielectric permittivity and low loss [19]. However, massive loadings of ceramic filler have been shown to significantly increase the dielectric permittivity of the polymer. High filler loadings improve the elastic modulus of the elastomer, resulting in a stiff and low actuation strain elastomer [20]–[22].

### 1.2 Objective

The purpose of this study is to achieve several objectives as follows:

1. To characterize the effect of the PDA-modification towards GO and $ZrSiO_4$ nanoparticle’s morphology, elemental and composition.