Uncertainty of the rate parameters in the wet CO combustion system

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In recent years, there has been an increased interest in studying the combustion of wet CO as a promising option towards cleaner combustion technologies for power generation from coal or biogenic feedstock in IGCC-CCS plants. Predictive CFD models used in combustor design require accurate kinetic mechanisms to describe the wet CO oxidation chemistry particularly at operating conditions predominant in gas turbines.

Description of the rate parameter k inherently possesses some uncertainty as an inevitable consequence of their experimental determination method or due to assumptions made in theoretical modeling. The uncertainty of even very accurate k measurements is about 30 %, while high level theoretical calculations have an uncertainty of factor 3 [1].

The aim of this work was to improve the performance of a wet CO combustion mechanism and to reduce the uncertainty of crucial rate parameters by applying the optimization methodology of Turányi et al. [2]. Starting from a slightly modified version of the Kéromnès 2013 [3] mechanism, 32 rate coefficients (Arrhenius parameters *A, n, E* and 3rd body collision efficiencies) of the 11 most influential reactions were optimized based on ignition delay time data (786 shock tube data points, 166 RCM data points for hydrogen ignition, 532/444 for wet CO) and reaction rate coefficient measurements (2012 data points). The joint uncertainty domain of all optimized parameters was determined.

Multiple CHEMKIN simulations were performed to compare the newly optimized mechanism to 15 other mechanisms published between 1999 and 2013 with respect to their reproduction of the 976 wet CO ignition delay time data points, 1711 flame velocity data points and 54 concentration profile data points. The mechanism performances were characterized using a sum-of-square normalized deviation-based error function [2].

Due to a large improvement in RCM simulations, the performance of the new mechanism is 19% better in comparison to the second best mechanism. The uncertainty of the rate parameters could be reduced by 50% or more (in terms of f values).

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