Investigations into Message Passing Neural Networks and Polymer Fouling

by

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Investigations are conducted on high-fidelity molecular property prediction with Message Passing Neural Networks (MPNNs) and nanoscale polymer fouling experimentation with Quartz Crystal Microbalances (QCMs).

Message Passing Neural Networks are promising deep learning architectures for chemical property prediction. The state-of-the-art MPNN known as Chemprop is adapted in this thesis for the prediction of infrared spectra. A novel loss function and architectural extensions are identified which increase the predictive performance of spectra relative to the original architecture. These extensions are collected into the software package Chemprop-IR. The Chemprop-IR architecture holds promise in the prediction of experimental infrared spectra with little to no peak shifts within the fingerprint region. Uncertainty estimation frameworks within Chemprop are also extended in this thesis. A pretraining procedure and expanded readout structure are identified which improve the accuracy of predicted properties and estimated aleatoric uncertainties while maintaining scalable predictivity.

Process fouling is a pervasive problem in ethylene plants. Foulant, or the undesired accumulation of material, has been identified throughout process units of ethylene plants. Fouling can result in a multitude of challenges in plant production and operation. A few challenges include reduced thermal separation efficiencies and increased process safety concerns. Identifying the root causes for various forms of process fouling is essential to identifying the appropriate mitigation strategies. In particular, polymer fouling within ethylene plants is probed experimentally by Quartz Crystal Microbalance with Dissipation monitoring (QCM-D). QCM-D is used to assess the potential for ethylene plant products to form polymer foulant or support growth of pre-existing polymer foulant at approximate process conditions. The effects of molecular oxygen and antioxidant inhibitors on foulant growth dynamics are also addressed.

The results of this thesis are twofold. Results regarding MPNNs illustrate increases in the prediction fidelity of deep learning algorithms. Advances in molecular property prediction with MPNNs support the integration of MPNNs into closed-loop frameworks for accelerated molecular discovery and design. Results regarding polymer fouling suggest the efficacy of examined antioxidants on mitigating polymer fouling. The experimental workflow developed around QCM-D could be applied and adapted for continued polymer fouling research.

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