

**UNIVERSITI TEKNOLOGI MARA**

**DIELECTRIC AND ELECTRICAL  
PROPERTIES OF BILAYER  
ZnO/PVDF-TrFE FILMS FOR LOW  
FREQUENCY CAPACITOR  
APPLICATION**

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**PhD**

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## AUTHOR'S DECLARATION

I declare that the work in this thesis was carried out in accordance with the regulation of Universiti Teknologi MARA. It is original and is the result of my own work, unless otherwise indicated or acknowledged as reference work. This thesis has not been submitted to any other academic institution or non-academic institution for any degree or qualification.

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## ABSTRACT

Recent studies focused on the development of polymer hybrid bilayer and multilayer films configuration as an improvisation to overcome agglomeration in nanocomposite system. However, the deposition of multilayer film usually requires complex processing parameters that requires longer time to produced. Hence, deposition of polymeric bilayer film was used as an alternative to overcome this limitation. The deposition of polymeric materials on metal-oxide layers were observed to improve the final properties of these hybrid configurations system. Polymeric material of Polyvinylidene Fluoride Trifluoro-Ethylene (PVDF-TrFE) copolymers were utilized due to its high dielectric properties. PVDF-TrFE dissolved in Methyl Ethyl Ketone (MEK) spin coated on aluminium-glass (Al-glass) substrates yield high  $\beta$ -crystal as observed in ATR-FTIR and XRD analysis. The spin coated single layer PVDF-TrFE thin films were annealed at temperatures of 100°C, 120°C and 140°C. PVDF-TrFE thin film annealed at 120°C (AN120) displayed a significant improvement in its crystallinity, dielectric, and electrical properties. This is evident by the 95% increment of the count per second (cps) value (XRD) with high dielectric constant ( $\epsilon'$ ) value of 13.8 with low tangent loss ( $\tan \delta$ ) of 0.03 at frequency of 1 kHz. AN120 also produced high resistivity ( $\rho$ ) value of  $1.71 \times 10^5 \Omega \cdot \text{cm}$  with low leakage current. Metal-oxide layer consist of ZnO with particle size of 130nm, revealed polydispersity index (PdI) value of 0.34 with Z-average of 207 d.nm. This indicates a uniform size distribution of 130nm particles in the ZnO 130nm film, thus favourable as the nano fillers to be utilized in this study. The spin coated unannealed and annealed (300°C, 400°C and 500°C) ZnO 130nm films illustrates a peak at  $2\theta = 31.9^\circ$ ,  $34.4^\circ$ , and  $36.2^\circ$  suggesting the presence of wurtzite hexagonal crystal structure of ZnO. Nonetheless, unannealed ZnO 130nm film showed highest cps value for all diffraction angles, which proposed that the film has high crystallinity, hence suitable to be utilized as film template. Upon deposition of PVDF-TrFE on unannealed metal-oxide ZnO film template, the bilayer ZnO/PVDF-TrFE film annealed at 100°C (bilayer ZnO/PVDF-TrFE (100°C)), showed 5% increment in the  $\epsilon'$  value with low  $\tan \delta$  value (0.3) measured at the same frequency. The  $\rho$  value of bilayer ZnO/PVDF-TrFE (100°C) showed a promising onefold increased ( $1.33 \times 10^6 \Omega \cdot \text{cm}$ ) compared to the single layer PVDF-TrFE film. Most importantly, these improvements in the bilayer film were gained at temperature 20°C less for annealing than that utilized in a single layer PVDF-TrFE film. Hence, this demonstrates that the optimized dielectric film attained in this study was spin coated PVDF-TrFE film deposited on spin coated ZnO film layer, and then annealed as a bilayer system at 100°C. The 1 kHz frequency utilized in this study suggests that the high resistivity bilayer ZnO/PVDF-TrFE (100°C) film obtained in this study can be utilized for application in low frequency capacitor with frequency range of 100 – 1 kHz.

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