Machine Learning-Enabled Prediction of Electronic Properties of Radical-Containing Polymers at Coarse-Grained Resolutions

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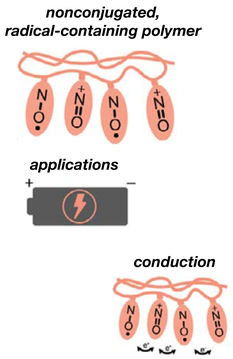
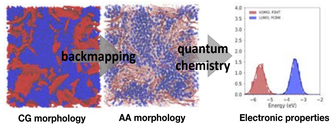
Introduction

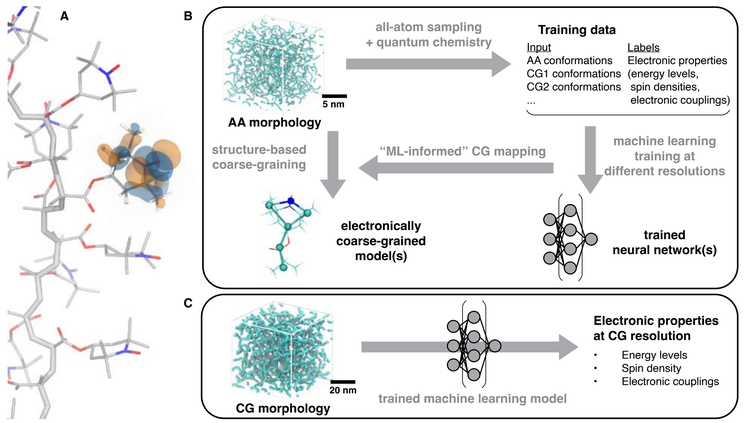


- Radical(-containing) polymers possess intriguing redox, optoelectronic, and magnetic characteristics.
- Applications range from energy or memory storage to optoelectronics and spintronics.
- Coupling of electronic and conformational degrees of freedom over a wide range of spatiotemporal scales determines their properties.
- State-of-the-art modeling to establish relationships between molecular structure, morphology and electronic properties:



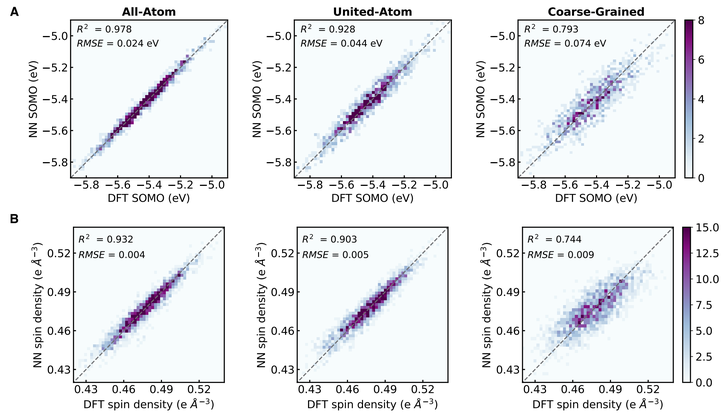
Wang, Easley, Lutkenhaus, *ACS Macro Lett.* **2020**
Tan, Hsu, Tahir, Dou, Savoie, Boudouris, *JACS* **2022**
Alessandri, Sami, Barnoud, de Vries, Marrink, Havenith, *Adv. Funct. Mater.* **2020**

The method



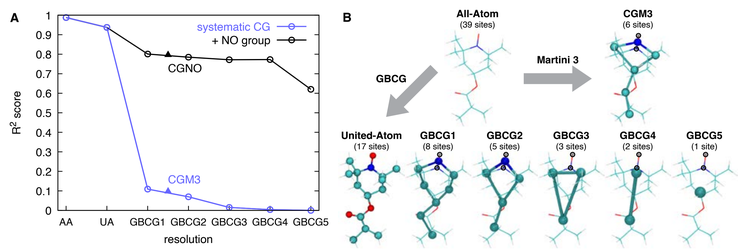
Reith, Putz, Muller-Plath, *J. Comput. Chem.* **2003**
Jackson, Bowen, Antony, Webb, Vishwanath, de Pablo, *Sci. Adv.* **2019**

Monomer conformation-dependent electronic properties



- Prediction of monomer conformation-dependent electronic properties at CG resolution.

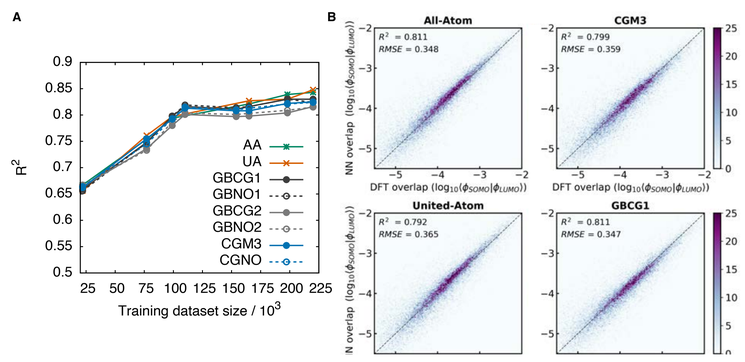
Impact of Coarse-Grained Mapping



- NO• description essential to predict energy levels of nitroxide-based radical polymers.
- Direction: design CG model that preserves the NO• orientation

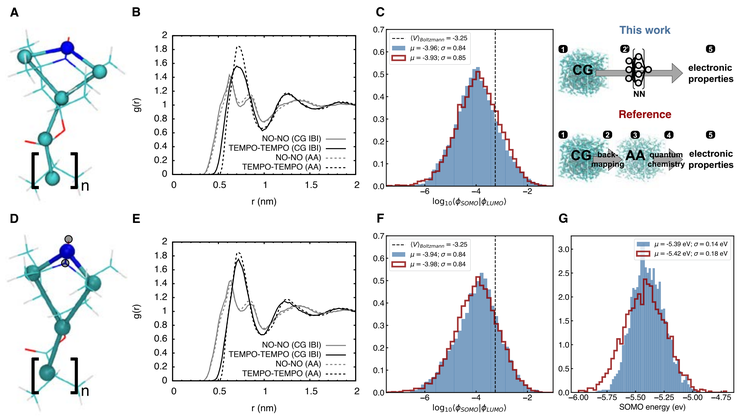
Graph-Based Coarse-Graining: Webb, Delannoy, de Pablo, *JCTC* **2019**
Martini 3: Souza, Alessandri, Barnoud, Thallmair, *et al.*, *Nat. Methods* **2021**

Dimer conformation-dependent electronic properties



- Prediction of electronic couplings is more data-intensive.
- NO• description not necessary.
- Smaller gap between AA/UA and CG resolutions.

Application and validation



- Predictions in agreement with reference backmapping-based approach.
- Structural accuracy of CG model is essential.