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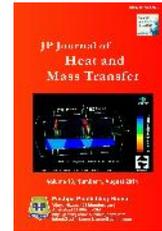
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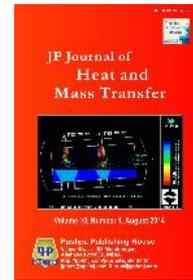
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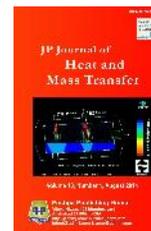
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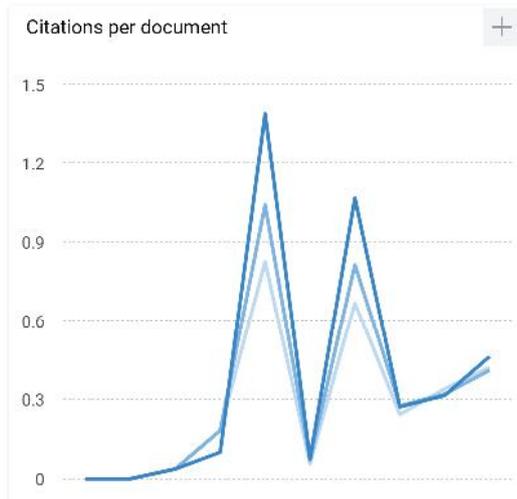
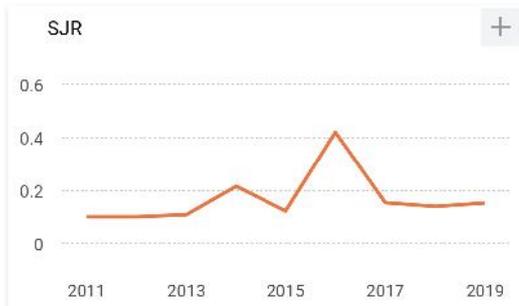
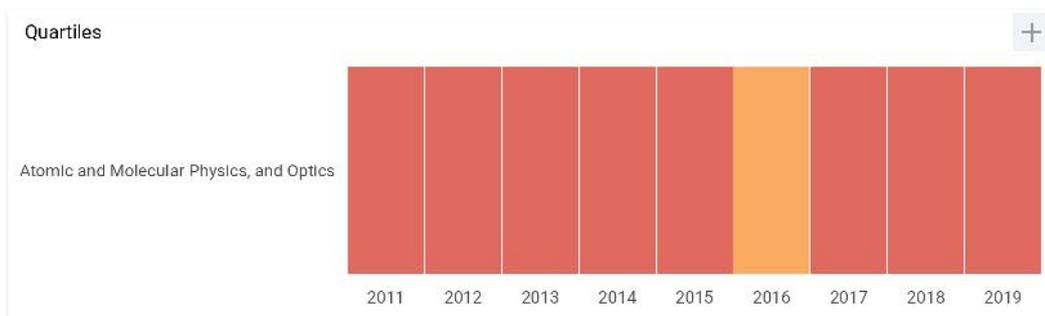
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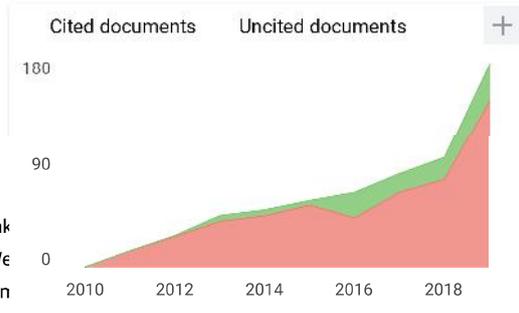
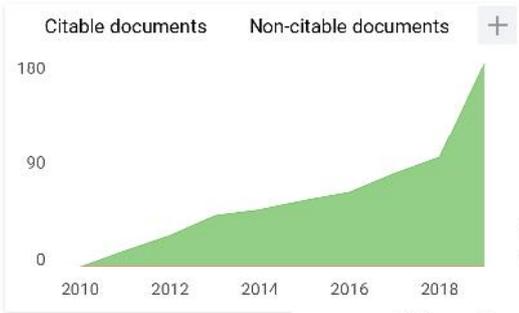
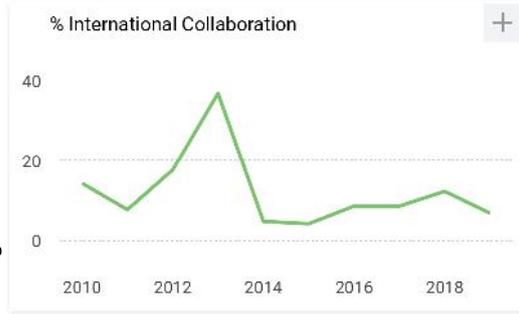
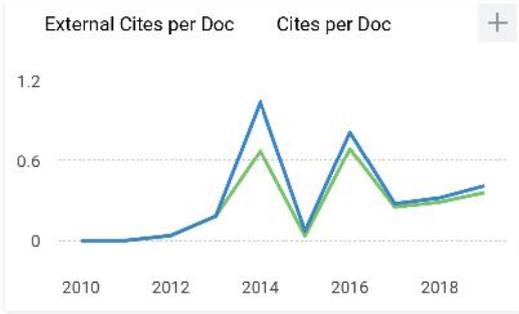
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PARAMETER SENSITIVITY ANALYSIS ON MATHEMATICAL MODEL OF METHANE OXIDATION USING REVERSE FLOW REACTOR WITH PERIODICALLY PERTURBED FEED GAS

