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# Computational Fluid Dynamics Simulation of Bio-Syngas Hetero-/Homogeneous Combustion over Platinum in Catalytic Micro-Combustors

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# ABSTRACT

Bio-syngas is a synthetic gas obtained from the thermal gasification of biomass. It contains carbon monoxide and hydrogen which is thought to be a cleaner fuel than biogas owing to its higher hydrogen content. In this work, the bio-syngas hetero-/homogeneous combustion over platinum in catalytic micro-combustors were studied through two-dimensional computational fluid dynamics model. Numerical simulations were carried out with a two-dimensional elliptic computational fluid dynamics code in conjunction with full-elliptic flow description, detailed hetero-/homogeneous chemistry, and all relevant heat transfer mechanisms in catalytic micro-combustors. In addition, we also studied the light-off of mixtures of carbon monoxide and hydrogen under lean conditions. The self-inhibition of carbon monoxide was found. Increasing carbon monoxide compositions result in an increase in the light-off temperature. Addition of hydrogen to the feed causes a reduction in light-off temperature for all compositions of carbon monoxide studied. The most significant shift in light-off temperature occurs with the addition of small amounts of hydrogen with only minor marginal enhancement occurring at higher hydrogen concentrations. Hydrogen alone in a lean atmosphere will oxidize at room temperature. Upon changing the activation energy between adsorbed hydrogen and oxygen, the carbon monoxide was observed to oxidize first, however, no enhancement of light-off was predicted.

Keyword: Bio-syngas; Homogeneous combustion; Heterogeneous combustion; Micro-combustion; Light-off; Platinum catalyst; Computational fluid dynamics

## **INTRODUCTION**

The profound environmental challenges faced by mankind at the dawn of the new millennium have made it abundantly clear that far-reaching changes in our management of energy are required. The use of renewable resources appears to be a necessary condition for achieving and upholding sustainability [1]. Among these, biofuels have to be considered and investigated with a meticulous care since they hold the promise of ensuring a transition from fossil fuels to sustainable ones [2]. Gaseous fuels from biological sources (such as biogas) are particularly promising since they can be produced from various wastes [3]. Biological syngas (generated from the gasification of biomass) is interesting owing to its thermal efficiency [4] and the cleaner character of its burning in comparison to other fuels [5].

Bio-syngas is a synthetic gas obtained from the thermal gasification of biomass. It contains carbon monoxide and hydrogen which is used for the synthesis of various products, and is thought to be a cleaner fuel than biogas owing to its higher hydrogen content. The high hydrogen content of bio-syngas is assumed to imply a cleaner combustion than that achieved with pure methane [6]. Several promising techniques are being developed for ensuring an economically viable production of biogas and bio-syngas [7] which could greatly contribute to sustainability since they are hoped to be carbon

dioxide-neutral. The major difference between bio-syngas and syngas is that the former contains a varying amount of methane, which is normally absent or negligible in syngas generated out of industrial sources such as coal.

Future utilization of bio-syngas catalytic combustion relies on the development of active and stable catalysts as well as on the understanding of the heterogeneous and homogeneous syngas kinetics under industrially-relevant operating conditions. In contrast to the extensive investigations of syngas homogeneous chemistry over broad ranges of mixture compositions and pressures [8], there is a clear lack of corresponding studies in combined hetero-/homogeneous syngas combustion. The non-negligible contribution of homogeneous chemistry, even in practical catalytic reactor geometries with large surface-to-volume ratios [9], requires additional validation of the gas-phase bio-syngas kinetic models that were developed in the absence of heterogeneous reactions.

Catalytic oxidation reactions of carbon monoxide and hydrogen mixtures over noble metals play a key role in the exhaust gas treatment, particularly for the abatement of emissions from the slow-reacting carbon monoxide component. Moreover, catalytic oxidation reactions can be used not only for tail-end emissions abatement, but also as a direct means for fuel combustion. The latter approach is driven by the enhanced catalytic combustion stability at very lean equivalence ratios and the ultra-low nitrogen oxides emissions due to the modest reaction temperatures [10]. In the conventional CST (catalytically stabilized thermal combustion) concept, fractional fuel conversion is achieved in a heterogeneous reactor, which is typically Pt-coated, while the remaining fuel is combusted in a follow-up homogeneous combustion region [11]. Such methods are appropriate for a wide range of fuels that include low calorific value biofuels [12], whereby flame stability is an issue, and hydrogen-rich syngas fuels in which conventional lean-premixed gaseous combustion entails the risk of flashback. Understanding the underlying heterogeneous carbon monoxide and hydrogen kinetics and their interactions over noble metals is thus crucial for the advancement of catalytic converters for emissions control and of catalytic syngas combustors for power generation.

