

Catalytic H₂ oxidation on palladium surface

Background

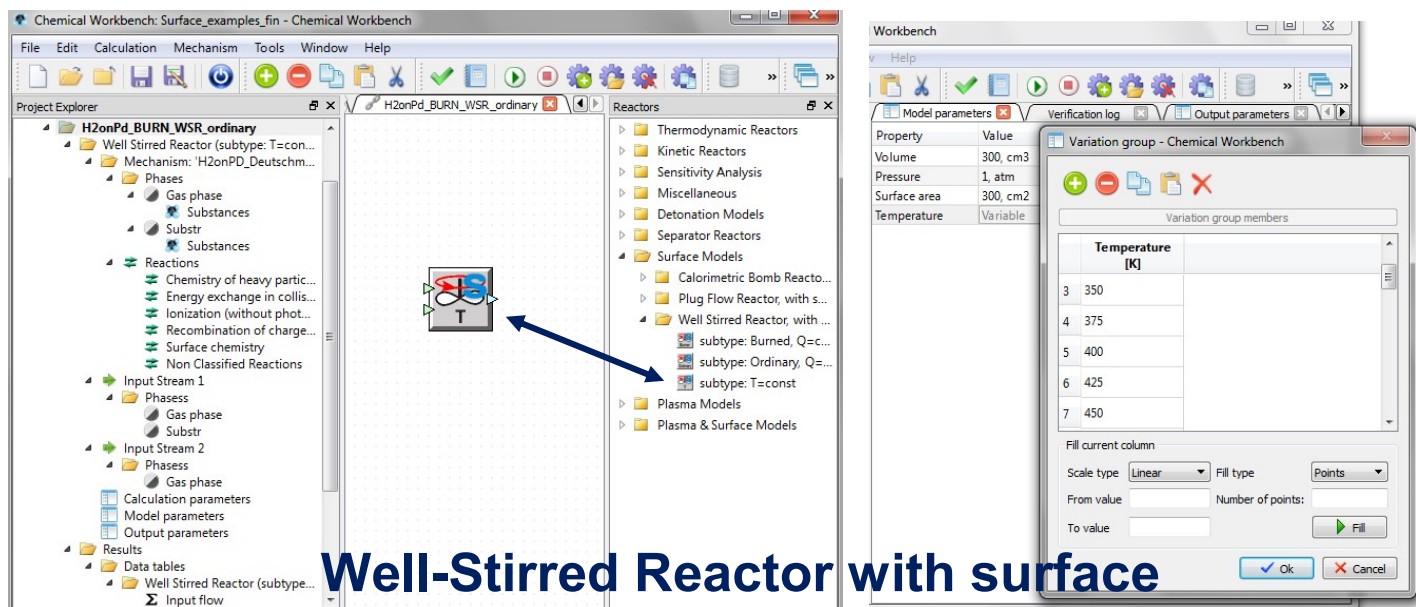
Accurate modeling of chemical processes on a surface is of fundamental importance for heterogeneous catalysis, semiconductor device fabrication, fuel cells, etc. CWB incorporates reactor models, which can be used for the modeling of heterogeneous processes like catalytic combustion. These models assume that chemical reactions can occur in the gas phase and on the catalytically active surface. A surface sites formalism was used to describe heterogeneous reactions that occur at the surface and between the surface and the gas phase. Within this formalism, the surface and condensed phases can possess several different types of sites at which heterogeneous reactions can occur, and the rates of these reactions are calculated according to formal kinetics rules. Catalytic ignition is an abrupt transition from a kinetically controlled system to one controlled by mass transport. Therefore, the complex interactions of chemical and transport processes in the gas phase as well as on the surface have to be included.

Problem statement

It is required to calculate the catalytic H₂ oxidation rate on palladium surface at atmospheric pressure for H₂/O₂/N₂ mixture and varying temperature in the range from 300 K to 1000 K. The ratio of oxygen/hydrogen is fixed 1/2 with 55% of N₂ in the mixture.

Problem setup in Chemical Workbench

The example describes the calculation of the catalytic H₂ oxidation on a Pd surface and it demonstrates an influence of gas temperature on the oxidation rate. The **Well-Stirred Reactor Model with constant temperature** is used, which is a 0 – dimensional reactor with two inlets and one outlet and ideal mixing of the gas mixture between inlets and outlets. The mixture of H₂/O₂/N₂ is set at the inlet. The steady-state composition of gas and surface phase is calculated, together with the rate of heterogeneous combustion.



