A non-stationary model for catalytic converters with cylindrical geometry

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Abstract

We prove some existence and uniqueness results and some qualitative properties for the solution of a system modelling the catalytic conversion in a cylinder. This model couples parabolic partial differential equations posed in a cylindrical domain and on its boundary.

1 Introduction

A gas containing $N-1$ different chemical species is flowing through a cylindrical passage with a parabolic speed profile. Chemical species are diffusing in the cylinder and are reacting only on the boundary of the cylinder.

We investigate the existence, uniqueness and qualitative properties of a non-stationary system of partial differential equations describing the evolution of the concentrations of the $N-1$ chemical species and of the temperature, both with respect to the time variable and along the cylinder. This model of catalytic converters starts with the contribution of Ryan, Becke and Zygourakis [5]. However, we have added an axial diffusion term on the boundary in the present model, see [1] for a more detailed description of this model.

Due to its internal symetry, the cylinder may be reduced to the domain $\Omega = [0,1[\times]0,1[$, the boundary of which is $\Sigma = \{1\} \times]0,1[$. The concentrations (resp. the temperature) inside the cylinder Ω are named C_{if}, $i = 1, ..., N-1$ (resp. C_{Nf}). The concentrations (resp. the temperature) on the boundary Σ are named C_{is} (resp. C_{Ns}). The problem is written in a normalized way as:

$$\left\{ \begin{array}{rcl} (1-r^2)\dfrac{\partial C_{if}}{\partial z}(r,z,t) & = & \dfrac{\beta_{if}}{r}\dfrac{\partial}{\partial r}\left(r\dfrac{\partial C_{if}}{\partial r}\right)(r,z,t), \\ \dfrac{\partial C_{is}}{\partial t}(z,t) & = & -\gamma_{is}\dfrac{\partial C_{if}}{\partial r}(1,z,t) + \delta_i \mathbf{r}_i\left(C_{1s}^+, \dots, C_{Ns}^+\right)(z,t) \\ & & +\theta_{is}\dfrac{\partial^2 C_{is}}{\partial z^2}(z,t), \end{array} \right. \quad (1)$$

for $i \in \{1, \dots, N\}$ and with $C_{is}^{+}(z,t) = \sup(C_{is}, 0)$. The initial and boundary conditions are:

$$\left\{ \begin{array}{rclrcl} C_{if}(r,0,t) & = & C_{i0}(r), & \dfrac{\partial C_{if}}{\partial r}(0,z,t) & = & 0, \\ C_{if}(1,z,t) & = & C_{is}(z,t), & C_{is}(z,0) & = & C_{is0}(z), \\ \theta_{is}\dfrac{\partial C_{is}}{\partial z}(1,t) & = & 0, & \theta_{is}\dfrac{\partial C_{is}}{\partial z}(0,t) & = & 0. \end{array} \right. \tag{2}$$

The functions $\mathbf{r}_i$, $i \in \{1, \dots, N\}$, are supposed to be Lipschitz continuous:

$$\left|\mathbf{r}_i\left(C_{1s}^1, \dots, C_{Ns}^1\right) - \mathbf{r}_i\left(C_{1s}^2, \dots, C_{Ns}^2\right)\right| \leq k_i \sum_{h=1}^{N} \left|C_{hs}^1 - C_{hs}^2\right|$$

and verify the following hypotheses:

(H1) $\forall x \in (\mathbb{R}^+)^N : \mathbf{r}_i(x_1, \dots, x_N) \geq 0$.

(H2) If at least one of the x_i, $1 \leq i \leq N$, is equal to 0, then $\mathbf{r}_i(x_1, \dots, 0, \dots, x_N) = 0$.

