

Laminar flow of a reacting gaseous mixture through a vertical duct with catalytic walls

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Abstract. Numerical analysis has been performed for predicting the onset and establishment of a steady state flow of a reactive hydrogen/air/vapour mixture through a two-dimensional vertical duct of finite length with its side walls coated by catalytic material. The flow is initiated by the exothermic reaction of hydrogen with air oxygen on the catalytic wall, that causes the hot gases to flow upwards through the vertical duct and by continuity sucks fresh mixture through the lower end of the duct. The flow is always laminar and the two-dimensional governing transport differential equations are solved by means of the numerical finite volume method, using a collocated variable arrangement. Comparisons between calculated and experimental data are presented, showing good agreement between them. The method is employed for various initial mixture compositions and duct geometries.

Laminare Strömung einer reagierenden Gasmischung durch einen vertikalen Kanal mit katalytischen Wänden

Zusammenfassung. Die Strömung eines reagierenden Wasserstoff-Luft-Wasserdampf-Gemisches durch einen zweidimensionalen, vertikalen Strömungskanal endlicher Länge mit katalytischen Seitenwänden wurde numerisch untersucht. Die Strömung wird durch die exotherme Reaktion von Wasserstoff mit Luftsauerstoff am Katalysator initiiert. Die heißen, gasförmigen Reaktionsprodukte strömen durch den senkrechten Strömungskanal aufwärts; aus Kontinuitätsgründen strömt von unten frisches Gemisch nach. Die Strömung ist laminar und die zweidimensionalen Transportgleichungen werden nach der Finite-Volumen-Methode auf einem nicht-versetzten numerischen Gitter gelöst. Vergleiche zwischen Rechnungen und Meßergebnissen zeigen gute Übereinstimmung. Die Methode wurde zur Untersuchung der Strömungsverhältnisse bei unterschiedlichen Gemischzusammensetzungen und Strömungskanalgeometrien eingesetzt.

Nomenclature

C_k	mass fraction of mixture component k
C_p	heat capacity
D	width
D_k	mass diffusion coefficient of mixture component k
f	body force vector
g	gravitational vector
h_r	reaction enthalpy
K_{react}	reaction velocity
Le	Lewis number
m	exponent for property evaluation
\dot{m}_k	mass flux of mixture component k
n	in normal direction

P	pressure
Pr	Prandtl number
S_T	source term in temperature equation
S_{C_k}	source term in mass fraction equation
Sc	Schmidt number
t	time
T	temperature
U	velocity vector
U, V	velocity in x - and y -direction
x, y	space coordinates

Greek symbols

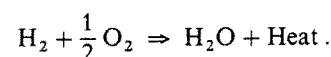
α_k	expansion coefficient due to mass fraction differences of component k $\left(= -\frac{1}{\rho} \frac{\partial \rho}{\partial C_k} \right)$
β	thermal expansion coefficient $\left(= -\frac{1}{\rho} \frac{\partial \rho}{\partial T} \right)$
η	steady state normalized hydrogen reaction rate $\left(= \frac{C_{\text{H}_2, \text{in}} - C_{\text{H}_2, \text{out}}}{C_{\text{H}_2, \text{in}}} \cdot 100\% \right)$
λ	thermal conductivity
μ	dynamic viscosity
ρ	density
τ	shear stress tensor
Φ	general transported quantity

Subscripts

bulk	at the 1st grid point away from the wall
in	at the inlet
k	for mixture component k
out	at the outlet
ref	reference value
wall	on the wall

1 Introduction

In an environment containing a hydrogen/vapour/air mixture the presence of a suitable catalytic surface will cause the hydrogen to react in a controlled manner with the oxygen of the air to form vapour, according to



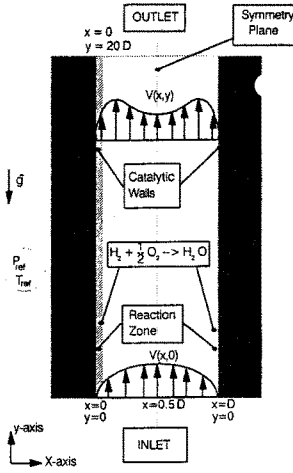


Fig. 1. Schematic of the general arrangement

Even if the mixture is at rest, the reaction enthalpy will heat the mixture near to the catalytic surface and will cause it (if possible) to flow upwards due to buoyancy.

