
Hashemi, Hamid; Christensen, Jakob Munkholt; Glarborg, Peter

Publication date: 2013

Document Version
Publisher's PDF, also known as Version of record

Citation (APA):
Hamid Hashemi, Jakob Munkholt Christensen, Peter Glarborg
Department of Chemical and Biochemical Engineering, Technical University of Denmark, 2800 Lyngby, Denmark
E-mail: pgl@ktdu.dk

Abstract
A series of experimental and numerical investigations into hydrogen oxidation at high pressures and intermediate temperatures has been conducted. The experiments were carried out in a high pressure laminar flow reactor at 50 bar pressure and a temperature range of 600–900 K. The equivalence ratio of the mixture was varied from oxidizing to reducing conditions. Moreover, a series of experiments in an oxygen atmosphere instead of a nitrogen atmosphere has been done. A reaction mechanism based on a recent work by Burke et al. has been developed. In addition to modelling of the present experiments, the mechanism is used to simulate published data on ignition delay time and laminar burning velocity of hydrogen. The flow reactor results show that at reducing, stoichiometric, and oxidizing conditions, conversion starts at temperatures of 750–775 K, 800–825 K, and 800–825 K, respectively. In oxygen atmosphere, ignition occurs at the temperature of 775–800 K. In general, the present model provides a good agreement with the measurements in the flow reactor and with recent data on laminar burning velocity and ignition delay time.

Experimental Setup – Laminar Flow Reactor
- Quartz reactor to minimize surface reactions
- Steel pressure shell to achieve high pressures
- Temperature: 600–900 K
- Pressure: 50 bar
- Flow: 3.06 NL/min
- Isothermal Zone Length: 42–44 cm
- Residence time: 6.3–8.0 s
- Measurement via a GC and a Gas Analyzer

Reaction Kinetics Model
- Developed based on the mechanism by Burke et al. [1]
- Updated rates for reactions of:
  - OH + O = O + H₂O
  - H₂O₂ + OH = H₂O + O₂
  - Solution via CHEMKIN-Pro

Results (The Flow Reactor)

Results (Comparison to Available Data)

Fig. 1: Schematic diagram of the high pressure laminar flow reactor

Results (The Flow Reactor)

Results (Comparison to Available Data)

Fig. 1: Schematic diagram of the high pressure laminar flow reactor

Fig. 2: Results of reducing experiments (0.95% H₂ and 0.04% O₂ in N₂, Φ=12.07) at 50 bar pressure.

Fig. 3: Results of reducing experiments (0.95% H₂ and 0.04% O₂ in N₂, Φ=12.07) at 50 bar pressure.

Fig. 4: Results of stoichiometric experiments (0.35% H₂ and 0.15% O₂ in N₂, Φ=0.03) at 50 bar pressure.

Fig. 5: Results of oxidizing experiments (0.18% H₂ and 1.60% O₂ in N₂, Φ=0.05) at 50 bar pressure.

Fig. 6: Results of experiments in oxygen atmosphere (0.17% H₂ and 5.01% N₂ in O₂) at 50 bar pressure.

Fig. 7: Major consumption path of hydrogen.

Fig. 8: Ignition delay time of H₂/O₂/Ar at Φ=0.5 (X₉₅=33.1%). Experimental results from [2].

Fig. 9: Ignition delay time of H₂/O₂/Ar and H₂/O₂/N₂ at Φ=0.5. Experimental results for Ar from [3] and for N₂ from [4].

Fig. 10: Ignition delay time of H₂/O₂/Ar at Φ=1.0 and 4.0. Experimental results from [4].

Fig. 11: Unstretched laminar burning velocity of H₂/Ar at initial conditions of 298 K and 1 atm. Lines denote model predictions and symbols mark experimental data from literature.

Summary
The flow reactor results show that at reducing, stoichiometric, and oxidizing conditions, conversion starts at temperatures above 750 K, 900 K, and 800 K, respectively. In oxygen atmosphere, ignition occurs at temperatures above 775 K. The changes in the model improve its prediction especially at oxidizing conditions. Ignition delay time of hydrogen shows a non-linear trend versus pressure especially at low temperatures. The present chemical scheme used in a constant u & v model is able to predict the trend reasonably well while for a more accurate prediction at low temperatures, it is required to consider device-dependent pressure (and temperature) rise before the ignition. Predictions of laminar burning velocity by the model are within the uncertainty of the experiments. In general, the present model provides a better agreement to the measurements comparing to the base model.

* References