(H3) For every $(x,y) \in (\mathbb{R}^+)^N \times (\mathbb{R}^+)^N$:

$$-\sum_{i=1}^{N} \delta_i \frac{\beta_{if}}{\gamma_{is}} \left(\mathbf{r}_i(x_1, \dots, x_N) - \mathbf{r}_i(y_1, \dots, y_N)\right)(x_i - y_i) \geq 0.$$

Remark 1.1 *We observe that:*

1. *Hypotheses (H2) and (H3) imply:*

$$\forall x \in \left(\mathbb{R}^+\right)^N : -\sum_{i=1}^{N} \delta_i \frac{\beta_{if}}{\gamma_{is}} \mathbf{r}_i(x_1, \dots, x_N)\, x_i \geq 0.$$

2. *We have:*

$$C_{if}(1,z,t) = C_{is}(z,t) \;\; ; C_{if}(r,0,t) = C_{i0}(r) \;\; ; C_{is}(z,0) = C_{is0}(z).$$

In order to ensure the continuity of the concentrations in Ω and on the boundary Σ at $z = 0$, we must have at $t = 0$, $C_{if}(1,0,0) = C_{is}(0,0)$, which implies : $C_{i0}(1) = C_{is0}(0)$; $C_{is}(0,t) = C_{i0}(1)$.

2 Existence of the solution

We establish the existence of the solution using the diagram:

$$\begin{array}{ccccc} & & \Gamma & & \\ & \tilde{C}_{if} & \rightarrow & C_{if} \\ \Phi & \downarrow & \nearrow & & \\ & & \Psi & & \\ & C_{is} & & & \end{array}$$

Indeed, the proof of the existence is decomposed in two steps:

1. Existence of a solution in Ω (given the solution on the boundary Σ) and existence of a solution on the boundary Σ (given the solution in Ω);
2. $\Gamma = \Psi \circ \Phi$ is a contraction in some appropriate functional space.

2.1 Preliminary results

We have the following continuous embeddings (see [3, p. 103]):

$$L^2(\Omega) \underset{\rightarrow}{\subset} L^2_r(\Omega) \underset{\rightarrow}{\subset} L^2_{r(1-r^2)}(\Omega) \ ; W^{1,2}(\Omega) \underset{\rightarrow}{\subset} W^{1,2}_r(\Omega) ; W^{1,2}(\Omega) \underset{\rightarrow}{\subset} W^{1,2}_{r(1-r^2)}(\Omega) .$$

Because $\frac{\partial C_{if}}{\partial r}(1,z,t) = \frac{1}{\beta_{if}} \int_0^1 \frac{\partial C_{if}}{\partial z} r(1-r^2)\, dr$, we can write the problem as:

$$\left\{ \begin{array}{rcl} (1-r^2)\dfrac{\partial C_{if}}{\partial z} & = & \beta_{if} \dfrac{1}{r}\dfrac{\partial}{\partial r}\left(r \dfrac{\partial C_{if}}{\partial r}\right), \\ \dfrac{\partial C_{is}}{\partial t} - \theta_{is}\dfrac{\partial^2 C_{is}}{\partial z^2} & = & -\dfrac{\gamma_{is}}{\beta_{if}} \displaystyle\int_0^1 \dfrac{\partial C_{if}}{\partial z} r(1-r^2)\, dr + \delta_i \mathbf{r}_i(C^+_{1s}, \ldots, C^+_{Ns}), \end{array} \right. \tag{3}$$

with $i \in \{1, \ldots, N\}$, and the initial or boundary conditions:

$$\left\{ \begin{array}{rclrcl} C_{if}(r,0,t) & = & C_{i0}(r), & \dfrac{\partial C_{if}}{\partial r}(0,z,t) & = & 0, \\ C_{if}(1,z,t) & = & C_{is}(z,t), & C_{is}(z,0) & = & C_{is0}(z), \\ \theta_{is}\dfrac{\partial C_{is}}{\partial z}(1,t) & = & 0, & \theta_{is}\dfrac{\partial C_{is}}{\partial z}(0,t) & = & 0. \end{array} \right.$$

2.2 Existence in the cylinder

Assuming that the C_{is} are known on the boundary and performing a change of function in order to have homogeneous boundary conditions at $r = 1$, we obtain the following problem in which we omitted the time variable t:

$$\left\{ \begin{array}{rcl} (1-r^2)\dfrac{\partial u_f}{\partial z} - \beta_f \dfrac{1}{r}\dfrac{\partial}{\partial r}\left(r\dfrac{\partial u_f}{\partial r}\right) & = & -\dfrac{\partial u_s}{\partial z}, \\ u_f(r,0) & = & u_0(r), \\ u_f(1,z) & = & 0, \\ \dfrac{\partial u_f}{\partial r}(0,z) & = & 0, \end{array} \right. \tag{4}$$

with $i \in \{1, \ldots, N\}$ and

$$\begin{array}{rclrcl} w_f & = & {}^t(C_{1f}, \ldots, C_{Nf}), & u_s & = & {}^t(C_{1s}, \ldots, C_{Ns}), \\ u_f & = & w_f - u_s, & \beta_f & = & diag\left(\beta_{1f}, \cdots, \beta_{Nf}\right). \end{array}$$