Despite previous work, there are a number of answered questions regarding heterogeneous carbon monoxide and hydrogen kinetics and their interactions over noble metals at the micro-scale. In this work, the heterogeneous chemistry coupling of carbon monoxide and hydrogen over platinum was investigated experimentally and numerically for carbon monoxide/hydrogen/oxygen/nitrogen mixtures in micro-combustors through two-dimensional fully elliptic simulations by treating explicitly heat transfer through the wall. Numerical simulations were carried out with a two-dimensional elliptic computational fluid dynamics code in conjunction with elementary heterogeneous and homogeneous chemical reaction schemes, detailed species transport, and heat transfer mechanisms in the solid wall.

Ghermay *et al.* [13] investigated experimentally and numerically The hetero-/homogeneous combustion of fuel-lean carbon monoxide/hydrogen/oxygen/nitrogen mixtures over platinum is investigated. It is shown that for wall temperatures in the range 975 to 1165 K the heterogeneous pathways of carbon monoxide and hydrogen are largely decoupled. However, for wall temperatures below a limiting value of 710-720 K and for the range of pressures and mixture preheats investigated, CO(s) blockage of the surface inhibits the catalytic conversion of both fuel components. The homogeneous ignition distance is well-reproduced by the model for inlet temperatures  $T_{in} > 426$  K, but it is modestly over-predicted at lower inlet temperatures.

Zheng *et al.* [14] investigated experimentally and numerically the gaseous oxidation of H<sub>2</sub>/CO/CO<sub>2</sub>/O<sub>2</sub>/N<sub>2</sub> mixtures in a platinum-coated channel at fuel-lean stoichiometry. Additional simulations have shown that gaseous oxidation was suppressed at atmospheric pressure due to the intrinsic slow gas-phase ignition kinetics in conjunction with the competition from the catalytic pathway for carbon monoxide and hydrogen consumption. At pressures p > 4 bar, homogeneous combustion was largely controlled by flame propagation characteristics due to the near-wall confinement of the established flames. The decrease in laminar mass burning rates at p > 4 bar led to a push of the gaseous combustion zregion close to the channel wall, to leakage of carbon monoxide and hydrogen through the flame and, finally, to subsequent catalytic conversion of the leaked fuel components.

Mantzaras [15] investigated numerically the catalytic combustion of syngas/air mixtures over Pt in a channel-flow configuration using two-dimensional steady and transient computer codes with detailed hetero-/homogeneous chemistry, transport, and heat transfer mechanisms in the solid. It is shown that the homogeneous chemistry of both carbon monoxide and hydrogen cannot be neglected at elevated pressures, even at the very large geometrical confinements relevant to practical catalytic reactors. The diffusional imbalance of hydrogen can lead, depending on its content in the syngas, to super-adiabatic surface temperatures that may endanger the catalyst and reactor integrity. On the other hand, the presence of gas-phase hydrogen combustion moderates the super-adiabatic wall temperatures by shielding the catalyst from the hydrogen-rich channel core.

Federici and Vlachos [16] investigated experimentally synthesis gas oxidation over a supported Pt/Al<sub>2</sub>O<sub>3</sub> catalyst in a novel microreactor fabricated for studying the intrinsic chemical kinetics of highly exothermic reactions. Carbon monoxide was found to significantly inhibit hydrogen oxidation. In contrast, hydrogen addition promotes carbon monoxide oxidation at low mole fractions but has a small promoting effect at high hydrogen mole fractions. As a result, the apparent reaction order of hydrogen changes from positive to zero. The change in hydrogen reaction order is associated with hysteresis. Possible mechanisms for the observed behavior are discussed.