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Abstract

In this paper, we investigate the effect of operation parameters to dynamic of dependent variables for catalytic oxidation reaction using reverse flow reactor (RFR). Here, we consider a 1-D pseudo-homogeneous model for RFR with cooling through the wall and periodically perturbed feed gas. By using finite difference method to the model, we construct numerical schemes. The simulations are conducted for various values of period of gas feeding, switching time, cooling capacity and superficial velocity.

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Keywords and phrases: switching time, pseudo-homogeneous model, periodic feed gas, cooling capacity.

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1. Introduction

Methane is the second largest greenhouse gas whose global warming potential reaches about 20 times greater than carbon dioxide and it becomes a wasted energy resource if not used. Methane includes the gas whose low and unstable concentration, is about 1 vol%, and constitutes the most arduous hydrocarbons to react whose adiabatic combustion temperature is about 1900°C in the traditional combustion mode [1]. Therefore, auto-thermal operation is hard to achieve a valuable without pre-heating the feed. Thus, conversion of methane to carbon dioxide will give two advantages: reduction of global warming effect and utilization of the heat output from the reaction to pre-heat the feed. On the other hand, a catalyst is required to accelerate the conversion and to carry out stable combustion for low concentrations.

One method used in methane combustion is catalytic reverse flow reactor (RFR). It was first proposed by Frank-Kamenetskii [2] and was reviewed by Matros and Bunimovich [3]. RFR is a packed-bed reactor in which the flow direction is periodically reversed to trap a hot zone within the reactor. If we compare RFR with a direct flow adiabatic reactor, then reactions using RFR may be run and proceed at lower temperature and higher conversions.

There are some methods in previous research to investigate dynamics of feed gas in RFR. In [4-17], there are three methods to investigate this problem. They are experimental, analytical and numerical or computational approaches. The last two methods are usually considered mathematical models that describe the dynamics of feed gas dynamics in RFR.

Various models were proposed to describe the dynamic behavior of the RFR. For example: Khinast et al. [4] and Salomons et al. [5] have used a one-dimensional pseudo-homogeneous model, while [6] proposed a one-dimensional heterogeneous model, and [7] used a two-dimensional

heterogeneous model. In those models, temperature and concentration of feed gas were assumed to be constant along time.

In [11, 12], the authors proposed an analytical approach to solve singular perturbation problem that derived from 1-D pseudo-homogeneous model at steady state condition under some assumptions. By using same model but at unsteady state condition, an analytical solution using homotopy perturbation method was studied [16]. Furthermore, Wibowo et al. [15] have reported solution unique existence of mathematical model of reverse flow reactor especially in the parabolic and hyperbolic equations.

Meanwhile studies of feed gas dynamics in RFR through numerical simulation approaches were reported in [8, 10, 14]. Budhi et al. have investigated RFR stability for constant and oscillating feed concentration [10]. With the same topic, Nuryaman et al. [14] have studied the impact of changes in operating parameter values to RFR dynamics for fixed feed gas temperature with periodic concentration like square-wave function.