In case the catalytic surfaces form a vertical duct with a finite length, the mentioned buoyant forces will induce an upwards directed flow anyway and, if there is enough hydrogen provided in the environment, this flow can become self-sustained and steady. The mixture leaving the vertical duct at its upper end will contain more vapour and less hydrogen and will have a higher temperature and velocity than the mixture entering the duct from its lower end.

In the present study the following problem has been simulated by numerical methods: A vertical square duct (Fig. 1) of finite length, whose walls are coated with a catalytic material, is in an environment containing a hydrogen/vapour/air mixture. The hydrogen reacts on the catalytic surface with the oxygen to form vapour. The released reaction heat of approx. 240 kJ/kmol H_2 heats the mixture layers close to the wall. The density of these layers decreases due to higher temperature and lower specific volume. This causes them to flow upwards through the duct and leave it through its upper end. By continuity, fresh, not yet reacted mixture (with a prescribed composition, temperature and pressure) enters the duct through its lower end, reacts at the wall and flows upwards. After some time this flow will become self sustained and steady.

The following parameters are believed to be important for the described problem:

1. Duct geometry
2. Initial mixture composition
3. Catalytic material.

The influence of (3) has not been investigated in this study, assuming a perfect catalytic material forcing every two hydrogen molecules, that reach the catalytic surface, to react with an oxygen molecule. The reaction velocity, when there is excess oxygen in the mixture can be given, for a

temperature range between 450–1070 K, by an Arrhenius-type formula [1]

$$K_{\text{react}} = 14.0 \cdot e^{\frac{463}{T_{\text{wall}}}} [1/\text{s}]. \quad (1)$$

T_{wall} being the temperature of the catalytic surface in [K]. This expression implies a 1st order reaction for the hydrogen.

Since the extension of the duct in Fig. 1 perpendicular to the xy -plane is much larger than its two other dimensions the problem is treated as two dimensional. The Reynolds number formed with the width D as characteristic length and with the inlet velocity lies in the range 300–500, i.e. the flow is always laminar.

2 Theoretical analysis

2.1 Governing equations

The two-dimensional governing equations can be expressed as follows.

- Continuity

$$\nabla U = 0. \quad (2)$$

- Momentum

$$\frac{\partial(\rho U)}{\partial t} + \rho U \nabla U = -\nabla P + \nabla \tau + \rho f. \quad (3)$$

τ being the shear stress tensor $\tau = -\mu \nabla U$ and f stands for the body force vector. The latter includes only the gravitational vector g . This buoyancy term, when modelled using the Boussinesq approximation, can be written as:

$$\rho f = \rho_{\text{ref}} \cdot \left\{ [1 - \beta(T - T_{\text{ref}})] + \left[1 + \sum_{k=1}^N \theta_k (C_k - C_{k,\text{ref}}) \right] \right\} \cdot g.$$

N being the number of the mixture components, i.e. buoyancy effects due to both temperature and mass fraction gradients are considered during the solution procedure.

- Temperature

$$\frac{\partial(\rho T)}{\partial t} + \rho U \nabla T = \nabla \left(\frac{\mu}{Pr} \nabla T \right) + \rho S_T. \quad (4)$$

- Mass fraction of a mixture component

$$\frac{\partial(\rho C_k)}{\partial t} + \rho U \nabla C_k = \nabla \left(\frac{\mu}{Sc} \nabla C_k \right) + \rho S_{C_k}. \quad (5)$$

Since pressure variations are only small, the molecular properties are assumed to be depending only on the temperature. For single mixture components the density is being calculated by the ideal gas equation of state, while the remaining relevant properties (i.e. molecular viscosity, specific heat capacity and thermal conductivity) are calculated by a “power law” assumption $q = q_{\text{ref}} \left(\frac{T}{T_{\text{ref}}} \right)^m$, q being an arbitrary prop-

erty and q_{ref} its reference value at the reference temperature T_{ref} . The exponent m receives different values for different properties according to [2]. The mixture's properties are calculated from formulas provided by [3].