We set :

$$\begin{array}{rcl} W & = & \left\{ u \in \left(L^2_r(0,1)\right)^N \mid \dfrac{\partial u}{\partial r} \in \left(L^2_r(0,1)\right)^N \right\}, \\ W_0 & = & \left\{ u \in \left(L^2_r(0,1)\right)^N \mid \dfrac{\partial u}{\partial r} \in \left(L^2_r(0,1)\right)^N, u(1) = 0 \right\} \end{array}$$

and let W_0' be the dual space of W_0.

Definition 2.1 *Assume that* $u_0 \in \left(L_r^2(0,1)\right)^N$ *and* $\frac{\partial u_s}{\partial z}(z) \in \left(L^2(0,1)\right)^N$. *A function* u_f *is a* ***weak solution*** *of (4) if and only if* $u_f \in L^2(0,1;W_0)$, $\frac{\partial u_f}{\partial z} \in L^2(0,1;W_0')$ *and if for every* $\varphi \in L^2(0,1;W_0)$, *we have:*

$$\int_0^1\int_0^1 \left\langle \frac{\partial u_f}{\partial z}, \varphi \right\rangle r(1-r^2)drdz + \int_0^1\int_0^1 \left\langle \beta_f r\frac{\partial u_f}{\partial r}, \frac{\partial \varphi}{\partial r} \right\rangle drdz$$
$$= -\int_0^1\int_0^1 \left\langle \frac{\partial u_s}{\partial z}, \varphi \right\rangle r(1-r^2)drdz.$$

Proposition 2.2 *Let* u_0 *and* $\frac{\partial u_s}{\partial z}$ *be as in the precedent definition. Then there exists at least one* ***weak solution*** *of (4). This solution verifies:*

$$\begin{aligned} \|u_f\|_{L^\infty\left(0,1;\left(L^2_{r(1-r^2)}(0,1)\right)^N\right)} &\leq C, \\ \int_0^1 \left\| \frac{\partial u_f}{\partial r}(z) \right\|^2_{\left(L_r^2(0,1)\right)^N} dz &\leq C, \\ \left\| \frac{\partial u_f}{\partial z} \right\|_{L^2\left(0,1;W_0'\right)} &\leq C, \end{aligned} \tag{5}$$

where C *is a positive constant which only depends on the data of the problem.*

Proof. We use a Galerkin approximation of u_f for which we establish the three above estimates, and pass to the limit in order to prove the Proposition, see [1] for the details. ∎

Proposition 2.3 *Under the conditions given in the Definition 2.1, the solution of (4) is such that:*

$$u_f \in L^2(0,1;W_0) \cap C\left(0,1;\left(L_r^2(0,1)\right)^N\right).$$

Proof. From the inclusion $W_0 \subset \left(L_r^2(0,1)\right)^N \subset W_0'$, and the fact that $u_f \in L^2(0,1;W_0)$, $\frac{\partial u_f}{\partial z} \in L^2(0,1;W_0')$, we deduce the result using Proposition 23.23 of [6, p. 422]. ∎

Remark 2.4 *In order to take into account the time variable* t, *all the above expressions* $v(.) \in H$ *have to be understood as* $v(.,t) \in L^2(0,T;H)$.

