Abstract

High-Performance Humidity Sensing in pi-conjugated molecular assemblies through the Engineering of Electron/Proton Transport and Device Interfaces

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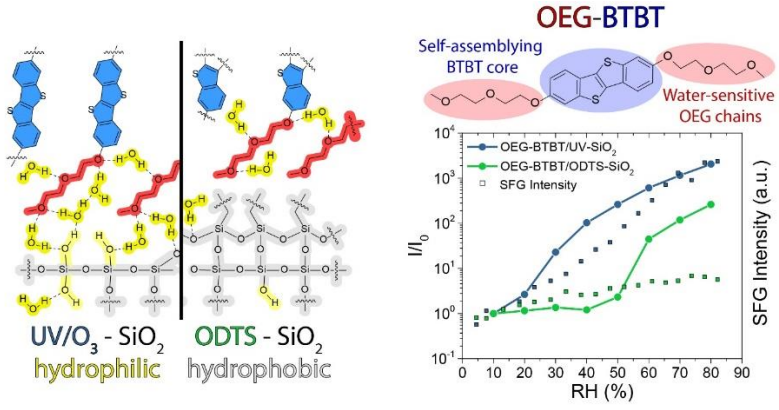
Achieving precise control over the chemistry of our environment is key for well-being and for guaranteeing high standards for industrial and technological processes. Environmental humidity is one of the ambient parameters that can severely affect the shelf life of goods, the efficiency of large-scale industrial systems and it thus needs to be finely controlled e.g., in surgery rooms in hospitals. Humidity can also unpredictably influence the outcome of laboratory experiments as it modifies the reactivity and various properties of materials at the nanoscale. Hence, the subtle control of the ambient humidity is pivotal for modern and sustainable technologies in diverse fields. The development of systems capable of responding to environmental changes, such as humidity, requires the design and assembly of highly sensitive elements that can operate as efficient transducers. Such a challenge can be mastered only by disentangling the role played by each component in the responsive device, thus ultimately achieving high performance by optimizing the synergistic contribution of all functional elements. We designed and synthesized a novel [1]benzothieno[3,2-b][1]benzothiophene (BTBT) derivative equipped with hydrophilic oligoethylene glycol lateral chains (OEG-BTBT). Such a molecular system electrically transduces subtle changes of RH with high current ratios (>10^4) at low voltages (2 V). A multi-scale structural, spectroscopical, and electrical characterization was performed to elucidate the role of each device constituent including the BTBT core and OEG side chains, the material's interfaces and the dielectric substrate. While the BTBT molecular core promotes the self-assembly into (semi)conducting crystalline films, the OEG side chains are prone to adsorb ambient moisture. These chains act as hotspots for hydrogen bonding with atmospheric water molecules that locally dissociate when a bias voltage is applied, resulting in a mixed electronic/protonic (H^+) long-range conduction throughout the film. Due to the OEG-BTBT molecules orientation with respect to the surface and structural defects within the film, water molecules can access the humidity-sensitive sites of the SiO_2 substrate surface, which can be further chemically modified to tune its hydrophilicity for an improved device response. Our findings provide insightful information on the synergistic chemical engineering of materials and interfaces for designing highly sensitive humidity-responsive electrical devices whose mechanism relies on the interplay of electron and proton transport.



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