In this paper, we explore the previous research in [14] by considering temperature is constant and concentration of feed gas which is periodically perturbed like trigonometry function. Here we consider one-dimensional pseudo-homogeneous model that describes cooled RFR behavior. By numerical simulation approach, we analyze the parameter sensitivity to RFR stability.

Here, we organize this paper as follows: A mathematical model for the dynamic of methane temperature and concentration and numerical scheme are described in Section 2. Next, in Section 3, numerical simulations are presented to describe the dynamic behavior of feed gas in reverse flow reactor. In the last section, conclusions are presented.

2. Mathematical Modeling and Numerical Scheme

In mathematical modeling of the RFR, we adopt 1-D pseudo-homogeneous model for the cooling RFR consisting of one panel in Khinast et al. [4] as follows:

$$\begin{aligned} ((1 - \phi)(\rho c_p)_s + \phi(\rho c_p)_g) \frac{\partial T}{\partial t} = \lambda_{ax} \frac{\partial^2 T}{\partial z^2} - u(\rho c_p)_g \frac{\partial T}{\partial z} \\ + (-\Delta H)g(T)C - U_w a_w (T - T_c), \end{aligned} \quad (1)$$

$$\phi \frac{\partial C}{\partial t} = \phi D \frac{\partial^2 C}{\partial z^2} - u \frac{\partial C}{\partial z} - g(T)C, \quad (2)$$

where $g(T) = \frac{\eta k_\infty k_c a_v \exp(-E_a/RT)}{k_c a_v + \eta k_\infty \exp(-E_a/RT)}$ which corresponds to reaction rate.

In equations (1)-(2), $T = T(z, t)$ and $C = C(z, t)$, respectively, denote the temperature (K) and concentration (mol/L) of feed gas at position z and time t . Beside that, equations system (1)-(2) be equipped with boundary conditions for flow to the right as follows:

$$-\frac{\lambda_{ax}}{u(\rho c_p)_g} \frac{\partial T}{\partial z} = T_{in} - T, \quad -\frac{\phi D}{u} \frac{\partial C}{\partial z} = C_{in} - C, \quad \text{at } z = 0, \quad (3)$$

$$\frac{\partial T}{\partial z} = 0, \quad \frac{\partial C}{\partial z} = 0, \quad \text{at } z = l. \quad (4)$$

In equations (3) and (4), parameters T_{in} denote inlet temperature and C_{in} correspond to inlet gas concentration which enter into RFR from left/right side of reactor. In this article, we assume that its temperature is constant along time and its concentration is periodically perturbed function like $C_{in} = A + \varepsilon \sin \omega t$, where ω corresponding with period for every time interval before the flow direction is reversed.

To describe the dynamic behavior of methane in reverse flow reactor, we use numerical approach since the model is in nonlinear form. Many numerical methods can be used to simulate the behavior of methane which is modeled by equations (1)-(4). Examples include finite difference method, finite volume method, Runge-Kutta method and others. Here we used finite difference method with forward difference for derivative with respect to time variable and central difference for second derivative and backward

difference for first derivative with respect to space variable. So we get numerical schemes as below:

$$T_j^{n+1} = T_j^n + a_1(T_{j+1}^n - 2T_j^n + T_{j-1}^n) - b_1(T_j^n - T_{j-1}^n) + c_1g(T_j^n)C_j^n - d_1(T_j^n - T_C), \quad (5)$$

$$C_j^{n+1} = C_j^n + a_2(C_{j+1}^n - 2C_j^n + C_{j-1}^n) - b_2(C_j^n - C_{j-1}^n) - d_2g(T_j^n)C_j^n, \quad (6)$$

where

$$a_1 = \frac{\lambda_{ax}\Delta t}{((1-\phi)(\rho c_p)_s + \phi(\rho c_p)_g)\Delta z^2}, \quad a_2 = \frac{D\Delta t}{\Delta z^2},$$

$$b_1 = \frac{u(\rho c_p)_g \Delta t}{((1-\varepsilon)(\rho c_p)_s + \varepsilon(\rho c_p)_g)\Delta z}, \quad b_2 = \frac{u\Delta t}{\phi\Delta z},$$

$$c_1 = \frac{(-\Delta H)\Delta t}{((1-\phi)(\rho c_p)_s + \phi(\rho c_p)_g)},$$

$$d_1 = \frac{U_w a_w \Delta t}{((1-\phi)(\rho c_p)_s + \phi(\rho c_p)_g)}, \quad d_2 = \frac{\Delta t}{\phi}.$$

Here $T_j^n = T(z_j, t_n)$, $C_j^n = C(z_j, t_n)$, $\Delta z = l/N_z$ and $\Delta t = t/N_t$ with N_z and N_t denote the partition number of spatial and time coordinate, respectively. Parameter values that were used in our simulations are given in Table 1.

