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Computer simulation approach in development of propane-air combustor microreactor

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Abstract

A mathematical model and simulation results of a new microreactor for propane-air combustion are presented. In this article we review 3D model of microreactor and solve problems of gas dynamics, heat transfer and chemical kinetics. The model is based on Navier-Stokes equations for a transient laminar flow of a compressible chemically reacting gas mixture, together with the mass transfer and energy balance equations. The simulation results were applied under development of microreactor geometry to solve a problem of fuel burning process stability. As a main result we have obtained temperature distribution in the microreactor during a fuel combusting process under different mass flow rates of propane and air. Flame shape and its long term stability were also investigated. To validate the model, we developed a microreactor prototype and performed a series of experiments varying fuel consumption. Experimental data have a high correlation to the simulation results and our model correctly predicts gas flows, flame shape and long term stability of oxidation process. The validated computational model will be used to investigate more complex microsystems base on combustion microreactors

Keywords: microsystems, microreactor, CFD, multiphysics simulation, micro combustor

1 Introduction

High-performance computing technologies and computer modeling is becoming an important, powerful and effective tool for design of advanced systems in modern world. Simulation approach effectively solves complex multiphysics problems and is widely used in engineering practice for development of innovation products. One of the most important classes of such products is microsystems, which include micro electromechanical systems (MEMS), microfluidics, electro optical devises, etc. According to resent research and forecasts, industry of microsystems has experienced a huge constant growth for the two last decades and has a strong base for its evolution and extension in future [1]. Computer simulation approach is one of the key components of the microsystems progress.
At first, modeling approaches were used to solve mechanical problems in elementary MEMS such as accelerometers, gyroscopes and pressure sensors. Nowadays in addition we should consider fluid dynamic problems for reacting flows, heat transfer problem and take into consideration some specific effects under development of new microsystems. It requires more complex models and high-performance-computing technologies. The best illustration of a computer simulation approach applied in engineering practice is studying the development process of new propane-air combustor microreactor.

As is known, modern portable communication electronic devises, micro robots, micro unmanned air vehicles and other numerous systems require reliable, low-weight, high capacity and cost effective source of electric energy. Currently, advanced lithium batteries are widely available to be used in microsystems. However, they are large, have a short operation time and low energy to weight ratio [2]. For these reasons, their usage in electronic and MEMS devices seems to be limited, so other available sources of energy should be developed and applied. Hydrocarbon fuels due to their high energy to weight density and their low cost are advantageous for some critical application. By these reasons, it is necessary to develop high efficient fuel burning microreactor, which is compatible with MEMS conditions [3].

2 State of the art in developing and modeling of combustion microreactors

Combustion at small scale reveals various technical challenges. Papers [4,5] present a short overview and consider some fundamental problems of combustion and power-generation systems at small scales. The main problems are oxidation reaction stability and avoidance of combustion quenching by the wall. This technical challenge for micro combustion implies from large surface to volume ratio that is inherent to microsystems. High ratio results in heat losses causing flame stability issues, which affects to the ratio of heat loss to heat generation. In general, micro combustors have small residence time, which makes it important to have small chemical times to make sure complete combustion of the reactants takes place. There are several ways to achieve this small chemical time which includes rising combustion temperatures, preheating of reactants, stoichiometric operating conditions and usage of fuels with high energy density [4,5]. Usage of catalytic burning is another way to increase stability of oxidation reactions, as the process involves diffusion of species to the wall, absorption/desorption at the wall that determines the residence time. There are a lot of research where catalytic combustion is used in microreactors [6-9]. The main drawbacks of catalytic burning are low temperature of the oxidation reaction, high cost of catalytic material and its low reliability including using contaminated fuel. By these reasons non-catalytic microcombustors are prevalently developed.

There are a lot of prototypes of microreactors for non-catalytic fuel burning which have a ‘swiss-roll’ structure [10,11]. In these devises, combustion chamber is located at the centre of the burner and structure provides heat transfer from the hot exhaust gases to the cold incoming reactants. These solutions allow obtaining stable combustion in thin tubes with diameters smaller than the quenching distances.

