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Gas Phase Sulfur, Chlorine and Alkali Metal Chemistry in Biomass Combustion
Concern about aerosols formation, deposits, corrosion, and gaseous emissions during biomass combustion, especially straw, continues to be a driving force for investigation on S, Cl, K-containing species under combustion conditions. These trace species contained in the biomass structure will be released to the gas phase during combustion and contribute to the problems generated during the process. The investigation during this PhD project is done to stepwise improve the understanding in the chemistry and reduce the uncertainties. In the present work, the detailed kinetic model for gas phase sulfur, chlorine, alkali metal, and their interaction has been updated. The K/O/H/Cl chemistry, S chemistry, and their interaction can reasonably predict a range of experimental data. In general, understanding of the interaction between K-containing species and radical pool under combustion conditions has been improved. The available K/O/H/Cl chemistry has been updated by using both experimental work and detailed kinetic modeling. The experimental work was done by introducing gaseous KCl to CO oxidation system under reducing conditions. The experiments were performed using a laboratory flow reactor at atmospheric pressure and temperatures in the range of 773-1373 K, varying the CO inlet concentration and the KCl level. The addition of KCl results in a strong inhibition of the CO oxidation. The inhibition increases with the KCl level, but the effect levels off at high concentrations. The experimental data were interpreted in terms of a detailed chemical kinetic model and used to update the K/O/H/Cl chemistry. The oxidation of SO2 to SO3 under combustion conditions has been suggested to be the rate limiting step in the gaseous sulfation process prior to aerosol formation. While the overall mechanisms for SO3 formation and destruction are fairly well known, kinetic modeling of the process still suffers from a lack of accurate kinetic data, and refinements are required in order to establish a reliable mechanism. In this study, the sulfur chemistry important for the SO2/SO3 ratio under combustion conditions has been updated. The uncertainties of the important rate constants have been minimized. Modeling predictions with a revised reaction mechanism for SO2/SO3 chemistry are in a good agreement with a range of experimental data from reactor experiments. The calculations indicate that oxidation of SO2 to SO3 with and without the presence of combustibles involve primarily recombination of SO2 with O and OH radicals. Reaction SO3+H may limit the SO3 concentration, while reactions SO3 with O or OH are unimportant for the SO2/SO3 ratio under most conditions of interest. The ratio of SO2/SO3 in the gas phase system is mainly determined by reactions SO2 + O/ OH, reactions involving intermediate HOSO2, and reaction SO3+H. There has been a controversy regarding the fundamental mechanism of the sulfate aerosol formation. Alkali sulfate aerosol formation may either be initiated in the gas phase by homogeneous mechanism, or it may only involve a heterogeneous mechanism. In this PhD project, the updated K/O/H/Cl and S chemistry, also the interaction of that chemistry are used to investigate the importance of homogeneous gas phase sulfation process prior to aerosol formation. The results of the present study suggest that homogeneous gas phase sulfation may be active. However, heterogeneous mechanism involving catalytic oxidation of SO2 by solid surfaces may also be active in the system investigated.

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