Table 1. Parameter values, [4]

Parameters	Value	Descriptions
ϕ	0.69	void fraction
$(\rho c_p)_s$	$904.55 \text{kJm}^3 \text{K}^{-1}$	volumetric heat capacity of solid
$(\rho c_p)_g$	$0.508 \text{kJm}^3 \text{K}^{-1}$	volumetric heat capacity of gas
λ_{ax}	$5.64 \text{kw m}^{-1} \text{K}^{-1}$	effective axial heat conductivity
u	0.3ms^{-1}	Superficial gas velocity
U_w	$0.1377 \text{kWm}^{-1} \text{K}^{-1}$	Heat transfer coefficient at reactor wall
a_w	272m^{-1}	Specific reactor wall surface
T_C	323K	Coolant temperature
$(-\Delta H)$	802000kJ/kmol	Heat of reaction
D	$6.52 \times 10^{-6} \text{m}^2 \text{s}^{-1}$	Gas diffusion coefficient
E_a	$9.629 \times 10^4 \text{kJ/kmol/K}$	Activation energy
R	8.3145kJ/kmol/K	Universal gas constant
k_∞	$7.34 \times 10^7 \text{s}^{-1}$	Frequency factor
k_c	0.115ms^{-1}	Mass-transfer coefficient
a_v	2000m^{-1}	Specific particle surface area
η	1	Effectiveness factor
l	0.26m	Reactor length
t_f	60s	Switching time

3. Results

In this section, we show some numerical simulations to analyze the sensitivity of operating parameters by means the effect of changing the parameter values on RFR stability or the continuation of the oxidation process.

Parameter sensitivity analysis of feed gas period

At first case for constant feed gas, the behaviors of two dependent variables T and C are shown in Figure 1. Furthermore, if we investigate up to six times reversed, we have Figure 2.

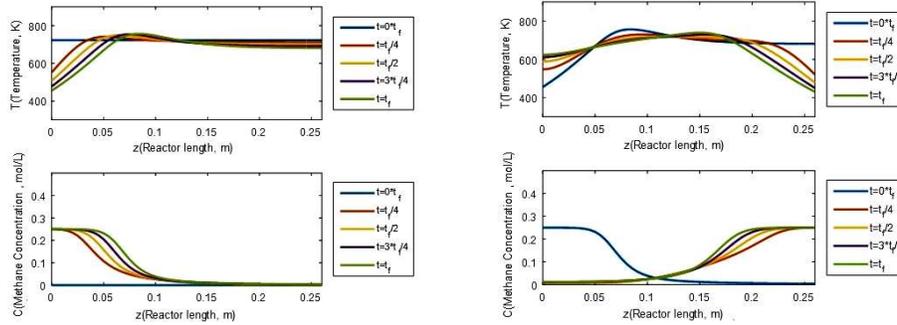


Figure 1. Temperature and concentration behavior of feed gas for various time values t with $A = 0.25$, $\varepsilon = 0.05$ and $\omega = 0$. Left figure, for flow from left to right and right figure after reversed.

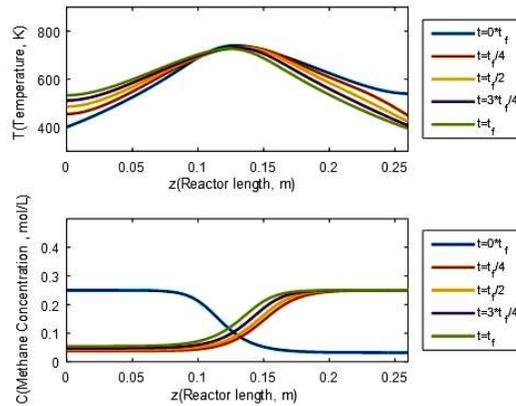


Figure 2. Temperature and concentration behavior of feed gas for various time values t with $A = 0.25$, $\varepsilon = 0.05$ and $\omega = 0$ after six times reversed.