Another technical issue is experimental investigation of the micro combustion systems, because they have small dimensions of critical parts - around several millimeters or less. Chamber is hidden from direct visual observation. Applying of temperature, heat flux, pressure and chemical sensors is limited by their dimensions, which have the same order of magnitude as microreactor elements. By these reasons, methods of computer simulation are widely used in development and investigation processes. Some papers dedicated to CFD simulation of fuel oxidation in catalytic reactors [6-9]. The models include heterogeneous reactions. Gas dynamic boundary layer and surface chemical processes should be simulate correctly to obtain a proper model of combustion processes. But these models
frequently does not contain volume chemical reactions that simplify mathematical model and, in general, reduce a computer resources.

Informative research [12] about flame dynamics simulation in catalytic and non-catalytic mesoscale microreactors allow comparing two types of micro combustors. In this paper hydrogen/air flames are investigated numerically in a 2-mm-height planar channel with platinum-coated walls, as a function of the inlet velocity and catalytic reactivity. The catalytic reactivity is controlled by varying the parameter, which denotes the ratio of the catalytically active area to the geometrical channel surface area. It was revealed that the non-catalytic case provides non-stationary repetitive ignition/extinction and oscillating flames, that shows stabilized effect of catalytic coating. In the other paper [13] dynamics and stabilization of CO/H₂CO/H₂/air atmospheric pressure flames in mesoscale channels was investigated numerically using detailed gas phase chemistry and transport. Numerical results revealed different flame modes that included oscillatory ignition, random ignition spots, as well as steady weak and V-shaped flames. Also the Computational Singular Perturbation (CSP) method was used for detailed investigation of flame behavior. In the work [14] authors experimentally and numerically investigate H₂/CO/O₂/N₂ premixed combustion in a 7-mm height mesoscale channel at atmospheric pressure. 2-D model including detailed species transport perfectly simulates all stationary combustion modes that included V-shaped and asymmetric (upper or lower) modes, in terms of flame shapes and flame anchoring positions. It was found that simulations of the oscillatory flames were very sensitive to specific boundary conditions, which can be adjusted basing on accurate experimental measurements. A model of combustion system with submillimeters channels for liquid fuel supplying was described in [15]. After comparison with experimental data, it was found that numerical simulation method application is reliable to investigate. Authors also note that radiation heat transfer from liquid droplet and boundary slip conditions have significant effect on numerical simulation. In paper [16] effects of different chemical kinetic models were numerically investigated for premixed methane/air micro flame. Authors of this work give overview of hydrocarbon oxidation reaction applied in many CFD models. It was found that some simple chemical schemes do not take into account all the kinetic phenomena for appropriate numerical simulations of the micro combustion. Unfortunately, this work does not include experimental data and their conclusions may be generalized by the following thesis: the model of chemical kinetic has some influence on the simulation results.

In this paper we present new microreactor for non-catalytic oxidation of gas fuel. Our design and computes model have three specific features. Firstly, we consider a model without fuel-air premixing. Propane and air are supplied independently of each other and we investigate a mixing process in the microreactor chamber. Secondly, our microreactor design does not allow axial or mirror symmetry so we apply 3D model for detailed investigation. Thirdly, we consider the microreactor surrounded by air that allows clarify boundary conditions and prepare a system to ordinary experimental measurements for model verification. In this paper, we describe a microreactor design, mathematical model and its implementation. In addition, we present simulation results for temperature distribution, reaction rate and other parameters and provide model verification by experimental data.

