Artificial intelligence and Automation in Chemical Kinetic Modeling

Florence H. Vermeire, Kevin M. Van Geem

Laboratory for Chemical Technology

Ghent University

florence.vermeire@ugent.be

The chemistry of simple molecules can be extremely complex. Thermal decomposition, combustion, and pyrolysis processes often involve hundreds of intermediate species and hundreds of thousands of elementary reactions between those species. In the last decade, computer-aided kinetic modeling software has been developed to deal with those large kinetic models. Examples of such software are Genesys developed at the Laboratory for Chemical Technology at Ghent University, and the Reaction Mechanism Generator (RMG) developed in the Green Group at Massachusetts Institute of Technology. Both have demonstrated success in automatically developing kinetic models for the pyrolysis of oils, high-temperature pyrolysis of natural gas, combustion of biofuels, *etc.* The number of significant species and reactions in gas-phase kinetic models increases exponentially with the number of heavy atoms in the fuels. One of today's challenges for automatic kinetic modeling software, and one of the highlights of this talk, is how to deal with detailed elementary-step kinetic models for molecules with more than ~8 heavy atoms or for surrogate mixtures. Most recently, RMG has been used to develop a kinetic model for the pyrolysis of a Jet-A surrogate, the profiles of the main product species are given in Figure 1, comparing experimental measurements and model simulations. Another challenge deals with the presence of hetero-atoms. The chemistry of hydrocarbon pyrolysis and oxidation are well-known, but for species containing O/N/S/Cl/… moieties, finding the right reactions and determining the thermodynamic and kinetic parameters poses a challenge. Genesys has been extended to deal with many different functional groups, most recently for the combustion of amines and oxymethylene ethers.

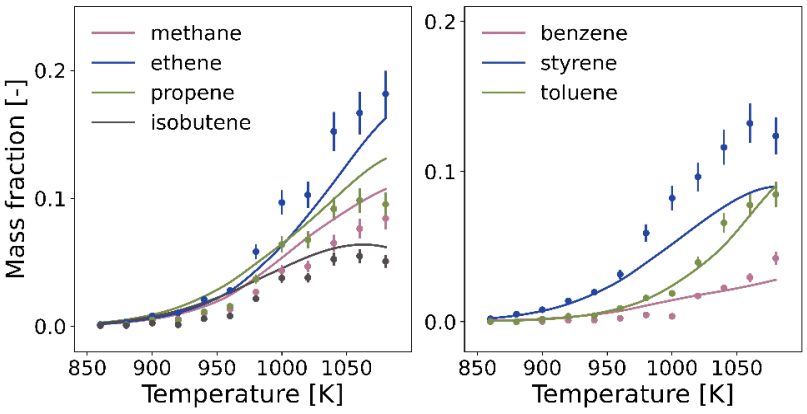

Figure 1. Mass fraction of the main product species from the pyrolysis of a Jet-A surrogate, comparing experimental measurements (symbols) and model simulations (lines).

The kinetic models contain so many thermodynamic and kinetic parameters (*e.g.* k's, K_{eq}'s) that they cannot all be determined experimentally. Instead, most of those parameters are determined automatically using structure-activity relationships. The few thermodynamic and kinetic parameters that are sensitive towards the concentration of certain desired products, are typically refined with high-level theoretical calculations or experimental measurements. Recent advances in artificial intelligence for applications in chemical engineering have opened a new route for the fast and more accurate prediction of such chemical properties. This talk will center around the application of machine learning using message passing neural networks for the fast predictions of thermodynamic and kinetic properties in the framework of computer-aided chemical kinetic model development. The typical machine learning model architecture for message passing neural network used for chemical property prediction is given in Figure 2.

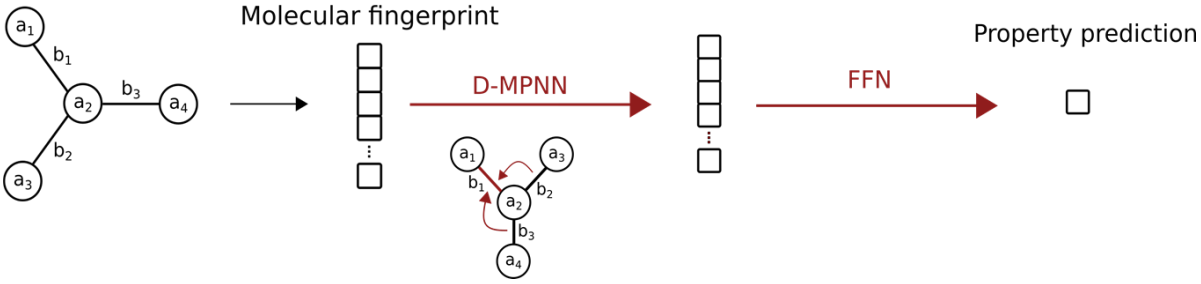

Figure 2. Machine learning model architecture of a message passing neural network