Schultze *et al.* [17] investigated experimentally and numerically the catalytic and gas-phase combustion of  $H_2/CO/O_2/N_2$  mixtures was investigated in a platinum-coated channel-flow reactor at fuel-rich equivalence ratios 2-7,  $H_2$ :CO volumetric ratios 1:2 to 5:1, and wall temperatures 750-1250 K. Even though the catalytic conversion of the limiting  $O_2$  reactant was nearly transport-limited, the competition between  $H_2$  and CO for  $O_2$  consumption allowed for evaluation of the catalytic kinetics. Under rich stoichiometries, the oxidation of CO(s) via the HCOO(s) reaction pathway was significant. The sensitivity of gaseous combustion on catalytic reactions was strong, exemplifying the need of accurate surface chemistry when modeling syngas hetero-/homogeneous combustion.

Salomons *et al.* [18] presented experimental and modelling results for the oxidation of mixtures of hydrogen and carbon monoxide in a lean atmosphere. Simulations performed using literature mechanistic models for the oxidation of these mixtures predicted that hydrogen ignites first, followed by carbon monoxide, a direct contradiction of the experimental evidence. Upon changing the activation energy between adsorbed hydrogen and oxygen, the carbon monoxide was observed to oxidize first, however, no enhancement of light-off was predicted. The effect cannot be explained by the mechanistic model currently under discussion.

## Numerical models and simulation approach

The channel-flow catalytic micro-combustor consists of two infinite parallel plates coated with platinum catalyst, 4.0 mm long, separated by a gap distance d, as shown in Figure 1. Due to the aspect ratio, the combustor is modeled as a two-dimensional system. Premixed carbon monoxide/hydrogen/oxygen/nitrogen mixtures are fed to the inlet of the micro-combustor, and hot product gases exit the

micro-combustor. The combustor wall is 0.2 mm thick with a constant thermal conductivity  $\lambda_s = 16$  W/m K, heat capacity  $c_s = 700$  J/kg K, and density  $\rho_s = 7200$  kg/m<sup>3</sup>, corresponding to FeCr alloy (a common material for catalytic honeycomb combustors in power generation) [19].



Fig. 1. Schematic diagram of the channel-flow catalytic micro-combustor geometry.

The governing equations were solved for a steady laminar reactive flow in two-dimensional homogeneous micro-combustors. Simulations were performed by using the CFD (computational fluid dynamics) software, FLUENT<sup>®</sup> Release 6.3 [20] was used to perform these simulations. A finite-difference method is used to discretize the two-dimensional continuity, momentum, energy and species conservation equations in the fluid and the two-dimensional energy equation in the wall.

Heterogeneous kinetics was described with a detailed chemical reaction scheme for carbon monoxide and hydrogen on Pt [21] consisting of 27 reactions, 8 gaseous and 9 surface species. The reaction scheme was originally developed for  $CH_4/CO/H_2$  oxidation on Pt [22] and then refined in the analytical work of Zheng *et al.* [23] with new H<sub>2</sub>/CO kinetic data. Adsorption reactions were modeled using a modified Motz-Wise correction formulation [24].

Gas-phase chemistry was modeled with the elementary carbon monoxide and hydrogen mechanism form Li et al. [25] (36 reactions and 13 species); in this mechanism the  $H_2/O_2$ chemistry part has been updated according to the latest kinetic data from Burke et al. [26]. The resulting gaseous mechanism has been recently shown in the experimental and numerical investigation of Brambilla *et al.* [27], who accurately reproduce the onset of homogeneous ignition measured with OH-LIF in the catalytic reactor for fuel-lean  $H_2/CO/air$  mixtures. The contribution of gaseous chemistry was evaluated for the cases with highest surface temperatures by comparing predictions with hetero-/homogeneous chemistry. The resulting differences were negligible and gas-phase kinetics was henceforth excluded in the simulations.

The boundary conditions used in this model are as follows. A fixed flat velocity profile is assumed at the inlet. For the species and energy equations, Danckwerts boundary conditions are employed; i.e., the convective portions of the equations are fixed, and the diffusive portions are calculated implicitly. At the interface between the solid and the fluid, no slip and no normal species diffusive flux boundary conditions are employed. The heat flux at this interface is calculated using Fourier's law and continuity in temperature and heat flux is ensured. A symmetry boundary condition is employed at the centerline between the two plates. At the exit, the pressure is specified and the remaining variables are calculated assuming

far-field conditions, i.e., zero diffusive flux of species or energy normal to the exit. In the bulk of the wall the two-dimensional energy equation is solved. The exterior/top surface of the wall is assumed to obey Newton's law of cooling. It is important to note that all the two-dimensional internal heat transfer within the fluid and the solid are calculated explicitly with the two-dimensional elliptic models without any further simplifications. The exterior convective heat-transfer coefficient is only used for the calculation of the heat flux of the exterior wall edge boundary condition. This heat-transfer coefficient lumps the details of heat loss from the micro-combustor and of the process that utilizes the heat generated by the combustor.

