**Dealing with Structural Uncertainties in Lumped Reaction Networks**

Zoltán Till1,\*, Tamás Varga1, János Sója2, Norbert Miskolczi2, Tibor Chován1

*1University of Pannonia, Department of Process Engineering
10 Egyetem Street, H-8200 Veszprém, Hungary*

*2University of Pannonia, Department of Hydrocarbon and Coal Processing
10 Egyetem Street, H-8200 Veszprém, Hungary*

*\*Corresponding author: tillz@fmt.uni-pannon.hu*

**Highlights**

* The definition of the pseudocomponents contributes to the uncertainty of the model.
* In principle, this effect can be separated from the other components of the model error.
* Therefore, the lumping scheme can be optimized within certain limits.

**1. Introduction**

Complex chemical processes are often modeled using lumped reaction networks [1]. Dealing with these models, however, involves some uncertainties. Firstly the overall predictive ability of the lumped kinetic model is limited (due to bias, variance, etc.). Secondly, the kinetic parameters can have broad confidence intervals, mainly because there are a number of alternative pathways in a lumped chemical reaction network that cannot be observed separately. Thirdly, there is an uncertainty in the lumping scheme, i.e. in how we define the pseudocomponents themselves.

In this work, we would like to deal with the latter. Given an experimental data set (of thermo-catalytic pyrolysis of real plastic waste) and number of pseudocomponents to follow (e.g. solids, at least two liquid fractions, gas) we measure the changes of model uncertainty as a function of how these pseudocomponents are actually defined.

The aim of our work is to measure and minimize the model error related to the lumping scheme in case of fixed structural complexity, i.e. for a certain number of lumps and reactions between them.

**2. Methods**

Pyrolysis of real plastic waste was carried out in a two-stage laboratory reactor system with 50 g shredded HDPE/LDPE/PP feedstock using various zeolite-based catalysts at different temperature levels (425/455/485 °C in the 1st stage and 380 °C in the 2nd stage). The gaseous product was driven through a water cooled heat exchanger. Condensate was collected and analyzed by gas chromatography at given time intervals, while the volume of the remaining gas was measured using a gas flow meter [2].

For the kinetic study, we propose a lumped chemical network in which the solid (polymer, coke), liquid (C6-C30+) and gaseous (C0-C5) products are distinguished. Here we would like to consider a straightforward lumping scheme where we group the liquid components into a lighter and heavier fraction (L– and L+) and investigate how this classification affect the uncertainty of the model. It is a somewhat improved model derived from our previous work [3].

**3. Results and discussion**

After minimizing the uncertainty in connection with the identification process and the lumped chemical reaction network and then carrying out the identification procedure consecutively by changing the boundary between the two defined liquid lumps, we were able to relate the change in the squared error between the experimental and calculated data to the uncertainty in the model structure. Apparently, the definition of these pseudocomponents does have an impact on the error of the model, as you can see in Figure 1.

**Figure 1.** Development of total model error while moving the boundary between the two liquid pseudocomponents.

Undoubtedly, there is a limitation in how we define these two lumps, given that we would like to follow the concentrations of specific components, e.g. the boundary between gasoline and Diesel fuel is somewhere between C10-C12; nevertheless, this effect worth not to look over.

**4. Conclusions**

Our results already indicate that the uncertainty inherent to the model structure can be quantified. Nevertheless, there is much to be done. We would like to find the adequate measure of the complexity of the model. There are plenty of these in the literature and we have to find a select the most representative indicator. Moreover, the remaining uncertainty in the reaction network needs to be separated from the uncertainty of the lumping scheme. In any case, the proposed approach appears to be particularly promising and feasible to continue.

**References**

[1] L. P. de Oliveira, D. Hudebine, D. Guillaume, J. J. Verstraete, Oil Gas Sci. Technol. – Rev. D’IFP Energ. Nouv. 71(3) (2016) 45.

[2] N. Miskolczi, J. Sója, E. Tulok, J. Anal. Appl. Pyrolysis 128 (2017) 1–12.

[3] Z. Till, T. Varga, J. Sója, N. Miskolczi, T. Chován, Energy Convers. Manag. 173 (2018) 320–330.