

## Enhancing Volumetric Capacitance in pgBTBT Polymers through Ethylene Glycol Side Chain Variation and Blending Approaches

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The development of efficient electrochemical devices necessitates a comprehensive understanding of how polymer structure influences their performance. In this study, we investigated the impact of ethylene glycol side chains on the volumetric capacitance of pgBTBT, a regiochemistry-driven organic electrochemical transistor<sup>1</sup>. Inspired by prior research on polymer design and mixed conductors<sup>2</sup>, we synthesized a series of pgBTBT polymers with varying percentages of hydrophilic, ethylene glycol side chains, ranging from 50% to 100%. Our findings revealed a positive correlation between the percentage of ethylene glycol side chains and the volumetric capacitance of the polymers, except at 90% side chain content.

Building upon these findings, we further explored the potential of blending as a method for enhancing the volumetric capacitance of the polymers<sup>3</sup>. Two distinct blending approaches were employed: pgBTBT with pBTBT (OR)<sub>2</sub>, and pgBTBT with pgBTBT-OEG-OR. The volumetric capacitance of the blend with pBTBT (OR)<sub>2</sub> was found to increase in accordance with the trend established by the varying side chain percentages. However, the blend with pgBTBT-OEG-OR demonstrated a consistent volumetric capacitance across the same ratios<sup>4</sup>, with both blends exhibiting lower volumetric capacitance than copolymers, except at a 90% ethylene glycol side chain ratio.

Additionally, we examined the effect of film thickness on the kinetics of doping and dedoping<sup>5</sup>. We discovered a direct relationship, with increased thickness leading to longer doping and dedoping times. This observation holds potential for future investigations into the interplay between side chain modifications and doping kinetics.

Our study provides valuable insights into the manipulation of polymer structure for improved electrochemical performance, serving as a foundation for the design of high-performing organic electrochemical transistors and mixed conductors.

1. Rawad K. Hallani, Bryan D. Paulsen, Anthony J. Petty, II, Rajendar Sheelamantula, Maximilian Moser, Karl J. Thorley, Wonil Sohn, Reem B. Rashid, Achilleas Savva, Stefania Moro, Joseph P. Parker, Oscar Drury, Maryam Alsufyani, Marios Neophytou, Jan Kosco, Sahika Inal, Giovanni Costantini, Jonathan Rivnay, Iain McCulloch, *Journal of the American Chemical Society*, **2021**, 143, 11007-11018.
2. Achilleas Savva, Rawad Hallani, Camila Cendra, Jokubas Surgailis, Tania C. Hidalgo, Shofarul Wustoni, Rajendar Sheelamantula, Xingxing Chen, Mindaugas Kirkus, Alexander Giovannitti, Alberto Salleo, Iain McCulloch, Sahika Inal, *Advanced Functional Materials*, **2020**, 30, 1907657.
3. Micah Barker, Tommaso Nicolini, Yasmina Al Yaman, Damien Thuau, Olga Siscan, Sasikumar Ramachandran, Eric Cloutet, Cyril Brochon, Lee J Richter, Olivier J Dautel, *Materials Horizons*, **2023**.
4. David Ohayon, Victor Druet, Sahika Inal, *Chemical Society Reviews*, **2023**.
5. Priscila Cavassin, Isabelle Holzer, Demetra Tsokkou, Olivier Bardagot, Julien Réhault, Natalie Banerji, *Advanced materials*, **2023**, 2300308.