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Influence of Inorganic Fillers on the Mechanical Properties of Polypyrrole Composite Thin Films

Alin-Alexandru Andrei[1,2], Izabell Craciunescu[1], Lucian Barbu Tudoran[1], George Marian Ispas[1], Gavril Ionel Giurgi[1], Alexandru Oprea[1], Mioara Zagrai[1], Cristian Sevcencu[1,2]

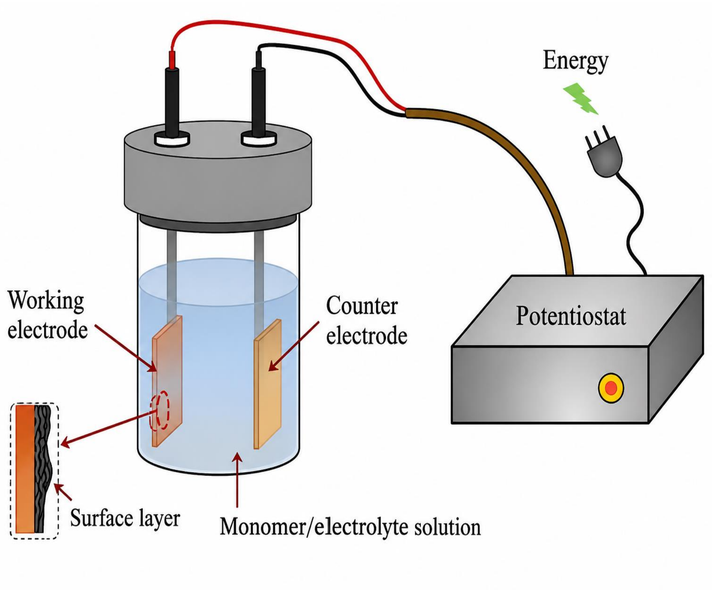
[1]National Institute for Research and Development of Isotopic and Molecular Technologies Cluj-Napoca, Romania
[2]Doctoral School in Integrative Biology, Faculty of Biology and Geology, Babes-Bolyai University, 400084 Cluj-Napoca, Romania

INTRODUCTION & AIM

Conductive polymers such as polypyrrole (PPy) have attracted significant interest for flexible electronic applications due to their electrical conductivity, chemical stability, and ease of synthesis. However, pure PPy films often exhibit limited mechanical robustness, which can restrict their long-term performance in flexible devices. Incorporating inorganic fillers into the polymer matrix represents a promising strategy to improve the mechanical stability of conductive polymer films while maintaining their functional properties.

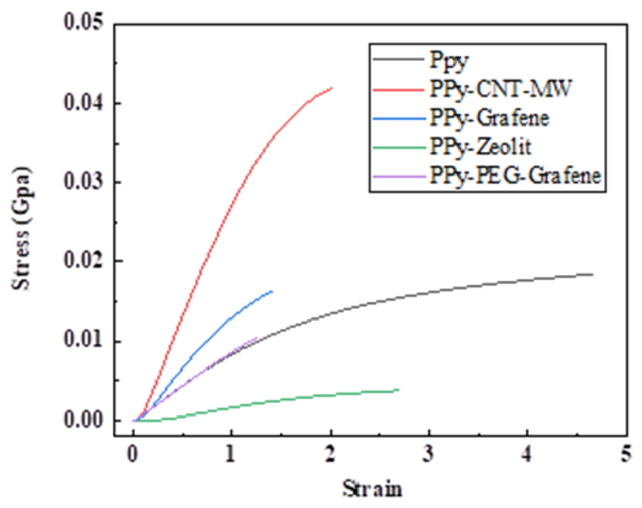
METHOD

PPy-TSA and composite films were prepared by galvanostatic electropolymerization in acetonitrile (ACN) containing pyrrole (0.1 M) and p-toluenesulfonic acid (p-TSA, 0.1 M) as dopant. Graphene (GR) and carbon nanotubes (CNT) were incorporated at 0.4 wt%, while zeolite (ZE) was added at 4 wt%. A hybrid formulation containing polyethylene glycol (PEG) and graphene (PPy-TSA-PEG-GR) was also prepared. All additives were dispersed in the electrolyte prior to electropolymerization, and all films were synthesized under identical experimental conditions.