The screenshot displays the Chemical Workbench interface. The Project Explorer on the left shows a 'Well Stirred Reactor (subtype: T=const...)' selected. The Reactors panel on the right lists various reactor models, with 'Well Stirred Reactor, with...' selected. A 'Variation group - Chemical Workbench' dialog box is open, showing a table of temperature values in Kelvin.

Temperature [K]	Value
3	350
4	375
5	400
6	425
7	450

The dialog box also includes options for 'Fill current column', 'Scale type' (set to Linear), and 'Fill type' (set to Points).

Well-Stirred Reactor with surface

Parameters setup in Chemical Workbench

The following parameters should be specified for **Well-Stirred Reactor** model: Kinetic mechanism, Input streams, Calculation parameters, Model parameters.

In this example the H₂/O₂/N₂ ratio in the Input Stream 1 is 0.3/0.15/0.55, T = 300 - 1000 K, P = 1 atm, flow rate 0.463 g/s, reactor volume 300 cm³ with surface area 300 cm².

The mechanism of heterogeneous hydrogen oxidation on Pd surface is taken from literature (O. Deutschmann et al, Int. Sym. on Combustion, V. 26, 1996, P. 1747-1754) and it includes 2 phases, 40 gas-phase reactions and 13 reversible surface reactions.

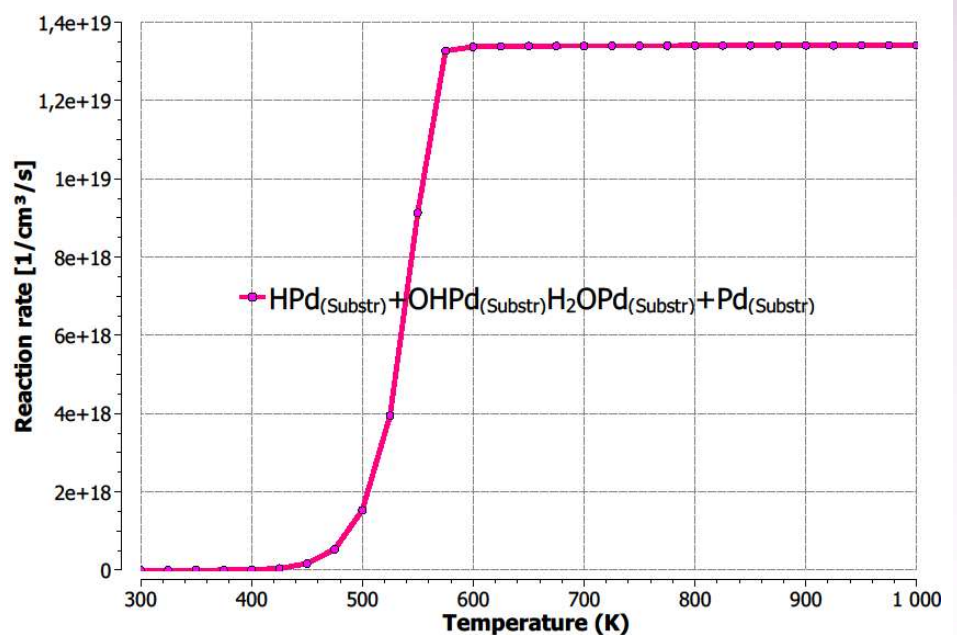
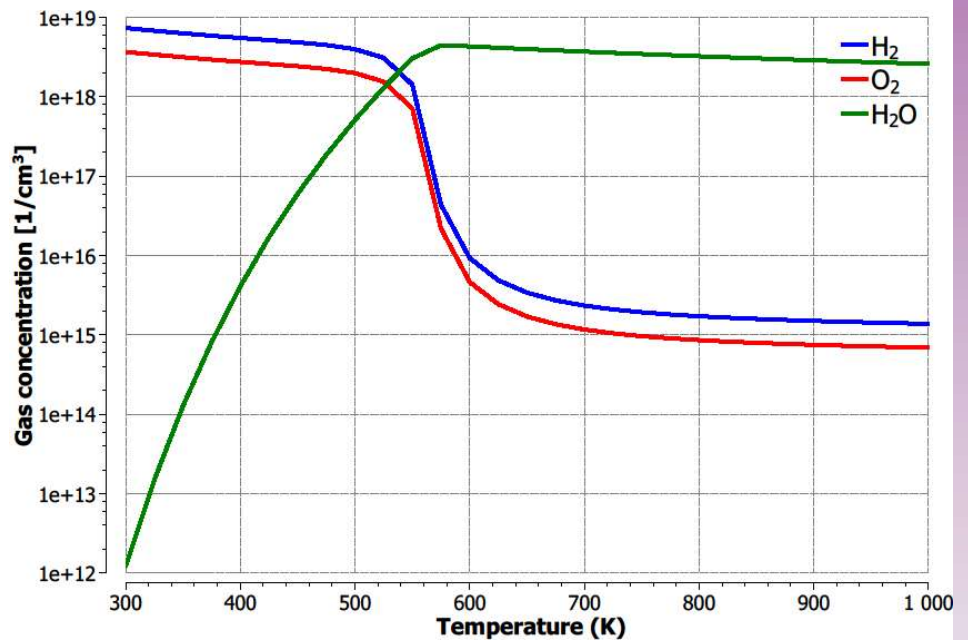
