Numerical modeling of subsonic axisymmetric reacting gas flows

E E Peskova
Ogarev Mordovia State University,
68 Bolshevistskaya Street, Saransk, 430005, Russia
Boreskov Institute of Catalysis SB RAS,
5 Lavrentyev Avenue, Novosibirsk, 630090, Russia
E-mail: e.e.peskova@mail.ru

Abstract. A numerical algorithm is developed and implemented for modelling axisymmetric subsonic reacting gas flows based on a previously created program for plane flows. The system of Navier-Stokes equations in the low Mach number limit is used as a mathematical model. Calculations of ethane pyrolysis for axisymmetric and plane flow of mixture at heat supply from the reactor’s walls are carried out. Through the interplay of the developed code and the code for plane flows it becomes possible to identify the geometric factor role at the presence of a large number of nonlinear physicochemical processes. We found that diffusion of synthesized molecular hydrogen mainly influences heat supply from the reactor’s walls to gas and pyrolysis products distribution along its length.

1. Introduction
Scientific knowledge about radical chain reactions of chemical transformations in substances has been accumulated for the last two centuries. Pyrolysis of hydrocarbons to produce ethylene and propylene is an important application of this knowledge for the modern chemical industry. These endothermic processes are carried out in pyrolysis pipes absorbing heat of the external sources. When studying the pyrolysis of ethane and other light hydrocarbons, it is often assumed that the radical reactions occur in the gas environment. The temperature of the reaction gas inside the pipe is lower than at its surface. Since the rate of radical reactions is increased with temperature raising, the radicals are firstly generated on the pipe’s hot walls in heterogeneous conditions. The importance of the chemical processes with a heterogeneous component has been emphasized before, in particular, in the articles of the collection devoted to N. Semenov’s 70th anniversary [1]. This component has been studied in reaction vessels by changing the ratio of the reactor’s surface area to its volume [1].

Presently the radical chain reactions of hydrocarbons have become more interested for scholars. This is caused by possibility of additionally influencing pyrolysis with powerful laser radiation, which resulted in the development of laser conversion of ethane [2, 3]. Therefore, some problems those hadn’t been studied before became relevant, i.e. the geometric shape of the reactor influences the chemical process of hydrocarbon conversion. Diffusion of radicals, especially of hydrogen atoms, heat transfer along with powerful absorption of thermal energy during the chemical reactions can influence hydrocarbon conversion differently in various flow geometry.

Currently computational experiments have become possible due to computer technology and development of numerical methods. The necessity to take into account viscosity, thermal conductivity,
diffusion of radicals those differ in molecular weight, and complexity of the reacting gas greatly consideration leads to extremely long calculations in three-dimensional space. However, it is possible to determine the geometric factor influence on the yields of the target products if we calculate and compare the transformation of ethane in the planar and cylindrical geometries of the calculated region with the energy flow from the heated walls of the reactor. To do this, you need to create two programs in two geometries for mathematical modelling of the dynamics of a multicomponent gas with chemical reactions, significant energy absorption and with a change in the volume of the gas. These programs should be created on the basis of the same numerical methods and similar computational algorithms in order to minimize the influence of the numerical methods themselves on the differences in solutions.

Recently a computational algorithm has been developed, implemented and validated to solve two-dimensional Navier-Stokes equations in the low Mach number limit [4,5]. The present paper describes the modification of the developed algorithms and the code if axisymmetric flows are using, as well as the comparison of the obtained calculation results for ethane pyrolysis in planar and cylindrical pipes.

2. Mathematical model

We use the system of Navier-Stokes equations in low Mach number limit [6] to describe the subsonic flow of a multicomponent gas mixture. We write the equations in the cylindrical coordinate system for the axisymmetric flow [7] in the following vector form:

\[
\frac{\partial U}{\partial t} + \frac{\partial}{\partial z} \left( F^{(1)}(U) - H^{(1)}(U) \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( r F^{(2)}(U) - H^{(2)}(U) \right) = Q. \tag{1}
\]