From Figure 2, when the feed gas flow is reversed several times, the highest temperature is in the reactor center position. This is caused around the end of the reactor where the feed gas enters, the temperature is still low so that only a small amount of feed gas reacts. While around the end of the reactor where the remaining feed gas or the reaction product comes out, the temperature drops again due to the same thing as in the start up conditions.

In the next step, we simulate the impact of changing the period parameter ω which is shown in Figure 3 as follows.

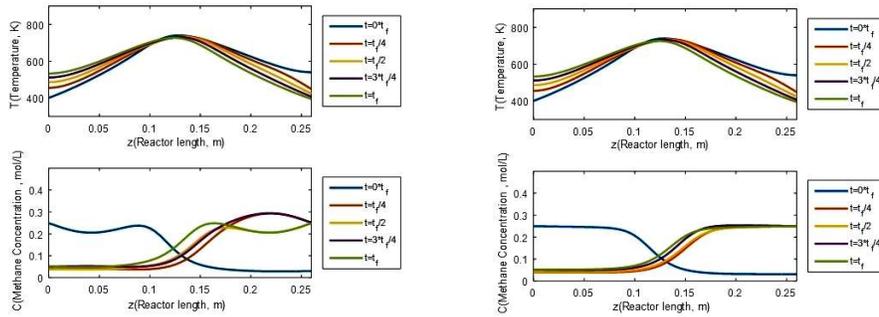


Figure 3. Temperature and concentration behavior of feed gas for various time values t with $A = 0.25$, $\varepsilon = 0.05$ after six times reversed. Left figure, for $\omega = 5\pi$ and right figure $\omega = \frac{\pi}{5}$.

In the case of feed gas being disturbed periodically, the parameters of the feed gas period affect the presence of waveforms in the plot of feed gas. The concentration of the feed gas which oscillates periodically has little effect on the temperature dynamics of the feed gas in the reactor. The existence of waveforms on the dynamics plot of feed gas concentration occurs for a large period. The similar conditions to constant feed gas concentrations occur when the feed gas period is small. In general, the size of the feed gas period does not make the reactor go out. This is indicated by the dynamic plot shape of the feed gas temperature in the reactor as shown in Figure 3.

Parameter sensitivity analysis of switching time

Switching time is one of important factors in RFR operation. Figure 4 and Figure 5 show the effect of switching time to dynamics of feed gas along RFR.

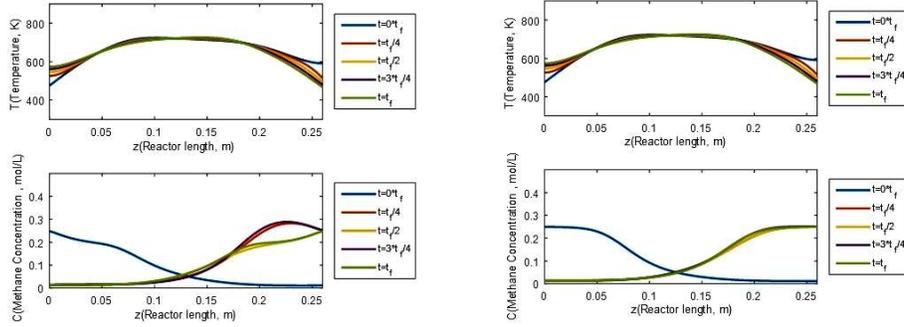


Figure 4. Temperature and concentration behavior of feed gas for various time values t with $A = 0.25$, $\varepsilon = 0.05$, $t_f = 20$ s after six times reversed.

Left figure, for $\omega = 5\pi$ and right figure $\omega = \frac{\pi}{5}$.

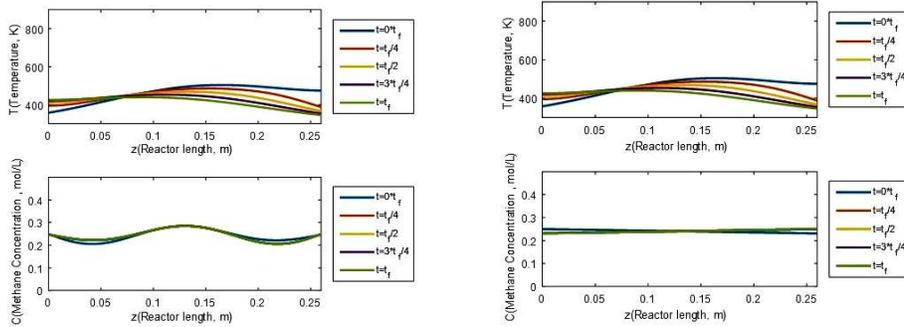


Figure 5. Temperature and concentration behavior of feed gas for various values t value with $A = 0.25$, $\varepsilon = 0.05$, $t_f = 120$ s after six times reversed.