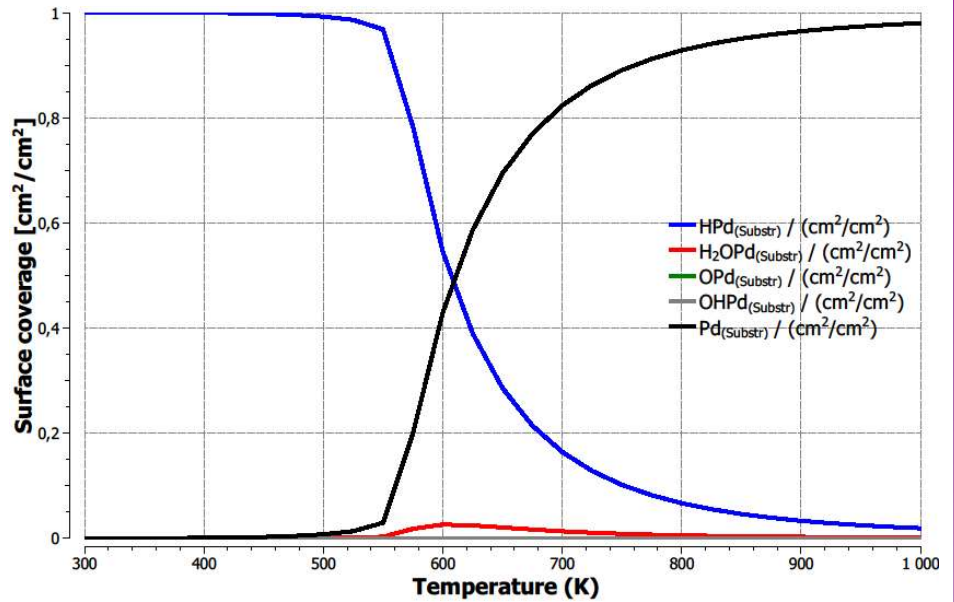
Results

Simulated results are shown in figures below:

Concentrations of surface species for stoichiometric H_2/O_2 ratio for different system temperatures showing heterogeneous oxidation of a H_2/O_2 system: initially surface is fully-covered with atomic hydrogen, for temperatures $> \sim 600$ K effective H_2 oxidation starts.

Corresponding concentrations of gas species for stoichiometric H_2/O_2 ratio for different system temperatures showing heterogeneous oxidation of a H_2/O_2 system: formation of gaseous water H_2O for temperatures $> \sim 600$ K.

Conversion rate in the H_2/O_2 system (the dominant reaction leading to formation of H_2O is shown). At low temperatures Pd surface is covered with hydrogen atoms, conversion rate is low. At temperatures $> \sim 600$ K effective H_2 oxidation starts, a new steady state is established, now controlled by mass transport of reactants toward and products away from the catalyst.



Next steps

This example can be extended further:

1. Change input gas composition (H_2/O_2 ratio) and reactor parameters such as reactor volume, surface area, gas flow rate etc.
2. Compare predictions of different chemical kinetic mechanisms, for conditions of interest for you.