Here

\[
U = \left( \begin{array}{c}
\rho Y_m \\
\rho u_z \\
\rho u_r \\
\rho h \\
\end{array} \right); \quad F^{(1)}(U) = \left( \begin{array}{c}
\rho u Y_m \\
\rho u u_z + \pi \\
\rho u u_r \\
\rho u h \\
\end{array} \right); \quad F^{(2)}(U) = \left( \begin{array}{c}
\rho u Y_m \\
\rho u u_z + \pi \\
\rho u u_r \\
\rho u h \\
\end{array} \right); \quad H^{(1)}(U) = \left( \begin{array}{c}
J_{mc} \\
\tau_{zz} \\
\tau_{rr} \\
q_z \\
\end{array} \right); \quad H^{(2)}(U) = \left( \begin{array}{c}
J_{mr} \\
\tau_{zz} \\
\tau_{rr} \\
q_r \\
\end{array} \right); \quad Q = \left( \begin{array}{c}
R_m \\
0 \\
0 \\
0 \\
\end{array} \right). \tag{2}
\]

The system is complemented with the equation of state for mixture of perfect gases and the equation for the enthalpy of the mixture:

\[
p = \rho RT \sum_m \frac{Y_m}{M_{uu}}; \quad h(T,Y_m) = \sum_m Y_m h_m(T). \tag{3}
\]

Here \( m = 1, \ldots, M \), \( M \) - a number of components in gas mixture, \( \rho \) – density, \( Y_m \) – mass fraction of species \( m \), \( u_z \), \( u_r \) - projections of the velocity vector \( \vec{u} \) on the axis \( z \) and \( r \), \( h \) – enthalpy of the mixture, \( \pi \) – perturbational pressure, \( R_m \) – range of production or consumption of \( m \) – component of the mixture, \( T \) – temperature, \( M_{uu} \) – molecular mass of \( m \) – component of the mixture, \( R \) – universal gas constant, \( h_m(T) \) – enthalpy of component \( m \).

The components of the diffusion flux vector \( \vec{J}_m \) are computed with the formulas:

\[
J_{mc} = \rho D_{m,uu} \frac{\partial Y_m}{\partial z}, \quad J_{mr} = \rho D_{m,ur} \frac{\partial Y_m}{\partial r}, \tag{4}
\]

where \( D_{m,uu} \) – coefficient of \( m \)-component diffusion, average in mixture.

The components of the heat flow vector \( \vec{q} \) are computed with the formulas:

\[
q_z = \lambda \frac{\partial T}{\partial z} + \sum_{m=1}^{M} h_m \rho D_{m,uu} \frac{\partial Y_m}{\partial z}, \quad q_r = \lambda \frac{\partial T}{\partial r} + \sum_{m=1}^{M} h_m \rho D_{m,ur} \frac{\partial Y_m}{\partial r}, \tag{5}
\]
where $\lambda$ – thermal conductivity of the mixture.

The components of the viscous tensor $\tau$ are given by:

$$
\tau_{zz} = \mu \left( \frac{2}{3} \frac{\partial u_z}{\partial z} + \frac{1 \partial u_r}{r \partial r} \right),
\tau_{r} = \mu \left( \frac{2}{3} \frac{\partial u_r}{\partial z} + \frac{1 \partial u_z}{r \partial r} \right),
\tau_{rr} = \mu \left( \frac{\partial u_r}{\partial r} + \frac{u_r}{r} \right),
$$

(6)

where $\mu$ – coefficient of dynamic viscosity of the mixture.

A detailed description and expressions for the coefficients of multicomponent mixtures finding are given in [8].

The system of equations (1) must be supplemented with initial and boundary conditions, those setting is determined with the specific task to be solved. The conditions of the gas stream inflow and outflow, wall no slip are set for calculations in planar and cylindrical geometry. The symmetry conditions are set on the axis of symmetry for axisymmetric geometry:

$$
\frac{\partial \rho Y_m}{\partial r} = 0, \frac{\partial u_z}{\partial r} = 0, u_r = 0, \frac{\partial \pi}{\partial r} = 0, \frac{\partial ph}{\partial r} = 0.
$$

(7)

3. Computational algorithm

To develop the discrete model, we introduce a grid of rectangles uniform in each direction:

$$
\Omega_n = \left\{ z_i = ih_z, r_j = jh_r, i = 1, N_z, j = 1, N_r, h_z = L_z, h_r = L_r \right\},
$$

(8)

where $L_{z(r)}$ – size of calculating domain along the axes $\{z, r\}$. All gas dynamic parameters and component concentrations of the mixture are considered at cell centers.