2.3 Existence on the boundary

Assuming that $\frac{\partial u_f}{\partial z}$ is known, we have:

$$\begin{cases} \dfrac{\partial u_s}{\partial t} - \theta_s \dfrac{\partial^2 u_s}{\partial z^2} &= \delta \mathbf{r}(u_s^+) - \Gamma_f \displaystyle\int_0^1 \frac{\partial u_f}{\partial z} r\left(1-r^2\right)dr, \\ u_s(z,0) &= u_{s0}(z), \\ \theta_s \dfrac{\partial u_s}{\partial z}(0,t) &= 0, \\ \theta_s \dfrac{\partial u_s}{\partial z}(1,t) &= 0, \end{cases} \tag{6}$$

with:

$$\begin{aligned} \mathbf{r} &= {}^{t}(\mathbf{r}_{1,} \ldots, \mathbf{r}_N), \qquad & \Gamma_f &= diag\left(\frac{\gamma_{1s}}{\beta_{1f}}, \cdots, \frac{\gamma_{Ns}}{\beta_{Nf}}\right), \\ \delta &= (\delta_1, \cdots, \delta_N), & \theta_s &= (\theta_{1s}, \cdots, \theta_{Ns}). \end{aligned}$$

Let

$$H^1(0,1) = \left\{ u \in \left(L^2(0,1)\right)^N \mid \frac{\partial u}{\partial z} \in \left(L^2(0,1)\right)^N \right\}$$

and let H^* be the dual space of H^1.

Definition 2.5 *Suppose that* $\frac{\partial u_f}{\partial z}$ *belongs to* $L^2\left(0,T; L^2\left(0,1; (L_r^2(0,1))^N\right)\right)$ *and that* u_{s0} *belongs to* $(L^2(0,1))^N$. *A function* u_s *is a* ***weak solution*** *of (6) if and only if* $u_s \in L^2(0,T; H^1(0,1))$, $\dfrac{\partial u_s}{\partial t} \in L^2(0,T; H^*(0,1))$, *satisfies* $u_s(z,0) = u_{s0}(z)$, *and if for all* $\varphi \in L^2(0,T; H^1(0,1))$, *we have:*

$$\begin{aligned} &\int_0^T \int_0^1 \left\langle \frac{\partial u_s}{\partial t}, \varphi \right\rangle dzdt + \int_0^T \int_0^1 \left\langle \theta_s \frac{\partial u_s}{\partial z}, \frac{\partial \varphi}{\partial z} \right\rangle dzdt \\ &\quad = \int_0^T \int_0^1 \langle \delta \mathbf{r}(u_s^+), \varphi \rangle \, dzdt - \int_0^T \int_0^1 \int_0^1 \left\langle \Gamma_f \frac{\partial u_f}{\partial z}, \varphi \right\rangle r\left(1-r^2\right) drdzdt. \end{aligned}$$

Proposition 2.6 *Let* u_f *and* u_{s0} *as in the preceding definition. Then there exists at least one* ***weak solution*** u_s *of (6).*

Proof. We prove the existence of a solution for the linearized weak formulation of the problem due to Theorem 2.2 of [4, p. 286] and then we use some fixed point argument. ■

Proposition 2.7 *Under the conditions given in the Definition 2.5, the solution of (6) is such that:*

$$u_s \in L^2\left(0,T; H^1(0,1)\right) \cap C\left(0,T; \left(L^2(0,1)\right)^N\right).$$

Proof. From the embeddings $H^1(0,1) \subset (L^2(0,1))^N \subset H^*(0,1)$ and the fact that $u_s \in L^2(0,T; H^1(0,1))$, $\frac{\partial u_s}{\partial t} \in L^2(0,T; H^*(0,1))$, we deduce the result using Proposition 23.23 of [6, p. 422]. ■

2.3.1 $\Gamma = \Psi \circ \Phi$ is a contraction

Consider the mapping $\Gamma : \tilde{U}_f \mapsto U_s \to U_f$, and let $\mu = \sup_i \left(\gamma_{is}/\beta_{if}\right)^{1/2} / \inf_i \theta_{is}$.

