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Influence of anionic and cationic polyelectrolytes on the conductivity and morphology of poly(3,4-ethylenedioxythiophene): poly(styrenesulfonate) films

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ABSTRACT

Conductivity of the poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS) strongly depends on the film morphology, which can be altered by the presence of polyelectrolytes. Aqueous dispersion of PEDOT:PSS was studied with anionic sodium polyacrylate (PA) and cationic poly(dimethyldiallylammonium chloride) (pDADMAC) polyelectrolytes that are typically used in papermaking as retention aids and dispersing agents in the paper pigment coating formulations. Spin-coated PEDOT:PSS films on a PA coated glass formed non-uniform layers with lowered conductivity compared to the reference PEDOT:PSS films on a clean glass substrate. On contrary, spin-coated PEDOT:PSS on a pDADMAC coated glass formed uniform layers with good conductivity. These results point out the importance of surface chemistry when using renewable and recyclable paper-based substrates with the PEDOT:PSS films.

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1. Introduction

Fabrication of electronic devices directly onto paper substrate has received increased attention in recent years. Besides printing conductive non-patterned or patterned areas on paper [1,2], an increasing number of devices and concepts have been demonstrated such as an all-organic active matrix display [3], a passive ultra-high radio frequency identification tag module embedded in a paper-on-paper structure [4], moisture and temperature sensors [5,6], an ethanol sensor [7], a piezoresistive microelectromechanical force sensor [8], a H_2S sensor [9], a low-cost platform for electrochemical analysis [10], an electrochemical detection device for paper-based microfluidistics [11,12], and a photovoltaic device [13,14].

The paper based electronics can be divided into two main categories utilizing either bulk or surface conductivity. In bulk conductivity, the entire paper is functionalized to become an electrically conductive integral device component. This can be achieved, for example, by immersing or coating an uncoated paper with conductive polymers, or by in-situ polymerization in the paper [15–18]. Bulk conductivity can also be achieved directly from wood pulp fibers that are coated by a multi-sequenced, layer-by-layer method with conductive polymers or particles [19–22]. Alternatively, adding silver plated carbon fibers or activated carbon fiber to the pulp or preparing conductive paper out of kraft pulp mixed with metalized polyester fibers provides alternative routes for conductive paper production [23,24].

Our work concentrates on the surface conductive paper electronics, which are typically fabricated on coated paper grades [25–27]. Pigment coating improves the paper performance with an increased compatibility with the end-use application. Improved printability and optical characteristics with well controlled barrier properties to avoid functional material penetration into paper can be achieved together with enhanced surface strength, opacity, surface smoothness, and controllable porosity [28,29]. Performance of printed functionality such as conductive tracks can significantly be improved with the use of optimized coating layers [30,31].

In this study we investigate the effect of polyelectrolytes in direct contact with conductive layers. Polyelectrolytes are widely used as retention agents in the papermaking process: addition of polyelectrolytes in the wet end of the paper machine improves retention of mineral pigment fillers embedded into the fiber matrix of the pulp as well as retention of starch, gum, or rosin sizing during the formation of the base paper [32]. Polyelectrolytes are also used as dispersing agents in coating formulations [33]. In the coating layer, pigment surfaces are almost fully covered by polyelectrolytes, which may interfere with printed conductive layers. Therefore, understanding the impact of polyelectrolytes on the conductive layers is crucial for optimizing the device performance when fabricated on paper substrates.

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2. Experimental

A homogenized, water-based conductive polymer poly(3,4-ethylenedioxythiophene):poly(styrenesulfonate) (PEDOT:PSS, Clevios PH 500, Heraeus Holding GmbH, DE) was studied in the presence of two polyelectrolytes that consisted of an anionic sodium polyacrylate (PA, molecular weight < 10 kDa, Polysalz S, Basf, DE) and a cationic poly(dimethyldiallylammonium chloride) (pDADMAC, molecular weight > 80 kDa). The PEDOT:PSS ratio of the Clevios PH500 was 1:2.5 (by weight) and a pH range of 1.0–2.5 that points the protons in the sulfonic acid groups of the PSS. Polystyrene itself is non-polar while sulfonic acid is a strong acid i.e. it dissociates effectively in water. The negatively charged sulfonate ions attract positive PEDOT-molecules. Therefore, the sulfonate sites in the PEDOT:PSS water dispersion particles are attracted by the positively charged pDADMAC molecules.

All studied materials were deposited and measured on a pristine glass substrate to eliminate additional variables and complexity induced by the rough and porous coated paper substrate i.e. the wide variability of coating pigment particle mixtures with varying absorption, porosity, and surface roughness characteristics. The polyelectrolytes were spin-coated (KW-4A, Chemat Technology Inc., US) onto hand cut 2.5 × 2.5 cm^2 glass slides for 60 s at 1050 rpm for PA and 1300 rpm for pDADMAC with the following concentrations: PA at 2.5 wt.%, 5 wt.%, and 10 wt.%, and pDADMAC at 1 wt.%, 2.5 wt.%, 5 wt.%, and 10 wt.%. These rather high concentration amounts were chosen as the glass samples could not be uniformly coated below a certain polyelectrolyte specific concentration that was required to create reproducible and uniform films. Spin-coating was selected instead of inkjet printing for deposition of thin films to avoid the complexity caused by the additional ink components i.e. lubricants and humectants such as ethylene glycol, propylene glycol, and glycerol needed to make the PEDOT:PSS polymer and polyelectrolytes inkjet printable. Therefore, spin-coating was used to coat pure polyelectrolyte and PEDOT:PSS films to study their interaction without any additional factors.