According to the approach of [4], the system of equations (1) is splitted into physical processes on chemical kinetics, the Navier-Stokes equations without regard to contribution of the perturbational pressure, the Poisson equation for the perturbational pressure and the velocity vector correcting.

The following system of equations is used to account for the chemical reaction contribution to the process under investigation. The second equation shows the temperature changing during the chemical reaction.

$$
\begin{align*}
\frac{\partial \rho Y_m}{\partial t} &= R_m, \\
\rho C_p \frac{\partial T}{\partial t} &= - \sum_m R_m h_m, m = 1, ..., M.
\end{align*}
$$

(9)

An economical second-order-accurate method is used to solve the equations of chemical kinetics, which has been suggested and analyzed in [9].

The following conservative differential-difference scheme is constructed to solve the Navier-Stokes equations without regard to contribution of the perturbational pressure:

$$
\frac{U_{ij}^{n+1} - U_{ij}^n}{\Delta t} + \frac{\tilde{F}_{ij}^{(1)}(U_{i+1/2j}, U_{i-1/2j})}{h_z} + \frac{\tilde{F}_{ij}^{(2)}(U_{ij+1/2}, U_{ij-1/2})}{h_z} - \frac{\tilde{H}_{ij}^{(1)}(U_{i+1/2j}, U_{i-1/2j})}{r} - \frac{\tilde{H}_{ij}^{(2)}(U_{ij+1/2}, U_{ij-1/2})}{r} = 0.
$$

(10)

Here $\tilde{F}_{ij}^{(1)} = \tilde{F}^{(1)}(U_{i+1/2j}, U_{i-1/2j})$, $\tilde{F}_{ij}^{(2)} = \tilde{F}^{(2)}(U_{ij+1/2}, U_{ij-1/2})$ – convective flows, that are computed with the Lax-Friedriks-Rusanov’s scheme [10,11], $U_{i+1/2j}$, $U_{i-1/2j}$ – vector of variates values $U$ on the left and on the right from the boundary between $i$ and $i+1$ units. Discrete flows at
the boundaries between units $\tilde{H}_{i+1/2}^{(1)}$, $\tilde{H}_{i+1/2}^{(2)}$, responsible for heat transfer and diffusive transfer, are evaluated according to the scheme with central difference.

Since we haven’t taken into account influence of the perturbational pressure $\pi$, which changes the equations of conservation of momentum to solve system (10), then for velocity vector correcting the following expression is used:

$$\ddot{u}^{n+1} = \ddot{u}^{*} - \frac{\Delta t}{\rho^{u}} \nabla \pi^{n+1}.$$  

(11)

Here $\ddot{u}^{*}$ is velocity vector predictor obtained from the solution of the system (10). Let us take divergence from both members (11) and get the equation for definition $\pi^{n+1}$:

$$\nabla \cdot \frac{1}{\rho^{u}} \nabla \pi^{n+1} = \frac{1}{\Delta t} \left( \nabla \cdot \ddot{u}^{*} - \nabla \cdot \ddot{u}^{n+1} \right).$$  

(12)

To simplify the calculations, since density changing in the process under investigation is not significant, we can convert (12) to:

$$\nabla \cdot \nabla \pi^{n+1} = G_{ij}, G_{ij} = \frac{\rho^{u}}{\Delta t} \left( \nabla \cdot \ddot{u}^{*} - \nabla \cdot \ddot{u}^{n+1} \right).$$  

(13)

Using the integro-interpolation method the equation (13) is approximated in the following way:

$$\frac{\pi_{i+1j} - 2\pi_{ij} + \pi_{i-1j}}{h_{i}^{2}} + \frac{\pi_{i+1j} - 2\pi_{ij} + \pi_{i-1j}}{h_{j}^{2}} + \frac{1}{r_{ij}} \frac{\pi_{i+1j} - \pi_{ij}}{2h_{j}} = G_{ij}. $$  

(14)

