Adsorption and diffusion in oxyfuel combustion – Linking experiment and MD simulation through graphite structures as a first example

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During combustion and gasification reactions of solid fuels, gaseous mass transfer is of utmost importance. Gas molecules diffuse through the boundary layer into the porous structure of the fuel, adsorb on the surface, and the gas-solid reaction takes place. Then, the reaction products desorb and diffuse back into bulk phase, where possible further oxidation reactions take place. In combustion modelling, this mass transfer is either neglected or considered in terms of effective diffusion coefficients. However, those coefficients are neither determined by adsorption or diffusion experiments nor modelling, they are derived by fitting the experimentally determined combustion rate to the used conversion model.

Within the framework of the Collaborative Research Centre 129 Oxyflame (CRC), funded by the German Research Foundation, we measure adsorption capacities and kinetics for different gases and solid fuels. Effective diffusion coefficients are calculated from the experimental data by using appropriate models for the adsorption kinetics. Moreover, the mass transfer is modeled using molecular dynamics (MD) simulations. The results of these atomistic simulations are compared to data from macroscopic adsorption experiments. To establish a sound basis for comparison, graphite (see Figure 1) of different porosity, which is also the most fundamental form of the main compound of solid fuels, was selected as adsorbent.

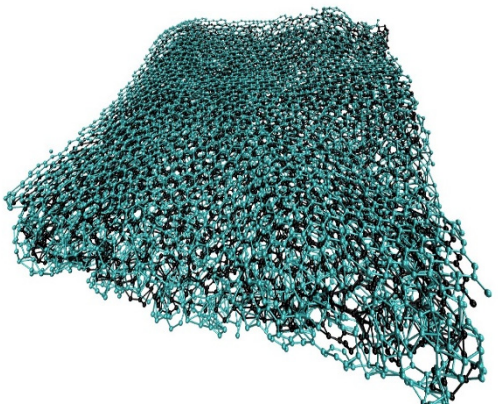

Figure 1: Graphitic structure.

In this work, we present adsorption capacities and kinetics for graphite determined with a commercially-available but deliberately modified gravimetric sorption analyzer (Rubotherm, Germany – since 2016, TA instrument, USA). These measurements are conducted with some of the most relevant gas species during oxyfuel combustion, which are carbon dioxide, oxygen and methane. Our investigations serve as base-line experiments for the evaluation of the MD simulations, since graphite has in contrast to natural solid fuels, like biomass or coal, a clearly defined structure.