Proposition 2.8 *1. The mapping* $\Phi : \tilde{U}_f \to U_s$ *is such that if we suppose* $\mu\alpha^2 < 4$, *then:*

$$\int_0^\tau \int_0^1 \left\| \frac{\partial U_s}{\partial z} \right\|^2 dzdt \leq \frac{4\mu}{\alpha^2(4-\mu\alpha^2)} \sup_{s\in[0,1]} \int_0^\tau \int_0^1 \left\| \tilde{U}_f \right\|^2 (r,s,t) r\left(1-r^2\right) drdt. \quad (7)$$

2. The mapping $\Psi : U_s \to U_f$ is such that:

$$\sup_{s\in[0,1]} \int_0^T \int_0^1 \|U_f\|^2 (r,s,t) r(1-r^2) dr dt \leq \frac{e}{4} \int_0^T \int_0^1 \left\| \frac{\partial U_s}{\partial z} \right\|^2 dz dt. \tag{8}$$

Let :

$$W_f = L^2(0,T; L^2(0,1;W_0)) \cap L^2\left(0,T; C\left(0,1; (L^2(0,1))^N\right)\right),$$
$$W_p = L^2(0,T; H^1(0,1)) \cap C\left(0,T; (L^2(0,1))^N\right),$$

and

$$W_g = \{(u,v) \in W_f \times W_p \mid u(1,z,t) = v(z,t)\}.$$

Theorem 2.9 *Under the hypothesis $(H3)$ the problem (1) admits a solution if:*

$$\frac{\sqrt{e}}{2} \sup_i \left(\frac{\gamma_{is}}{\beta_{if}}\right)^{1/2} < \inf_i \theta_{is}.$$

This solution belongs to W_g.

Proof. Using (7) and (8), we obtain:

$$\|U_f\|_f \leq \sqrt{\frac{e\mu}{\alpha^2(4-\mu\alpha^2)}} \left\|\tilde{U}_f\right\|_f \; ; \; \|U_f\|_f^2 := \sup_{s\in[0,1]} \int_0^T \int_0^1 \|U_f\|^2 (r,s,t) r(1-r^2) dr dt.$$

The α which minimizes the Lipschitz constant is given by $\alpha^2 = 2/\mu$, which leads to $\|U_f\|_f \leq \mu\sqrt{e}\left\|\tilde{U}_f\right\|_f/2$. This proves that $\Gamma : \tilde{U}_f \mapsto U_f$ is a contraction if and only if $\mu < 2/\sqrt{e}$. ∎

3 Uniqueness of the solution

Theorem 3.1 *Assuming that the solution of (1) is smooth enough, the system (1) has at most one solution.*

Proof. We suppose the existence of two couples of solutions $\left(C_{if}^1, C_{is}^1\right)_{i=1,\ldots,N}$ and $\left(C_{if}^2, C_{is}^2\right)_{i=1,\ldots,N}$ of (1), and define: $W_{if} = C_{if}^1 - C_{if}^2$; $W_{is} = C_{is}^1 - C_{is}^2$. We multiply the i-th equation of the system verified by W_{if} by rW_{if}, integrate on $[0,1]\times[0,1]\times[0,T]$ and use the equation verified by W_{is} on the boundary. Thanks to $(H3)$, one can deduce that W_{if} is equal to zero in the cylinder because W_{if} is equal to zero at the inlet $(z=0)$ and at the outlet of the cylinder $(z=1)$ and its partial derivative with respect to r is equal to zero too, which implies, thanks to the equation verified by W_{is} on the boundary, that the partial derivative of W_{if} with respect to z is equal to zero in the cylinder. We also deduce that $\frac{\partial W_{is}}{\partial z}$ is equal to zero on the boundary. W_{is} is equal to zero at time T and at time 0 because of the initial conditions. Because of T is arbitrarily chosen, W_{is} is equal to zero on the boundary Σ at any time. ∎

4 Qualitative properties of the solution

4.1 Nonnegativity of the solution

Proposition 4.1 *For almost every* (r,z,t) *in* $]0,1[\times]0,1[\times]0,T[$, *and for* $i\in\{1,\ldots,N\}$, *we have :* $0\leq C_{if}(r,z,t)$, $0\leq C_{is}(z,t)$.