2.2 Boundary conditions

The computations include only the flow of the mixture in the duct. The computational domain (CD) is shown in Fig. 1. Because of the axial symmetry only half of the duct is being considered. The southern boundary of the CD is treated as inlet and the northern boundary as outlet plane. The eastern boundary is a symmetry line and the western boundary is treated as rigid wall, upon which the chemical reaction occurs.

The set of elliptic partial differential equations mentioned above can be solved with the following boundary conditions:

Inlet

The normal velocity $V(x, y=0)$ is prescribed, following the distribution for laminar viscous flow in a duct

$$V(x, y=0) = V(D/2, y=0) \left(1 - \left(\frac{2x}{D}\right)^2\right).$$

The average value $\bar{V}(y=0)$ can be calculated from the exiting mass flow at the upper end of the duct in the previous time step and

$$V(D/2, y=0) = \frac{3}{2} \bar{V}(y=0).$$

The tangential velocity $U(y=0)$ is set to 0. The temperature and the mixture composition are set equal to their ambient values.

Outlet

The gradients for all flow quantities normal to the outlet plane (y -direction) are set to 0.

Symmetry

The normal velocity $U(x=D/2, y)$ is set to 0 as well as the gradients of all other quantities normal (in x -direction) to the symmetry plane.

Wall

All velocities are set to 0 due to friction on a rigid wall. In this study the wall is coated by a catalytic material and a reaction occurs on its surface. The temperature at the wall can be calculated by a formula provided in [4].

$$T_{\text{wall}} = T(x=0, y) = T_{\text{bulk}} + \frac{h_r C_{\text{H}_2, \text{bulk}}}{\rho C_P \sqrt{Le}} \cdot \frac{1}{1 + \frac{D_{\text{H}_2} x_{\text{bulk}}}{K_{\text{react}}}}. \quad (6)$$

K_{react} can be calculated from Eq. (1). The subscript "bulk" denotes values close to the wall (from the numerical point of view, values at the closest to the wall grid node in the CD in x -direction). Le denotes a Lewis-number for the hydrogen mass transfer to the wall

$$Le = \frac{\lambda}{\rho C_P D_{\text{H}_2}}.$$

In cases there is excess oxygen in the mixture the wall hydrogen concentration is always set to 0 (assumption of a perfect catalyst) $C_{\text{H}_2}(x=0, y) = 0$.

Due to mass diffusion there is a hydrogen mass flux directed towards the wall, which can be written, according to Fick's law as:

$$\dot{m}_{\text{H}_2} = -D_{\text{H}_2, \text{air}} \frac{\partial C_{\text{H}_2}}{\partial x} = -D_{\text{H}_2, \text{air}} \frac{C_{\text{H}_2, \text{bulk}}}{x_{\text{bulk}}}. \quad (7)$$

The corresponding oxygen mass flux towards the wall is

$$\dot{m}_{\text{O}_2} = 8 \cdot \dot{m}_{\text{H}_2} = -D_{\text{O}_2, \text{air}} \frac{\partial C_{\text{O}_2}}{\partial x}. \quad (8)$$

This relationship provides the boundary condition for the wall oxygen concentration gradient if the hydrogen mass flux in Eq. (7) is computed with values of the previous iteration. An analogous relation holds for the vapour mass flux towards the wall and the vapour concentration gradient

$$\dot{m}_{\text{H}_2\text{O}} = -9 \cdot \dot{m}_{\text{H}_2} = -D_{\text{H}_2\text{O}, \text{air}} \frac{\partial C_{\text{H}_2\text{O}}}{\partial x}. \quad (9)$$

These relations hold analogous when there is excess hydrogen in the mixture. In this case the wall oxygen concentration is set to 0 and the concentration gradients for hydrogen and vapour are calculated from the oxygen mass flux computed with values from the previous iteration.