3 System design

Propane-air combustor microreactor is shown in Figure 1. The microreactor is represented by a chip with dimensions of 20x10x1 mm. It consists of silicon (1) and glass (2) parts bond together. In silicon wafer microchannels and chamber (3) are etched by RIE process at the depth of 300 μm. Fuel and air are supplied into the chamber from feeder microchannels (4, 5) and are mixed into its volume. Reagents go outside from chamber through outlet fins (6). Flame and zone of oxidation reaction (7) are situated after the chamber outlet.
Thermal energy of the reaction increases the chip temperature in which heat transfers from outlet to inlet. Some part of the energy is spent on increasing of temperature of the fuel-air mixture in the chamber. Another part is wasted in the surrounded environment (8) because the chip is located in air and its surfaces are cooling by free convection.

The core aim of our propane-air combustor microreactor investigation is to observe a flame shape and temperature distribution in the chip under different fuel consumption.

4 Mathematical model

4.1 Governing equations

In our system we consider two types of volumes: solid silicon-glass chip and other liquid area for chamber and surrounding air. For all the volumes where gas flows, a mathematical model is based on continuum approach. The approach is justified at Knudsen numbers $Kn < 10^{-4}$ calculated for our conditions. Gas media consist of some chemical species from which we take into account five components: $C_3H_8$ (fuel), $O_2$ and $N_2$ (air), $H_2O$ and $CO_2$ (reaction products). Under these conditions the transport equation for species $i$ is:

$$\frac{\partial}{\partial t}(\rho Y_i) + \frac{\partial}{\partial x_j}(\rho u_j Y_i) = \frac{\partial}{\partial x_j} J_{ij} + M_i \omega_i$$

where $\rho$ is density, $Y_i$ is mass fraction of species $i$, $J_{ij}$ is diffusive flux, $M_i$ is molar mass of species $i$, $\omega_i$ is production rate of species $i$.

To this equation we join equation of impulse conversation for viscous Newtonian fluid:

$$\rho \frac{dV}{dt} = \rho F - \text{grad} p + \mu \Delta V$$

where $V$ is velocity vector, $p$ is pressure, $F$ is mass force (in our model it is gravity force), $\mu$ is mixture gas viscosity.

To calculate the mixture viscosity equation (3) is used:

$$\mu = \sum_{i=1}^{N} \frac{\Phi_{ij} \mu_i}{\sum_{j=1}^{N} \Phi_{ij}}$$

where, $\Phi_{ij}$ dimensionless quantity given by (4), $\mu_i$ is viscosity of species $i$, defined by Sutherland’s Law (equation (5))

$$\Phi_{ij} = \left(1 + \frac{M_i}{M_j}\right)^{\frac{1}{2}} \left[1 + \left(\frac{\mu_i}{\mu_j}\right)^2 \left(\frac{M_j}{M_i}\right)^{\frac{1}{2}}\right]^{\frac{3}{2}}$$

$$\mu_i = \frac{A_i T^\frac{3}{2}}{(B_i + T)}$$

where $A_i$ and $B_i$ are Sutherland’s constant for species $i$.

Density of gas mixture calculated by ideal gas state equation (6)

$$\rho = \frac{P \cdot M_{mix}}{R \cdot T}$$

where, $M_{mix}$ is molar weight of gas mixture, $R$ is gas constant, $T$ is temperature. Molar weight of gas mixture defined at equation (7)
The system is completed by thermal energy balance equation (8)

\[ \frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\rho \mathbf{v} h) = \nabla \cdot (k \nabla T) + \frac{dp}{dt} + S_h \]

where \( h \) is total enthalpy, \( k \) is thermal conductivity, \( \tau_{ij} \) viscous stress tensor, \( S_h \) is additional sources due to chemical reactions.

Thermal conductivity calculated from Prandtl number (9), which in our model was constant 

\[ \operatorname{Pr} = 0.707 \]

Thermal parameters of species are determined by JANNAF method by equations (10, 11)

\[ C_p/R = a_1 + a_2 T + a_3 T^2 + a_4 T^3 + a_5 T^4 \]  

\[ H/RT = a_1 T^2/2 + a_2 T^3/3 + a_3 T^4/4 + a_4 T^5/5 + a_5/T \]

For each species constants \( a_1 - a_5 \) are specified in NIST-JANAF Thermochemical Tables.

