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Water dynamics in conductive PEDOT:PSS/cellulose nanocomposite films

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PEDOT:PSS is a water-dispersable and electrically conductive polymer blend that is increasingly applied in numerous fields such as batteries and super-capacitors. While many studies focus on performance optimization, degradation issues because of humid environments are rarely discussed: PEDOT:PSS absorbs significant amounts of water (~50 wt%), which leads to a pronounced swelling factor of up to 1.6.
The integration of PEDOT:PSS into a cellulose nanofibril (CNF) matrix enhances significantly the mechanical integrity and limits water absorption. Moreover, a complex nanocomposite morphology is generated, which changes in dependence on the ambient humidity: high humidity leads to de-wetting of PEDOT:PSS from CNF bundles. As a result, the conductivity decreases. Upon drying, this behavior is reversible, however only after a first drying/humidifying cycle, which we refer to an initial kinetically trapped film morphology.
By studying the water dynamics via QENS, we identified two water species inside the films: fast-moving bulk water and slow-moving hydration water. In dry conditions, bulk water is completely released from the films, while parts of the hydration water remain inside the films. The remaining hydration water fraction provides a certain mobility for the PEDOT:PSS chains and supports their wetting on the CNF bundles. In addition, the QENS measurements provide detailed information about the diffusive behavior and the hydrogen-bonding environment of water molecules.

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