Proof. This is obtained multiplying the equations of (1) by the non-negative parts of C_{if} or C_{is}, respectively. ■

4.2 Upper and lower bounds of the concentrations

Proposition 4.2

1. *Let* $\delta_i=-1$. *For almost every* (r,z,t) *in* $]0,1[\times]0,1[\times]0,T[$, *we have :* $0\leq C_{if}(r,z,t)\leq A_{i0}$; $0\leq C_{is}(z,t)\leq A_{i0}$, *with:*

$$A_{i0}=\max\left(\sup_{r\in[0,1]}C_{i0}(r)\,,\ \sup_{z\in[0,1]}C_{is0}(z)\right).$$

2. *Let* $\delta_i=1$. *For almost every* (r,z,t) *in* $]0,1[\times]0,1[\times]0,T[$, *we have:* $a_{i0}\leq C_{if}(r,z,t)$, $a_{i0}\leq C_{is}(z,t)$, *with:*

$$a_{i0}=\min\left(\inf_{r\in[0,1]}C_{i0}(r),\ \inf_{z\in[0,1]}C_{is0}(z)\right).$$

3. *Let* $\delta_i=1$. *We have for almost every* (r,z,t) *in* $]0,1]\times]0,1[\times]0,T[$: $C_{if}(r,z,t)\leq a_{i0}e^{\lambda t}$, $C_{is}(z,t)\leq a_{i0}e^{\lambda t}$, *with:*

$$a_{i0}=\max\left(\sup_{r\in[0,1]}C_{i0}(r),\ \sup_{z\in[0,1]}C_{is0}(z)\right)\ ,\ \lambda=\sup_i k_i,$$

   k_i *being the Lipschitz constant of the function* $\mathbf{r}_i$.

4. *Let* $\delta_i=1$. *There exist two positive constants* a *and* b *such that we have for every* l *in* $]0,1[$ *and for every* $T>0$:

$$\begin{aligned}\int_0^T\int_0^1(C_{if})^2(r,l,t)\,r\,(1-r^2)\,drdt&\leq aT+b,\\ \int_0^1(C_{is})^2(z,T)\,dz&\leq aT+b.\end{aligned}$$

Proof. The verification of these qualitative properties of the solution is essentially obtained multiplying the equations of (1) by the appropriate non-negative or non-positive parts of the corresponding test-functions. ■

5 Numerical simulation for the reaction $CO + O_2 \to CO_2$

In the fluid, we use the following discretization method:

- If i is different of N (chemical species), we directly solve $(1)_1$ using the method of finite differences or that based on finite elements. This requires that the equation with $i = N$ has already been solved in order to put the appropriate values of u_N.

- If i is equal to N (temperature) we evaluate the coefficients at the step before. This possibly requires the use of some fixed point argument.

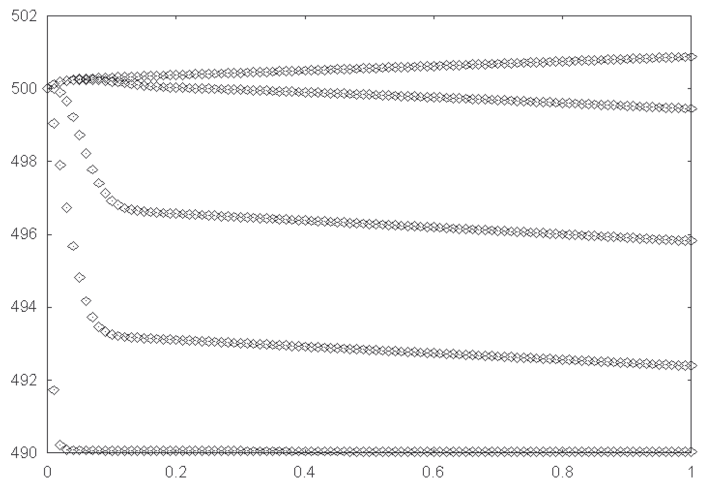
On the boundary, we still use some finite differences method or some finite element method (see [2] for the details).

Let us take the following initial and boundary conditions:

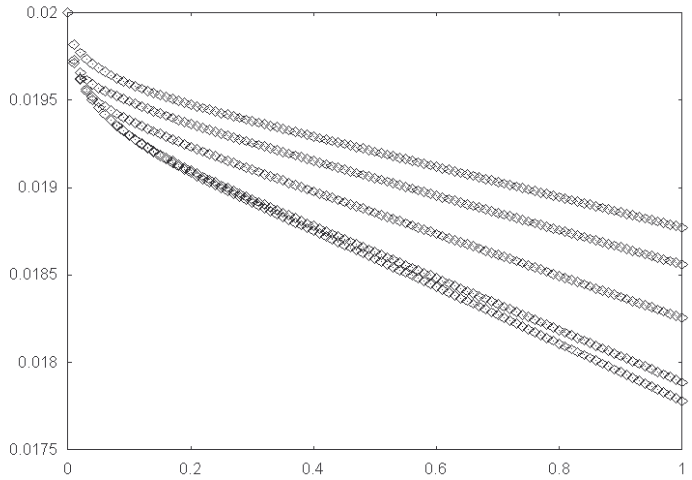
in the cylinder:
$$\begin{aligned} CO\,(r,0,t) &= 0.02 \\ O_2\,(r,0,t) &= 0.05 \\ CO_2\,(r,0,t) &= 0 \\ T\,(r,0,t) &= 500, \end{aligned}$$
on the boundary:
$$\begin{aligned} CO\,(z,0) &= 0.02 \\ O_2\,(z,0) &= 0.05 \\ CO_2\,(z,0) &= 0 \\ T\,(z,0) &= 490, \end{aligned}$$

We have the following graphs at $t = 0.3s$, $12s$, $24s$, $36s$ and $60s$. We observe that the CO and O_2 concentrations are decreasing and that the temperature and the CO_2 concentration are increasing:

- Temperature

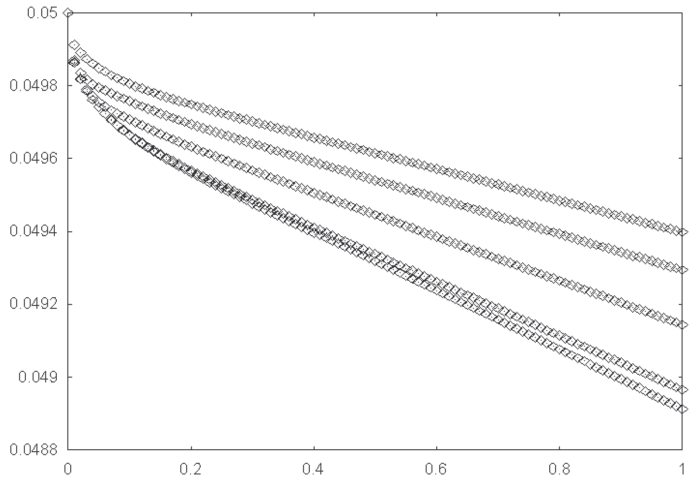


- CO



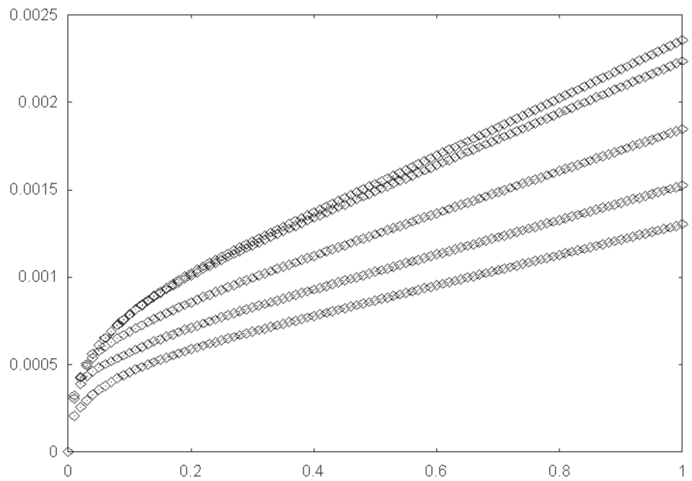
0.02
0.0195
0.019
0.0185
0.018
0.0175
0
0.2
0.4
0.6
0.8
1

- O_2



0.05
0.0498
0.0496
0.0494
0.0492
0.049
0.0488
0
0.2
0.4
0.6
0.8
1

- CO_2



0.0025
0.002
0.0015
0.001
0.0005
0
0
0.2
0.4
0.6
0.8
1

Remark 5.1 *The reaction ends after 54s with the following values at the outlet of the cylinder* $(z = 1)$*:*

$$\begin{aligned} CO\,(1,54) &= 0.017777, \\ O_2\,(1,54) &= 0.048912, \\ CO_2\,(1,54) &= 0.002355, \\ T\,(1,54) &= 500.873497. \end{aligned}$$

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