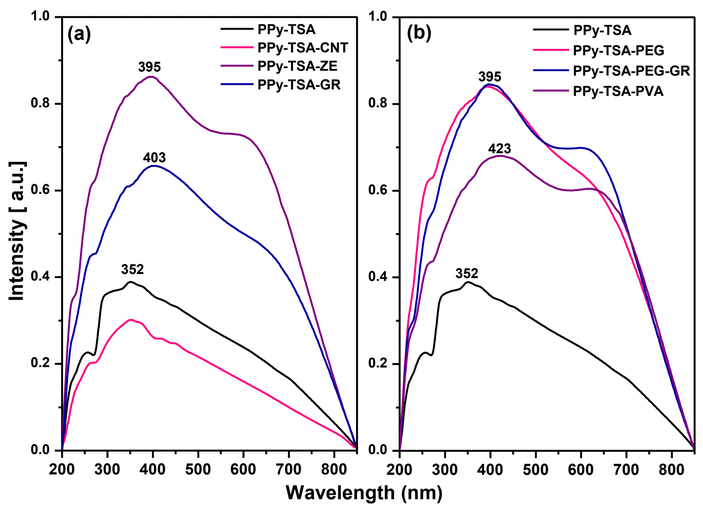


RESULTS & DISCUSSION

Mechanical properties. The incorporation of inorganic fillers significantly influenced the mechanical behavior of PPy-TSA films. The reference PPy-TSA film exhibited a Young's modulus of approximately 0.008 GPa, indicating a compliant material. CNT incorporation increased the modulus to ~0.026 GPa, demonstrating a strong reinforcing effect, while GR produced a moderate increase (~0.016 GPa). In contrast, PPy-TSA-ZE exhibited the lowest modulus (~0.003 GPa), suggesting a highly compliant structure, likely due to the porous nature of zeolite and reduced stress-transfer efficiency within the composite matrix.



Electrical properties. The PPy-TSA reference film showed the highest electrical conductivity (208 ± 16 S·cm⁻¹). The addition of GR and CNT reduced conductivity to 62 ± 3 and 58 ± 8 S·cm⁻¹, respectively, indicating that the filler concentration was insufficient to establish an effective conductive network. The most pronounced decrease was observed for PPy-TSA-ZE (4.5 ± 0.6 S·cm⁻¹), attributed to the insulating character of zeolite and the increased film thickness. Despite these reductions, all composites retained semiconducting behavior and stable conductivity over the investigated temperature range (25–85 °C).



Optical properties. UV–Vis analysis revealed that CNT incorporation produced only minor changes in the electronic structure of PPy-TSA. In contrast, GR- and ZE-containing films exhibited a redshift of the π–π* transition from ~352 nm to ~397 nm, indicating increased effective conjugation length and modifications of the PPy electronic structure. Optical band-gap values ranged from 2.01 eV (PPy-TSA-ZE) and 2.12 eV (PPy-TSA-GR) to 2.33 eV (PPy-TSA-CNT), compared with 2.25 eV for PPy-TSA, confirming that filler incorporation modifies the electronic structure without disrupting the semiconducting nature of the polymer matrix.

CONCLUSIONS

Overall, graphene provided the most balanced combination of mechanical compliance and electrical performance, whereas CNT mainly improved stiffness and zeolite induced significant structural changes, including increased film thickness and reduced conductivity.

Among the investigated systems, PPy-TSA-PEG-GR exhibited the most favorable combination of flexibility, conductivity, and signal stability, making it the most promising candidate for flexible, gel-free bioelectronic electrodes.

ACKNOWLEDGMENT

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