In our model we consider a reaction of propane oxidation (12) [17]

\[ C_3H_8 + 5O_2 \rightarrow 3CO_2 + 4H_2O \]  

From the reaction it is possible to calculate a production (consumption) rate of species \( i \) by modified Arrhenius equation (13)

\[ \omega_i = A_p \exp \left( -\frac{E_a}{RT} \right) \Pi_{j} \left( \frac{P_j}{M_j} \right)^{\alpha_i} \]

where \( A_p \) is pre-exponential constant, \( E_a \) is activation energy, \( \alpha_i \) is power factor for species \( i \). Constants using for the reaction in our model are presented in Table 1.

| Reaction                     | \( A_p \)       | \( E_a \)       | \( \alpha_{C3H8} \) | \( \alpha_{O2} \) |
|------------------------------|----------------|----------------|-------------------|-----------------|
| \( C_3H_8 + 5O_2 \rightarrow 3CO_2 + 4H_2O \) | \( 5.62 \cdot 10^9 \) | \( 1.256 \cdot 10^5 \) | 0.1               | 1.65            |

Table 1. Reaction parameters. Units are [kmol-m-s-kJ-K]

For solid volumes in our model (silicon and glass parts) we consider a heat transfer problem by equation (14)

\[ q = -\lambda \text{grad} \ T \]

where \( q \) is heat flux and \( \lambda \) is thermal conductivity. Properties of the solid materials are presented in Table 2

| Properties                   | Silicon | Glass |
|------------------------------|---------|-------|
| Density, kg/m\(^3\)         | 2330    | 2220  |
| Specific heat, J/kg\cdot K   | 714     | 820   |
| Thermal conductivity W/m\cdot K | 149     | 1.12  |

Table 2. Properties of the solid materials

### 4.2 Boundary and initial conditions

Boundary conditions for volumes surrounding the microreactor set a pressure of 100000 Pa and temperature of 300 K and provide a free air convection model around the microreactor. To supply a fuel and air mixture into the chamber we set a constant mass flow rate \( Q_{fuel} \) and \( Q_{air} \). All walls of microreactor have sticking conditions for velocity.

The model of propane-air combustor microreactor is unsteady so we should set initial conditions. But the reaction of propane oxidation requires an activation energy (or an ignition source). For this reason, we used the next approach. Firstly, we calculated a steady model, set as initial conditions a high temperature for all volumes. It allowed to “start” the oxidation reaction and to obtain temperature distribution in the flame area. Secondly, we created an unsteady model set an initial temperature 300 K for all volumes except for the flame area for which we applied a previous solution as initial conditions.
This approach solved the problem of ignition source and provided accurate results of temperature distribution in microreactor in due course.

5 Implementation

5.1 Model discretization

Finite Volume Method is used for model discretization. Equations from mathematical model can be expressed in the form of a generalized transport equation (generic conservation equation) for a quantity $\Phi$.

$$\frac{\partial \rho \phi}{\partial t} + \nabla (\rho \mathbf{V} \Phi) = \nabla \cdot (\Gamma \nabla \Phi) + S_{\Phi}$$

(15)

Integrating this equation over a control-volume cell, we have:

$$\int_V \left( \frac{\partial \rho \phi}{\partial t} \right) dV + \int_V \nabla \cdot (\rho \mathbf{V} \Phi) dV = \int_V \nabla \cdot (\Gamma \nabla \Phi) dV + \int_V S_{\Phi} dV$$

(16)

The transient term in equation (16) is integrated as follows

$$\int_V \left( \frac{\partial \rho \phi}{\partial t} \right) dV = \frac{\rho_0 \mathbf{V} \Phi^o - \rho \phi \mathbf{V} \Phi}{\Delta t}$$

(17)

where the superscript $o$ denotes an older time, while no superscript denotes the current or the new time. The convection term is discretized as follows

$$\int_V \nabla \cdot (\rho \mathbf{V} \Phi) dV = \sum_e \rho \mathbf{V} \Phi \cdot \mathbf{n} dA = \sum_e \left( \rho_e \Phi_e V_e^n \right) A_e = \sum_e C_e \Phi_e$$

(18)

where subscript $e$ denotes one of the faces of the cell, $A_e$ is the area of face $e$, $V_e^n$ represent the velocity component in the direction that is normal to the face, $C_e$ is the mass flux across the face.