Non-uniform node spacing is employed in this work, with more nodes in the reaction region. The number of nodes varies depending on dimensions, but the simplest one consists of 80 axial nodes by 60 transverse nodes, totaling approximately 4800 nodes. Typical fluid node spacing is 50  $\mu$ m in the axial direction and 8  $\mu$ m in the transverse direction. Typical wall node spacing is 50  $\mu$ m in the axial direction and 20  $\mu$ m in the transverse direction, where the temperature does not vary (only a few nodes are placed in the transverse direction within the wall).

The fluid density is calculated using the ideal gas law. The fluid viscosity, specific heat, and thermal conductivity are calculated from a mass fraction weighted average of species properties. The species specific heat is calculated using a piecewise polynomial fit of temperature [28].

The conservation equations were solved implicitly with a two-dimensional steady-state segregated solver using an under-relaxation method. The segregated solver first solves the momentum equation, then the continuity equation, and then updates the pressure and mass flow rate. The conservation equations are then checked for convergence. Convergence is determined from the residuals of the conservation equations as well as the difference between subsequent iterations of the solution. The pressure was discretized using a "Standard" method. The pressure-velocity coupling was discretized using the "Simple" method. The momentum, species, and energy equations were discretized using a second-order upwind approximation.

The simulations were performed on a Beowulf cluster consisting of  $6 \times Xeon X5670$  processors and 80 GB of RAM. When parallel processing was used, the message passing interface (MPI) was used to transmit information between nodes. In order to achieve convergence as well as compute extinction points, natural parameter continuation was implemented. The calculation time of each simulation varied between 8 hours and several days, depending on the difficulty of the problem and the initial guess.

## **RESULTS AND DISCUSSION**

Figure 2 shows the simulation results for the following conditions: 6 vol.% carbon monoxide in air with 8 vol.% hydrogen addition. All the carbon monoxide and hydrogen is completely consumed within the first few millimeters of the catalytic micro-combustor. The amounts of carbon monoxide and hydrogen is varied in the inlet mixture. Without hydrogen addition, no significant amount of carbon monoxide is

converted, and the catalyst temperature remains at 300 K at these conditions. The catalytic surface is covered with oxygen inhibiting the adsorption and subsequent reaction of carbon monoxide. When a lean hydrogen/air mixture is fed, the catalyst ignites and all the hydrogen is consumed leading to a rapid temperature increase of the catalytic micro-combustor. The reaction starts immediately at the catalyst entrance and hydrogen is completely consumed within few millimeters. At steady state, the catalyst temperature equals the adiabatic temperature. After hydrogen is ignited, carbon monoxide is added. If the catalyst temperature is lower than approximately 750 K, because the heat loss is too large or the initial hydrogen mixture is too lean, the onset of carbon monoxide combustion depends on the carbon monoxide concentration fed. The catalyst temperature slowly increases with increasing carbon monoxide/air ratio until the ignition of carbon monoxide combustion occurs leading to a jump of the catalyst temperature. If the catalyst temperature is high enough (over 800 K for the conditions used here), light-off of carbon monoxide combustion will occur immediately. In this case, all the carbon monoxide is completely consumed even for very low carbon monoxide concentrations.



Fig. 2. Two-dimensional distributions of temperature along with the streamwise profiles of temperature for the nominal combustor geometry. The parameters are l = 4.0 mm,  $u_{in} = 2.0$  m/s,  $T_{in} = 300$  K, h = 20 W/(m<sup>2</sup> K).



Fig. 3. Light-off curves for different inlet concentrations of carbon monoxide.

Transient simulations were conducted to examine the ignition curves for various feed concentrations. We present the curves and data for oxidation of carbon monoxide alone and for mixtures of carbon monoxide and hydrogen. In all cases, the ignition point is defined as the inlet temperature at which the outlet conversion is 50%.

