

selectivity of CO, with increasing temperatures leads to a decline of the overall hydrogen production. The slope for the higher alkanes is steeper than for methane and their yields for a given S/C are closer together. The curve for the hydrogen yield from natural gas follows below 450 °C the curves of the higher alkane propane and approaches the yield from SR of methane as soon as methane in the natural gas is converted.

Fig. 19 shows the numerically predicted spatial distribution of the species in the gas phase inside the catalytic channel for SR of natural gas with S/C 4. At 327 °C (Fig. 19a) no conversion of methane and only low to moderate conversion of the higher alkanes is observed, nearly all carbon is converted to CO. Low conversion of methane is found at 452 °C (Fig. 19b), while the higher alkanes are already completely converted at this temperature. The rapidly produced CO is re-adsorbed and further reacts to CO₂ via the water-gas shift reaction (19). Further discussion on that issue can be found in our recent paper [47]. At 577 °C (Fig. 19c) the conversion of all reactants is nearly complete. CO₂ is still the main product, but since the influence of the slightly exothermic water-gas shift reaction declines with increasing temperatures, CO is increasingly found in the product stream. A comparison of all

temperatures reveals that the reaction zone is moving to the front of the catalyst with increasing temperatures.

Fig. 20 shows the calculated temperature dependence of the surface coverage in SR of natural gas with S/C 4. At high temperatures, the surface is mostly empty denoted by the vacant sites Rh(s). Molecular hydrogen is available on the surface in the percent range with a maximum coverage of 6.8% at 390 °C and declines with increasing temperatures to 0.8% at 1000 °C. The primarily found species is CO(s) (Fig. 20). At first view, this result produced in terms of the reaction kinetics developed in this study seems to be contradictory to literature [48] claiming the CO coverage to be negligible at elevated temperatures. Other authors [39,49,50] observed in temperature programmed desorption (TPD) experiments a high coverage of CO at low temperatures and desorption temperatures of CO between 182 and 267 °C depending on surface type and experimental set-up. The coverage of CO in our simulation decreases with increasing temperature, from 76% at 200 °C to 2.5% at 1000 °C, which still is a significantly high value and definitely larger than one may expect. This apparent inconsistency can easily be resolved: the very active Rh catalyst is exposed to a hydrocarbon stream that continuously produces new

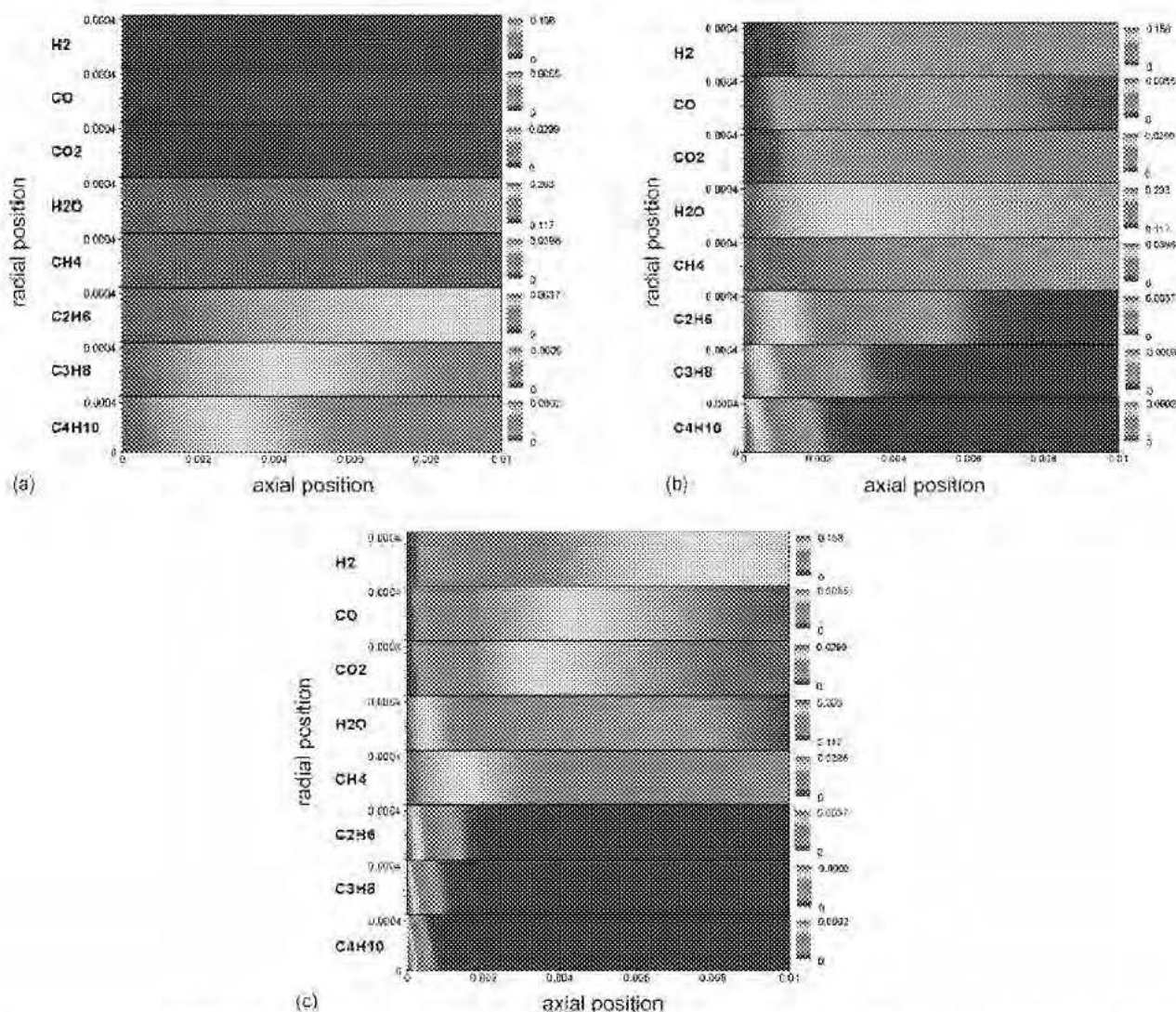


Fig. 19. Spatial distributions of species in the gas phase in the single channel in SR of natural gas with S/C 4 at (a) 327 °C, (b) 452 °C, (c) 577 °C; red indicates the highest and blue the lowest concentration of each species at all temperatures.