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Kinetic analysis and modeling of evaporation and urea decomposition processes in exhaust gas after treatment systems

Marion Börnhorst, Anna Bertótiné Abai, Steffen Tischer, Olaf Deutschmann

Karlsruhe Institute of Technology, Institute for Chemical Technology and Polymer Chemistry,

Engesserstraße 20, 76131 Karlsruhe, Germany

*Corresponding author: marion.boernhorst@kit.edu

Highlights
- Kinetic measurements of urea decomposition are performed.
- Experimental results indicate deficits of existing kinetic models.
- Based on experimental results a kinetic model for urea decomposition is developed.

1. Introduction

In exhaust gas aftertreatment systems, urea-water solution is injected as a precursor for ammonia, which is used for the selective catalytic reduction (SCR) of nitrogen oxide emissions. Solid deposits formed from urea and its by-products in the tailpipe lead to performance loss of the system. To predict the formation of urea derived deposits at various operating conditions a comprehensive kinetic model is desirable. In this work, kinetic analysis of urea decomposition is performed and experimental data is used for model development.

2. Methods

Deposit samples are derived at a hot gas test bench at different operating conditions. Kinetic measurements of derived deposits and pure components deliver data for modeling urea decomposition.

A laboratory test rig was designed and set up to investigate the formation of urea derived deposits at realistic conditions. The setup includes a hot gas supply, a commercial urea dosing system and a measurement chamber with optical access. Variable parameters of the test rig are the gas flow and temperature, the urea dosing strategy and the wall temperature in the measurement chamber.

Kinetic measurements are performed with pure components (e.g. urea, biuret, cyanuric acid) and the derived deposit samples from the test rig. Thermogravimetric (TG) analyses of the samples combined with Fourier-Transform Infrared Spectroscopy (FTIR) provide kinetic data of the decomposition process and give information about the deposit composition. Analyses are performed using a coupled setup of Netzsch Jupiter STA 449 F5 and Bruker Vertex 70 FT-IR. The experimental conditions are varied concerning the crucible geometry, incident flow direction, flow rate and initial sample mass.

Kinetic measurement data are used to develop and validate a model of the urea decomposition process in DETCHEM. The model represents a 0-D batch type reactor. Evaporation and urea decomposition is implemented based on an existing kinetic model [1].

3. Results and discussion

The decomposition process of urea and its by-products is investigated by TG measurements. As shown in Fig. 1, urea decomposition can be classified by three characteristic decomposition stages. In the first stage, urea decomposes and the by-products biuret and cyanuric acid are formed. Biuret is decomposed in the same stage. The second stage represents the decomposition of cyanuric acid, minor by-products (e.g. ammelide) decompose in the third stage. As shown in Fig. 1, numerical simulation of the decomposition process is in good agreement with the thermogravimetric measurement of sample mass loss. Simulations are performed using an existing kinetic model [1] together with DETCHEM^{EVAPORATOR}. Further experimental results show a strong dependency of the decomposition kinetics of urea to experimental boundary conditions, namely incident flow direction, initial sample mass and surface area. As shown in Fig. 2, great differences in the
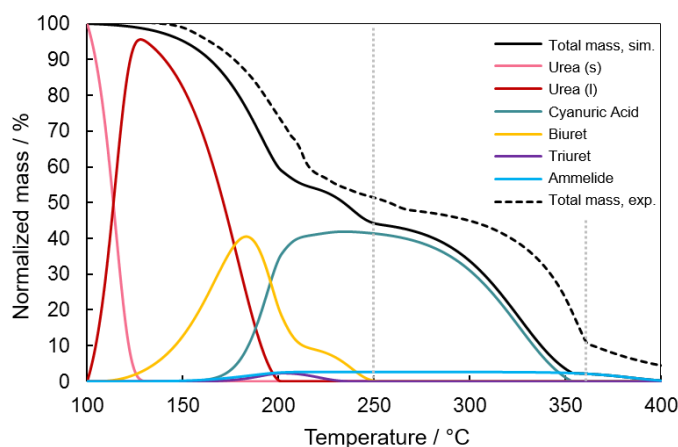
Figure 1. Thermogravimetric measurement and simulation of urea decomposition.
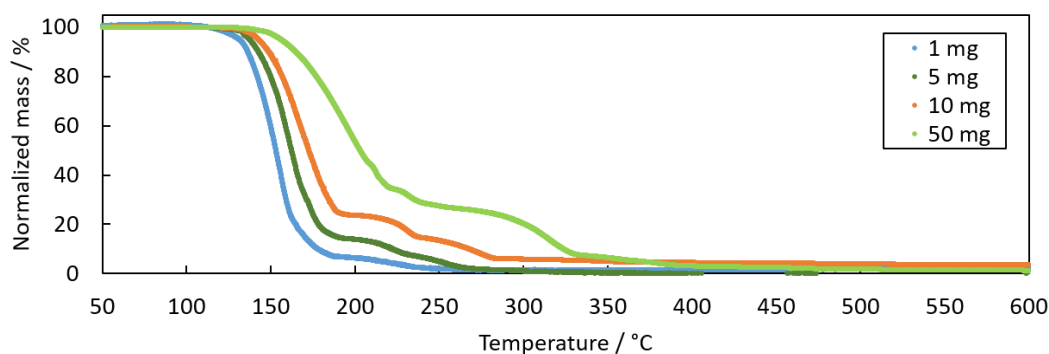

Figure 2. Thermogravimetric measurements of urea decomposition with different initial sample mass.

decomposition rate and in the extent of characteristic decomposition stages can be observed for a variation of the initial sample mass. These differences indicate a strong influence of evaporation processes and surface reactions on the decomposition kinetics. Due to the neglection of gas phase concentrations in the currently used model, the effects of variable surface-to-volume ratios or gas flow rates cannot be predicted by the simulation. This emphasizes the need for comprehensive kinetic modeling, taking surface area dependent reactions and evaporation effects into account.

4. Conclusions

In this work, kinetic measurements are used to develop a comprehensive model for urea decomposition, which is used for the prediction of undesirable deposit formation in exhaust gas after treatment system. Good qualitative agreement of experimental data and simulations is achieved using literature data for kinetic model parametrization. However, effects of important parameters like the surface-to-volume ratio and the gas flow rate, are not considered by the current model. To achieve a reliable prediction of deposit formation in applications, comprehensive experimental studies and model development is necessary.

References

[1] W. Brack, B. Heine, F. Birkhold, M. Kruse, G. Schoch, S. Tischer, O. Deutschmann, Chem. Eng. Science 106 (2014), 1-8.

Keywords
Selective catalytic reduction, Urea decomposition, Thermogravimetric analysis, Kinetic modeling