The diffusion term is discretized as follows:

$$\int_V \nabla \cdot (\Gamma \nabla \Phi) dV = \sum_e \Gamma_e \left( \frac{\partial \Phi}{\partial n} \right)_e A_e$$

(19)

where the term $\partial \Phi/\partial n$ can be written as:

$$\frac{\partial \Phi}{\partial n} = \frac{1}{\delta} \left( \frac{\partial \Phi}{\partial e} - \vec{e} \cdot \nabla \frac{\partial \Phi}{\partial \tau} \right)$$

(20)

where

$$\frac{\partial \Phi}{\partial e} = \frac{\phi_e - \phi_p}{\delta_{p,e}}$$

(21)

$$\frac{\partial \Phi}{\partial \tau} = \frac{\phi_{c2} - \phi_{c1}}{\delta_{c1,c2}}$$

(22)

where $\delta_{p,e}$ and $\delta_{c1,c2}$ represent the distance between $E$ and $P$, and $C2$ and $C1$, respectively (See Figure 2)

5.2 Geometry model

To provide numerical calculations of discrete mathematical model we create a 3D geometrical model of propane-air combustor microreactor in accordance with geometry of chip. We used an unstructured tetragonal mesh for all volumes. Figure 3 presents the mesh in the model. To investigate some details of gas flow and fuel-air mixing into the chamber we create some additional geometric models, which include only the microreactor chamber. Figure 4 presents a structured mesh in chamber
volume and microchannels between outlet fins. This trick represents an engineering approach to computer simulation of complex systems under limiting computational resources.

Figure 3. Mesh in 3D geometry model.  
Figure 4. Structures mesh in chamber volume for detailed investigation.

6 Simulation results and discussion

As we note above an independent fuel and air supply is a distinguishing feature of our microreactor. By this reason, we firstly investigate gas flow and mixing process into the microreactor chamber. Figures 5 and 6 show a propane distribution in the chamber volume and stream lines near the inlets under three different inlet velocities. These figures illustrate changes of diffusion mixing where we increase flow velocity. High flow rates provide creating 2D eddies that have an influence on C\textsubscript{3}H\textsubscript{3} distributions near the inlet, as it shown in Figure 6

Figure 5. C\textsubscript{3}H\textsubscript{8} distribution in the mixing chamber (left) under different mass flow rates of C\textsubscript{3}F\textsubscript{8} and air a) 1 \times 10^{-7} kg/s, b) 2 \times 10^{-7} kg/s, c) 6 \times 10^{-7} kg/s (XOY cross section)

Figure 6. Stream lines near the inlets (right) under different mass flow rates of C\textsubscript{3}F\textsubscript{8} and air a) 1 \times 10^{-7} kg/s, b) 2 \times 10^{-7} kg/s, c) 6 \times 10^{-7} kg/s (XOY cross section)
Fins located near chamber outlet reduce a cross section and increase a velocity of the flow. It restricts a probability of flame propagation into the mixing chamber because the velocity is higher than flame speed. Basing on this simulation results we obtain a range of suitable propane and air flow rates which then are used in the entire model of combustor microreactor.

The general view of flame simulation in the microreactor model is presented in Figure 7 where temperature distribution is depicted. Maximum temperature of 2183 K in the flame corresponds with the means values from others models described in literature, [16] for example.