2.3 Numerical method

The whole set of partial differential equations for continuity, momentum and scalar quantities with their initial and boundary conditions are first reduced to algebraic difference equations using the finite-volume method [5] by integrating them over small discrete control volumes formed in the arranged numerical grid. A collocated grid arrangement is used, i.e. all variables are stored in the center of the control volume. In order to avoid checkerboard pattern oscillations, when solving the pressure correction equation, which are likely to occur when using this variable storage scheme [5], a special interpolation procedure for the calculation of the velocities at the cell faces is adopted [6–9].

The values of convective and diffusive fluxes through the cell faces are calculated by using upwind and central differences respectively and weighing their contributions to the coefficients of the resulting algebraic equation for each point P by means of the deferred correction scheme [10]. The

general structure of the final finite volume equation for a general variable Φ is:

$$A_P \Phi_P = \sum_N A_N \Phi_N + S^U + S^P \Phi_P. \quad (10)$$

A_P , A_N are the finite volume coefficients for the point P and its 4 neighbours, S^U and S^P are the integrated source terms, S^P being the linearized part. Equation (10) is solved with the iterative SIP¹ method [11]. The equation set, consisting of Eq. (10) written down for each unknown (U , V , P , T , and C_k) is solved with the SIMPLE² algorithm [12].

The following convergence criterion is used at each time step: The normalized residuals for each equation should decrease by a factor 1000 in order to accept the solution for a discrete time step as converged [13].

3 Computational results and discussion

Figure 2 shows for a mixture with an initial composition of 3 vol-% hydrogen, 50 vol-% vapour and air the distribution patterns (iso contour plots) for the concentration of hydrogen, vapour and for the temperature after the flow through the duct has reached a steady state (approx. 7–8 minutes after initiation). The ambient pressure is 2 bar and the ambient temperature 400 K. The Reynolds number for the steady state flow (using the duct-width D as the characteristic length) is 330, i.e. the flow is laminar. One can see, that the reaction occurs mainly close to the lower end of the duct, while in the upper part of the duct almost no additional vapour is produced. This is due to the high near wall velocities of the already produced vapour, that do not allow hydrogen to come close to the wall in the upper duct regions.

The following Fig. 3 shows the temporal development of the averaged inlet velocity for the same initial and ambient conditions like before. The inlet velocity has been averaged over the duct width D . The steady state flow is reached after approx. 7–8 minutes. Also shown are experimental data (from [1], marked as \square in the figure). The experimental data were obtained using a velocimeter, located in the center of the duct, just below its lower end. However the measured values lie in the lower end of the measuring range of the used instrumentation, so they can not be regarded as accurate enough. The qualitative agreement of calculated and measured data however seems to be quite good.

Figure 4 illustrates the computed and measured [1] temporal development of the temperature in the duct's center at the outlet plane for 2 different inlet hydrogen concentrations (5 vol-% and 7 vol-%) in the mixture. The inlet vapour concentration for both cases is 50 vol-%. The agreement is thought to be good.