We present first ignition curves for different inlet carbon monoxide concentrations. The reactor and feed gas had an initial temperature near 350 K. The temperature of the feed gas was then increased at approximately 0.133 K/s from 350 to 623 K, where it was held constant for about 30 minutes. In all cases the feed contained 6% (v/v) oxygen, with either 500, 1000, 1500 or 2000 ppm of carbon monoxide. In each case, the start-up procedure was first to flush with nitrogen for 2 minutes, followed by addition of oxygen, which was allowed to flow for a further minute, and finally addition of carbon monoxide. This condition was held for 8 minutes before commencing the temperature ramp. The ignition curves are shown in Figure 3. It is seen that increasing concentration of carbon monoxide causes a higher light-off temperature.

Figure 4 shows the ignition curve for carbon monoxide in the presence and absence of hydrogen. The key new observation is that a small amount of hydrogen enhances significantly the ignition point, but the marginal benefit of additional hydrogen is small. The light-off enhancement with 500 ppm hydrogen is 22 K. We also note that hydrogen ignites at the same temperature as the carbon monoxide when the mixtures were present, as shown in Figure 5. The results here show carbon monoxide beginning to oxidize before hydrogen, consistent with the selectivity of Pt toward carbon monoxide oxidation. However, once light-off is achieved, and active site turnover increases, total conversion of both species is attained. Observations reported here of carbon monoxide inhibiting hydrogen ignition and hydrogen promoting carbon monoxide are consistent with the results reported in the literature.



Fig. 4. Light-off curves for different inlet concentrations of hydrogen with 1000 ppm carbon monoxide.



Fig. 5. Light-off curves for a system of 500 ppm hydrogen with 1000 ppm carbon monoxide.

The results are in general consistent with other experimental results reported in the literature. Self-poisoning of carbon monoxide oxidation is well known, hence the increasing light-off temperature as concentration increases. For mixtures of carbon monoxide and hydrogen, the surface of the catalyst is initially predominately covered by carbon monoxide, which inhibits hydrogen adsorption. We can propose a competitive adsorption between carbon monoxide and hydrogen, with the rate determining step for reaction being the adsorption of oxygen. As the temperature increases, carbon monoxide may begin to react, thus opening the surface. At some extent of reaction, sufficient surface is available for significant adsorption of oxygen, and the conversion of carbon monoxide becomes rapid, and at this point the hydrogen undergoes ignition. It not, however, clear what the mechanism of enhancement of the carbon monoxide ignition is. It has been proposed that the addition of hydrogen to carbon monoxide will reduce the light-off point for carbon monoxide owing to the additional exotherm provided by hydrogen oxidation. However, this proposal is not consistent with our observations. Indeed, since carbon monoxide is preferentially oxidised before the hydrogen, it is difficult to see how the exotherm could be significant. Secondly, and more importantly, the degree of enhancement cannot be accounted for by the exotherm alone. For example, 500 ppm hydrogen would add only a few degrees to the fluid temperature, yet the light-off temperature was reduced by 22 K. Secondly, if the exotherm alone were responsible, then the addition of 2000 ppm hydrogen should have produced a much lower light-off temperature for carbon monoxide. The carbon monoxide light-off temperature curve in Figure 6 shows this graphically, where the degree of enhancement does not vary directly with the hydrogen concentration. Therefore, it is proposed that the enhancement must be a function of the surface reaction mechanism.



Fig. 6. Carbon monoxide light-off temperature as a function of hydrogen concentration for the different activation energy.

## CONCLUSIONS

The bio-syngas hetero-/homogeneous combustion over platinum in catalytic micro-combustors were studied through

two-dimensional computational fluid dynamics model. Numerical simulations were carried out with a two-dimensional elliptic computational fluid dynamics code in conjunction with multicomponent transport and elementary homogeneous and heterogeneous chemical reaction schemes, and heat transfer mechanisms. In addition, we also studied the light-off of mixtures of carbon monoxide and hydrogen under lean conditions. We have found that hydrogen was observed to reduce the temperature at which carbon monoxide light-off occurred, while carbon monoxide inhibited the hydrogen oxidation. Carbon monoxide is oxidized first, with hydrogen oxidation occurring when the carbon monoxide was significantly depleted. The largest enhancement of light-off temperature was achieved at lower hydrogen compositions, and further addition of hydrogen up to higher compositions gave only a slight further reduction. The release of thermal energy by hydrogen oxidation cannot account for the reduction in carbon monoxide light-off temperature.

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