![Figure 7. Temperature distribution shows a flame area in 3D model.](image)

Figure 7. Temperature distribution shows a flame area in 3D model.

![Figure 8. Temperature distribution in silicon part (a), chamber (b) and glass part (c) (X0Y cross section)](image)

Figure 8. Temperature distribution in silicon part (a), chamber (b) and glass part (c) (X0Y cross section)

Figure 8 shows a temperature distribution in silicon, glass parts and chamber. Silicon has a high thermal conductivity and by the reason has a more uniform temperature distribution than glass part with low thermal conductivity. Cold gases inlet the chamber, cooling a central area of glass. Though propane-air mixture increases its temperature during the flow through the chamber, the mixture is heated before burning. Maximum temperature gradient on microreactor surface is 262 K on glass and 65 K on silicon. Maximum temperature is 660 K on glass surface. Nonsymmetrical distribution of temperature can be explained under a flame shape consideration. Behavior of flame under different mass flow rate of propane is illustrated in Figure 9. It shows three cases and depicts oxidation reaction rate and \( \text{C}_3\text{H}_8 \) distribution. For all cases propane distribution is nonsymmetrical that is result of mixing between fuel and air that was described above. In addition, we note an area of reaction in right corner under the main flame zone. This area is a result of air dilution of fuel mixture in the chamber. Rate of oxidation reaction has a strong dependence of oxygen concentration so the air supply in the right side of chamber provides an oxygen rich zone after chamber outlet where we observe this addition flame.

Changes of the flame shape are the result of increasing gas velocity at chamber outlet. As it is known, stable flame is realized when gas velocity and flame propagation speed are equal. For propane-air mixture mean flame propagation speed is around 0.3 m/s and dependent from oxygen concentration and total pressure. In considered system design, flame area is not restricted by walls and so we have a wide range of fuel consumption where flame is stable. Increasing a propane mass flow rate only changes a flame shape and dimensions. For the lowest value the high of flame is 2.2 mm for the highest is 3.4 mm.
Non-steady simulation shows a stability of propane oxidation process for considering range of fuel consumption during a long time. Flame does not have any oscillations or wave processes. This indicate a high quality of microreactor design which allows applying it for future development of power-MEMS devises.

7 Model verification

We carried out several experiments with microreactor prototype for the model verification. The prototype was produced by microsystem technology according to described design. Then capillary tubes were connected to silicon-glass chip to provide fuel and air supply into the chamber. This system was installed into the test equipment that controls mass flow rate of air and propane with high accuracy. To investigate a flame area a high-resolution photo camera with advanced optics was used.

Photo of the flame in the microreactor is shown in Figure 10 a). There we see a nonsymmetrical flame shape. For proper investigation of experimental photo we converted original data to black-and-white image (Figure 10 b) and compared it to the simulation result presented in Figure 10 c. It is easy to note a full correlation of flame shape and dimensions between experiment and simulation results. Long time (more than 5 minutes) experimental investigation of flame stability also proves simulation results for non-steady model. This facts shows our modes is right predict gas flows and flame area. It allows apply the model for future optimization of microreactor geometry and for investigation of operation processes.
8 Conclusions and future work

We have developed and validated a mathematical model of propane-air combustor microreactor. The model is based on Navier-Stokes equations for a transient laminar flow of a compressible chemically reacting gas mixture, together with the mass transfer and energy balance equations. The model was implemented for 3D geometry of silicon-glass microreactor. Mixing processes of propane and air into the chamber was investigated in details. This feature of microreactor defines a nonsymmetrical CH₃H concentration distribution, flame shape and temperature gradient on the chip surface. Increasing of propane mass flow rate leads to growth of flame area in dimensions. Non-steady simulations showed a high stability of flame. The microreactor prototype was made for model verification. Experimental data have a high correlation to simulation results. The model predicts gas flows, flame shape and long-term stability of oxidation process correctly. In the future developed model will be applied to microreactor optimization and for detailed investigation of operation process.

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