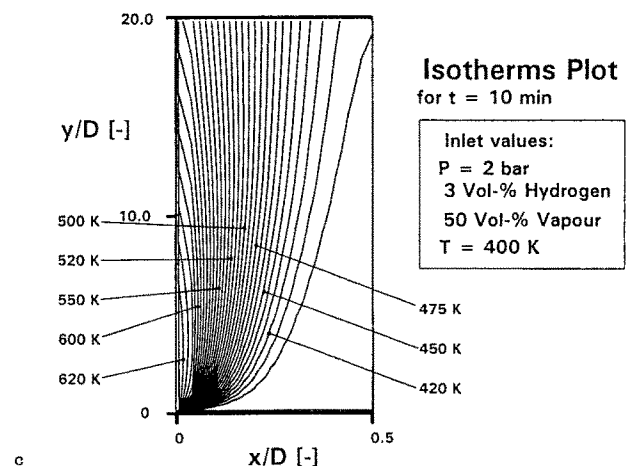
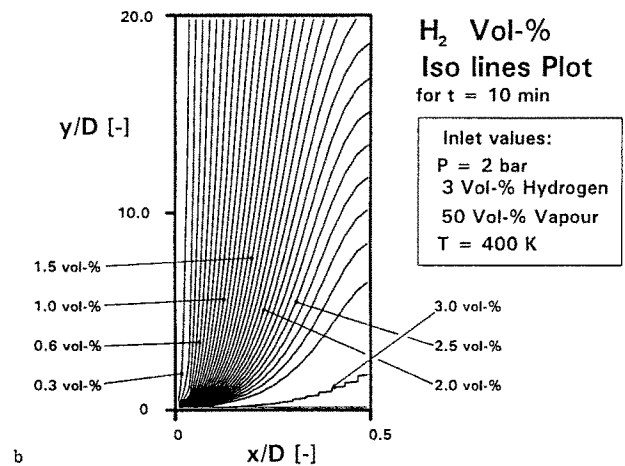
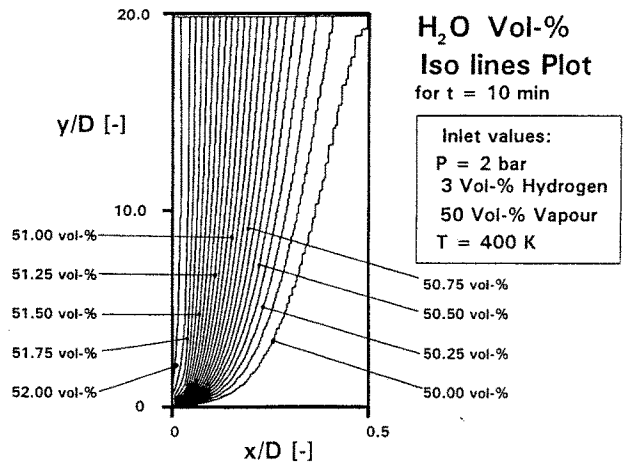


Fig. 2a–c. Iso contour plots of the steady state distribution patterns for (a) hydrogen, (b) vapour and (c) temperature of a mixture with an initial composition of 3 vol-% hydrogen, 50 vol-% vapour and 47 vol-% air. Ambient pressure 2 bar, ambient temperature 400 K, Reynolds number 330

¹ Strongly Implicit Procedure

² Semi Implicit Method for Pressure Linked Equations

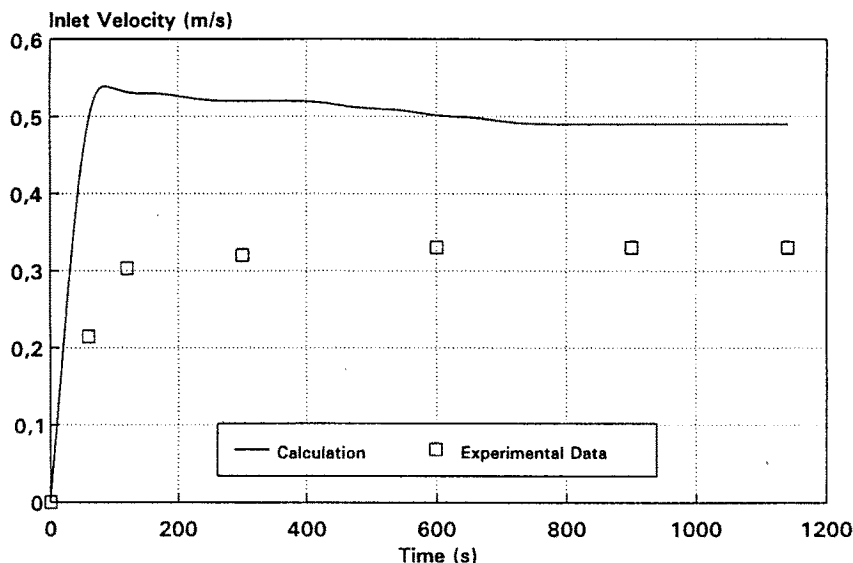


Fig. 3. Calculated (—) and measured (□) temporal development of the averaged inlet velocity

