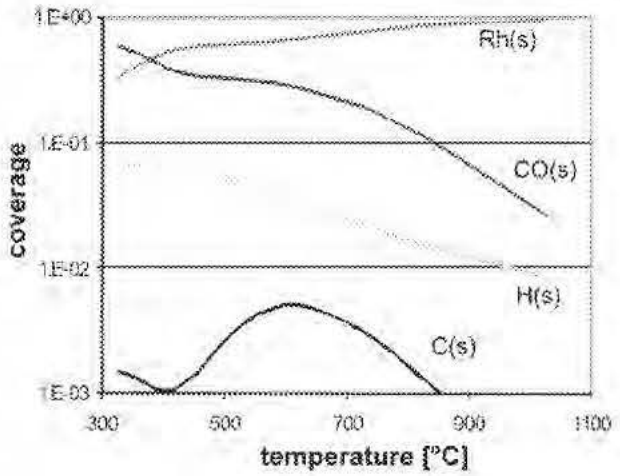

Fig. 20. Computed surface coverage of hydrogen, CO, and carbon in SR of natural gas with S/C 4 und 900 cpsi.

CO(s). An adsorption–desorption equilibrium cannot be reached. If, however, constant ambient conditions, temperature of 927 °C, a pressure of 1 bar and the inlet composition of SR of natural gas with S/C 4 are chosen, an equilibrium calculation using our reactions mechanism (approx. 100 s (=infinite) integration time of the kinetics) leads to a CO surface coverage of 3.5×10^{-5}. Hence, the surface coverage with adsorbed species in reactive systems can be much higher than those observed in TPD studies.

The coverage of the surface with carbon is worth mentioning: C(s) coverage reaches values up to 0.5% at 600 °C, which is also the temperature range that promotes coking as observed in aging studies.

7. Conclusion

Based on extensive experimental studies of SR of the major single alkane components of natural gas and SR of a sulfur-free natural gas mixture, a detailed reaction mechanism for the catalytic conversion over a Rh-based catalyst was developed and evaluated by comparison of experimentally derived and numerically predicted conversion and selectivity. The mechanism was implemented into a two-dimensional flow field description in a single monolith channel and also coupled with an elementary step reaction mechanism for potential modeling homogeneous SR in the gas phase. The mechanism can now be used to predict product distribution in SR of natural gas mixtures with varying compositions. Furthermore, the simulation tools developed allow the numerically simulation of chemical species profiles and surface coverage within catalytic monoliths.

SR of ethane, propane, and butane show almost identical conversion and selectivity as function of temperature and S/C. These dependencies are different for SR of pure methane, which, for instance, is converted as much higher temperature. The higher alkanes were found to have a positive impact on methane conversion in SR of natural gas. In the presence of a catalyst gas-phase reactions are not significant for conversion in SR of natural gas at the conditions studied.

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