Left figure, for $\omega = 5\pi$ and right figure $\omega = \frac{\pi}{5}$.

The choice of the duration of reversal (switching time) is very influential on the stability of the RFR in operation. From Figure 4 and Figure 5, it is indicated that the reversal time that is too short (case $t_f = 20$ s) or too long (case $t_f = 120$ s) will make the RFR go out. This situation is shown by the dynamics of temperature which decreases closer to the feed gas temperature. It shows that there is no reaction in the reactor. This result is in agreement with the results that reported in [10] and [17].

Parameter sensitivity analysis of superficial gas velocity

The effects of superficial gas velocity value to behavior of feed gas along RFR are shown in Figure 6 and Figure 7. When in the operation of the RFR, the feed gas flow velocity is small, it is indicated that the reactor will eventually go out. A slow feed gas flow rate will cause more feed gas to act in the reactor end position where the feed gas enters. Thus, getting to the end of the reactor, there is no additional heat from the reaction product. Simultaneously, the contribution of reactor cooling continues. As a result, over time the overall temperature becomes low and eventually goes out. The opposite condition occurs when the feed gas flow rate is increased as shown in Figure 7.

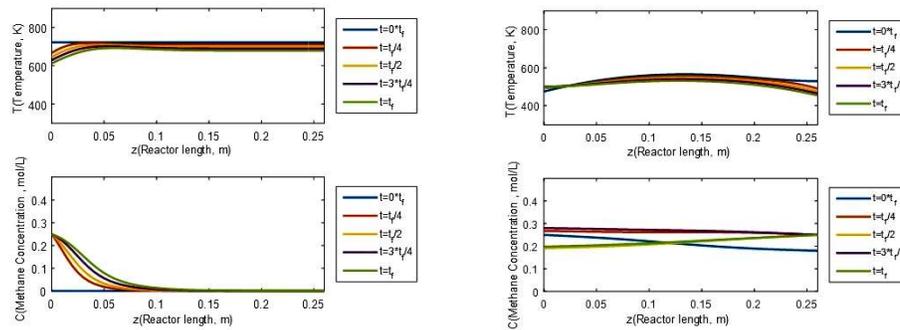


Figure 6. Temperature and concentration behavior of feed gas for various time values t with $A = 0.25$, $\varepsilon = 0.05$, $u = 0.1$ after six times reversed. Left figure, for $\omega = 5\pi$ and right figure $\omega = \frac{\pi}{5}$.

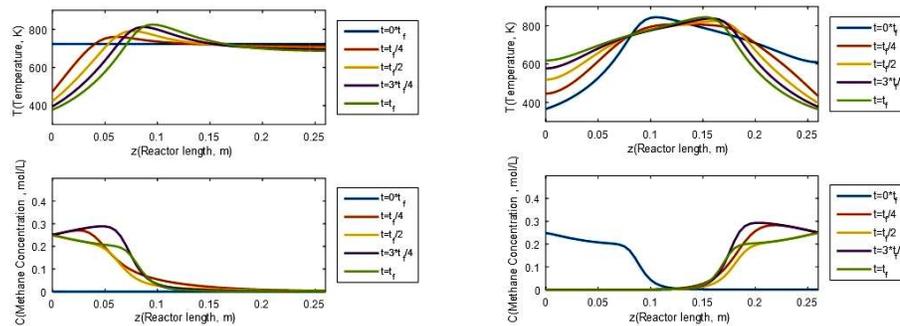


Figure 7. Temperature and concentration behavior of feed gas for various time values t with $A = 0.25$, $\varepsilon = 0.05$, $u = 0.7$.

4. Conclusion

RFR stability investigations have been shown for the oxidation process with periodically disturbed feed gas. The parameter sensitivity analysis process through a numerical approach was carried out to see the effect of the feed gas period, switching time and superficial gas velocity. The results of the analysis showed that the choice of switching time and superficial gas velocity greatly influences the sustainability and stability of RFR operations.

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