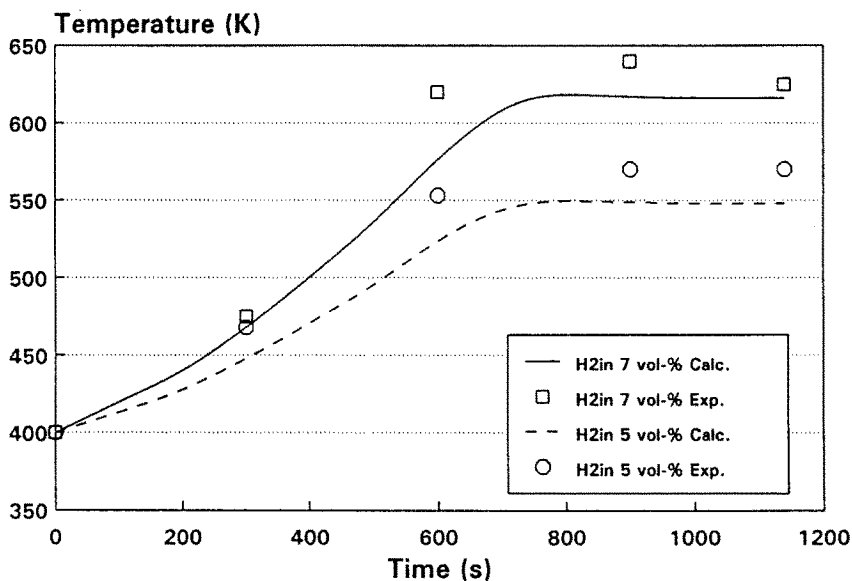


Fig. 4. Comparison between calculated and measured ([1]) temporal development of the outlet temperature in the duct's center for ambient hydrogen concentrations of 5 vol-% and 7 vol-%

The next Fig. 5 shows the effect of variations of the ambient hydrogen and vapour concentration on the steady state normalized hydrogen reaction rate

$$\eta = \frac{C_{H_2, in} - C_{H_2, out}}{C_{H_2, in}} \cdot 100\%$$

When varying the ambient hydrogen concentration the ambient vapour concentration measured constant 50 vol-% and when varying the ambient vapour concentration the ambient hydrogen concentration was constant 3 vol-%. The effect on the hydrogen reaction rate is quite small, when varying the ambient concentrations of hydrogen and vapour within these ranges.

Figure 6 shows the effect of geometry variations on the steady state values of η , velocity at the inlet and temperature

at the outlet. One can see that increasing the duct's height results into an increment of the overall reaction rate, the inlet velocity and the outlet temperature. This result is reasonable since increasing the duct's height is equivalent with a larger catalytic surface and more reaction possibilities for the mixture components. Widening the duct allows more of the mixture to flow through inactive regions (i.e. the duct's center) and as a consequence the overall reaction rate decreases. The values for η , the inlet velocity and the outlet temperature are therefore lower in that case.

All computations were performed on orthogonal numerical grids with uniform spacing and grid points varying between 4000–7200 on the CRAY Y-MP supercomputer system of the Technical University of Munich.