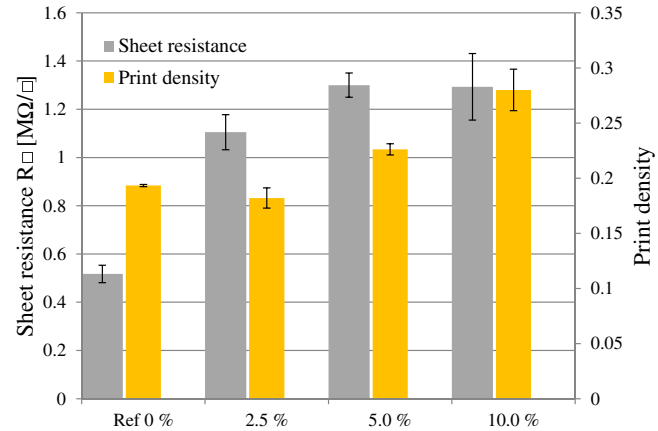

Fig. 2. Sheet resistance (a) and print density (b) values of spin-coated PEDOT:PSS on PA coated glass.

Homogenized standard PEDOT:PSS was spin-coated on top of the polyelectrolyte films and annealed at 130 °C for 15 min according to the manufacturer specifications. Sheet resistance $R_{sq} = R \times (W/L)$ is a measure (Ω/□) over a dimensionless unit (□), where R is the measured resistance over a given electrode gap distance L with an electrode width W, which allows comparison between measurements having different areas. However, in this study the sheet resistance of PEDOT:PSS was measured across two parallel conductive hand painted silver stripes with a constant electrode gap distance of 10 mm with a digital multimeter (Keigthley 2100, US) at ambient conditions (24 °C, relative

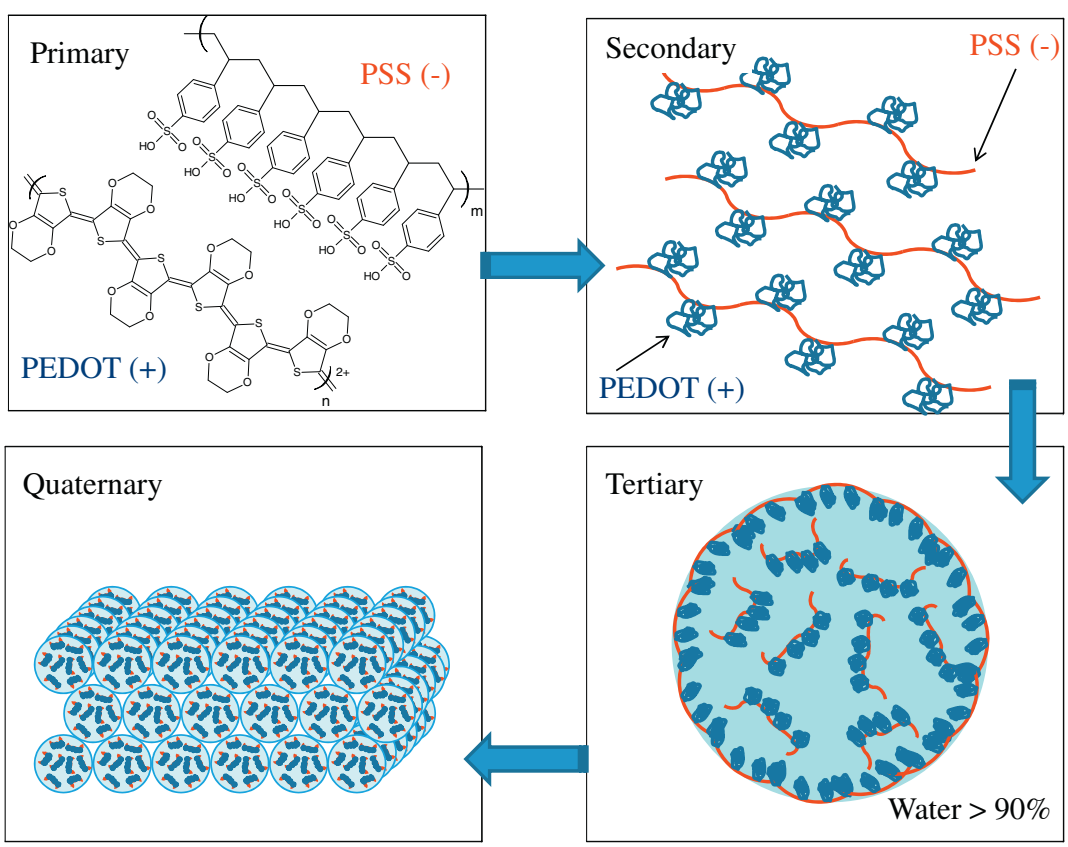

Fig. 1. Hierarchical structure of PEDOT:PSS.

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