Chemical and thermomechanical characterization of green, and ecofriendly PAFS-free battery separator films

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In 2024, global energy demand increased by 2.2%, driven by record-high temperatures and digitalization - exceeding the average annual growth rate observed over the past decade [1]. Li-ion batteries play a pivotal role in the transition towards renewable energy systems by allowing efficient short-term energy storage. Recently, polymethylmethacrylate (PMMA) coatings have been employed as an alternative to polyvinylidene fluoride (PVDF) based binders in porous polyolefin battery separators, enabling the development of eco-friendly, PFAS-free Li-ion batteries [2]. The chemical composition and the thermomechanical properties of a PMMA-PE-PMMA battery separator film were characterized using Fourier transform infrared microscopy (FTIR), dynamic mechanical analysis (DMA) and uniaxial tensile testing. FTIR microscopic images were recorded in attenuated total reflection (ATR) mode using a Spectrum 100 and a Spotlight 400 imaging system (Perkin Elmer, USA) equipped with a Ge-crystal. The thermomechanical properties were deduced by amplitude sweep tensile-mode DMA (-40 to 130°C, 3 K/min, 1 Hz, 4 N), using a TMA/SDTA 861 E (Mettler Toledo, USA). Additionally, tensile tests were performed on a Z005 (Zwick Roell, Germany) with the digital image correlation (DIC) system VIC-3D (Correlated Solutions, USA). FTIR microscopy revealed a thickness of about 10 µm for the PE core layer and 5 to 40 µm for the PMMA coating layer (see **Fig. 1a)**). Furthermore, absorption peaks related to PE and PMMA were confirmed by FTIR for the core and coating layers, respectively. Interestingly, absorption bands related to PMMA were also discernible within the porous PE core layer. The DMA thermogram, shown in **Fig.1b)**, revealed a decrease of the storage modulus (E') from 1800 at -25°C to 230 MPa at 100°C. Furthermore, the loss factor (tan(δ)) exhibited a small peak at -24°C associated with the glass transition temperature, and a broad peak ranging from 25 to 90°C presumably related to mobility in hinge segments (amorphous phase between two crystalline phases). The tensile modulus (E_t), maximum strength (σ_{max}) and strain at break (ε_b) values were ascertained at 1040 MPa, 70 MPa and 45%, respectively (see **Fig.1c)**). Overall, tensile (E_t) and storage (E') modulus values were well in agreement with a deviation of about 8%.

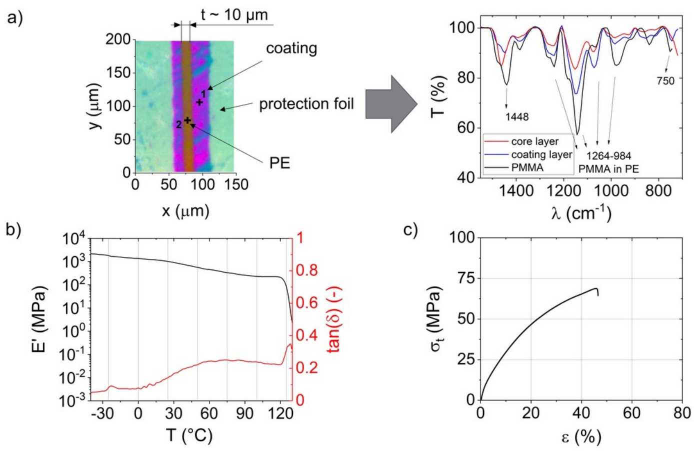

Fig. 1: Chemical and thermomechanical characterization of a PMMA-PE-PMMA based battery separator film: a) FTIR microscopy image and spectras, b) amplitude sweep tensile-mode DMA thermogram, and c) stress-strain curve obtained from tensile testing.

References

[1] Martinos, A., Spencer, T., Tapia, V. G., Roge, A. Global Energy Review 2025. IEA **2025**, Online available: https://www.iea.org/reports/global-energy-review-2025.

[2] Choi, J., Kim, P. J. A roadmap of battery separator development: Past and future, Current Opinion in Electrochemistry **2022**, 31, 100858.