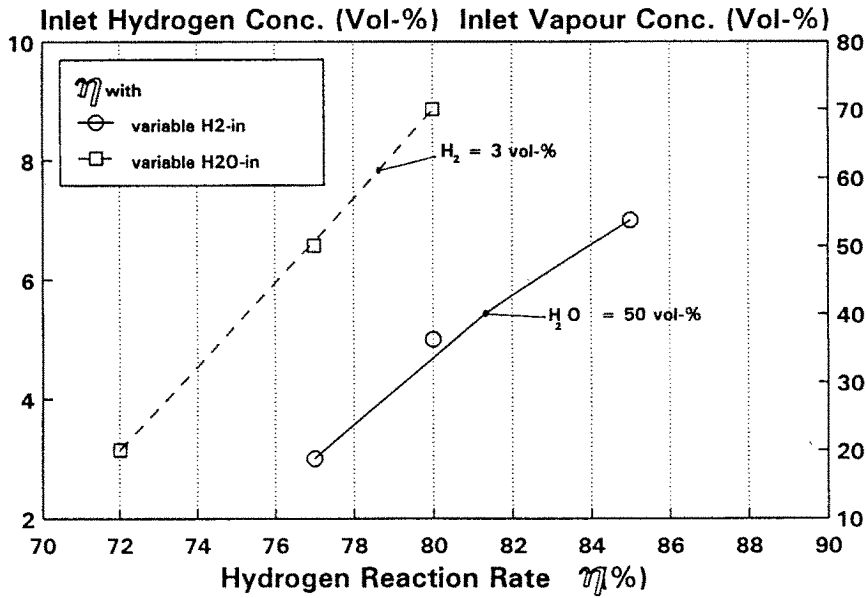


Fig. 5. The effect of variations of the ambient hydrogen and vapour concentration on the steady state normalized hydrogen reaction rate η

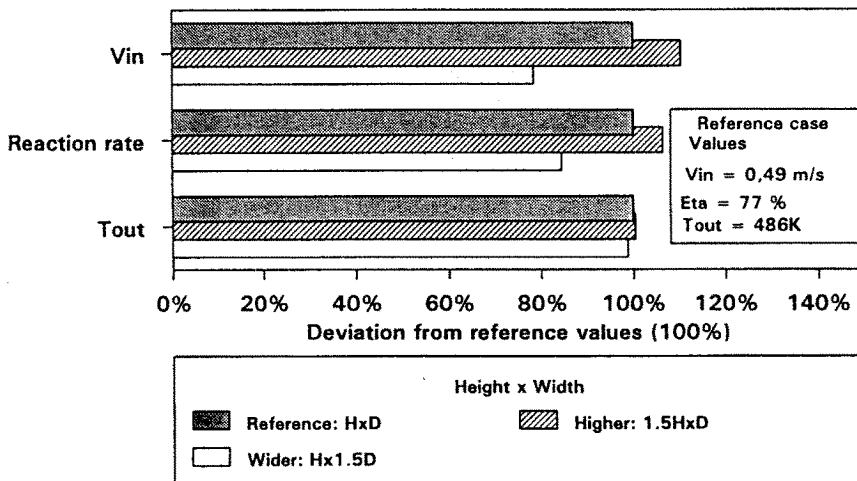


Fig. 6. The effect of geometry variations on the steady state values of η , inlet velocity and outlet temperature

4 Concluding remarks

A method is presented, able to calculate the onset and establishment of a steady state flow of a reacting gaseous mixture in a vertical duct with walls coated by a catalytic material. A finite-volume scheme is used for the numerical treatment of the governing equations.

Calculations were performed for different geometries and inlet conditions. The nature of the resulting flow was always laminar. The reaction on the duct's walls has been accounted for by a simple model, presented in [4]. Comparisons between computed and measured data show the method's ability to predict both the temporary development as well as the steady state values of the flow field.

The significance of the concrete geometry on the hydrogen reaction rate is shown to be crucial, whereas variations of the mixture composition at the inlet (however within nar-

row ranges, as far as the initial hydrogen concentration is concerned) is shown not to have such a great effect on the normalized reaction rate for the hydrogen.

The presented simulations demonstrate the method's feasibility and capability of producing correct results, within the ranges of engineering accuracy requirements.

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