To find the variable $G_{ij}$ in (14) it’s necessary to know the value of the velocity vector divergence at the next step according to time $\nabla \cdot \ddot{u}^{n+1}$. It is obtained by the equation of continuity in a non-divergent form converting, the energy conservation equation written relatively the temperature, and the state equation for the gases mixture [6]:

$$\nabla \cdot \ddot{u} = \frac{1}{\rho C_{p} T} \left( \nabla \cdot \lambda \nabla T + \sum_{m} \rho D_{m,m} \nabla Y_{m} \nabla h_{m} \right) + \frac{1}{\rho} \sum_{m} \frac{M_{m}}{M_{mm}} \left( \nabla \cdot \rho D_{m,m} \nabla Y_{m} \right) + \frac{1}{\rho} \sum_{m} \left( \frac{M_{m}}{M_{mm}} - \frac{h_{m}}{C_{p} T} \right) R_{m}. $$  

(15)

All variables of expression (15) are obtained after system (10) solving at the next step according to time. Using Jacobi’s iterative method, it’s possible to find the pressure dynamic component $\pi$, and then to correct the velocity vector using expression (11).

The code for problems with axisymmetric geometry has been developed and a number of computational experiments have been conducted using the constructed computational algorithm.

4. Computational experiment
Ethane pyrolysis is considered in cylindrical reactors of 220 mm length, of 20 mm diameter in a plane pipe of the same gap. Initially the pipes have been filled with methane. The initial mixture of 870.15 K temperature consists of ethane and methane (40% C2H6, 60% CH4). It enters the pipes on the left at a flow rate of 40 l/h. The outlet pressure on the right is 101325 Pa, and the inlet pressure of the gas mixture has been calculated with regard to flow rate and outlet pressure according to the Poiseuille equation. A 15-stage radical kinetic scheme [12] has been used as ethane pyrolysis reaction scheme which had been showed sufficient completeness of the process under investigation in [4,8]. The calculations have been made for three different wall temperatures with other fixed initial conditions.
Figure 1. Distribution of temperature (on the left) and ethane (on the right) along the axis, T=915 K. Solid line - planar pipe, dotted line - cylindrical pipe.

Figure 2. Distribution of ethane along the axis, T=965 K (on the left), T=1015 K (on the right). Solid line - planar pipe, dotted line - cylindrical pipe.

The gas mixture heating has been more intensive in the cylindrical pipe despite significant energy absorption close to the walls in the endothermic reaction, as shown in Figure 1, where one can see the temperature and ethane distributions along the reactor’s axis. The gas heating inside the reactor is influenced by the coefficient increasing thermal conductivity of the mixture due to the actively diffusing molecular hydrogen in the reactor. Its thermal conductivity is several times higher than that of other components of the mixture. Heat transfer increasing from the walls inside the reactor using a cylindrical pipe (Figure 1, Figure 2) has a significant effect on ethane conversion in the initial pipe sections at moderate temperatures. However, ethane conversion tends to be complete before reaching the central part of the reactor at 1015 K temperature. There is accumulation and diffusion equalization of the reaction products on the right side of it. Consequently, the geometric factor influence ethane conversion is weakened in case of the reactor’s walls high temperature and big sized reactors.

Conclusions
In the present work a numerical algorithm for solving the multi-component Navier-Stokes equations in the low Mach number limit for axisymmetric problems has been developed and implemented. Through the interplay of the developed code and the code for problems in the planar pipes has made it possible to identify the geometric factor’s role when a large number of nonlinear physicochemical processes have been at work. There has been obtained a picture showing the flow of the gas mixture in a volumetric cylindrical pipe. The computational experiments have resulted in coinciding the flow pattern in the planar and cylindrical geometries, but the pyrolysis product yields and the reacting gas conversion are different. This fact indicates the determining influence of molecular hydrogen diffusion, that appears in radical chain reactions, heat transfer in the mixture and kinetics of chemical transformations on the temperature distribution, density, diffusion and ethane pyrolysis product yields.
The implemented algorithm is suggested using in the technological processes occurring in the cylindrical pipes research, when the chemical reactions occur under the influence of the reactor’s walls external heating. In future it is planned to expand it for calculating reaction gas flows at laser radiation absorbed by the environment, which is accompanied by a sharp change of the gas-dynamic characteristics of the flow.

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