MODELING HEAT AND MASS TRANSFER WITH CHEMICAL KINETICS FOR THE PYROLYSIS OF OIL SLUDGE

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Abstract. *Several activities in the oil industry, such as drilling, production, refining, transport, processing and distribution, create considerable amounts of hazardous waste, such as oil sludge, which has one of the highest generation rates in refineries. Gasification can be considered a promising technology for the thermochemical conversion of waste from the oil industry since it is a process with minimal environmental impact. It allows the production of syngas, which can have different potential applications, such as the production of liquid fuels, electricity generation, hydrodesulfurization, among others. Thus, this work aims to theoretically evaluate oil sludge gasification seeking to use syngas produced in hydrodesulfurization processes. The theoretical analysis consists of modeling the heat and mass transfer, considering the chemical kinetics involved in the different stages of the gasification process. A one-dimensional model to describe the heat and mass transfer in the zones of the gasification process was elaborated, from mass balances for each component, as well as for the energy balance for the mixture of components. Different chemical reactions, which may be present at various stages, were considered in the model. The resulting system includes 30 non-linear coupled PDEs for describing the transport phenomena in the gasification process. A simplified version of the model, consisting of a steady state operations without axial diffusion was then considered, and computational implementation of the solution was implemented on the Wolfram Mathematica platform. The analysis is focused on the first two-regions of the gasifier, namely drying and pyrolysis. Finally, illustrative results are presented to analyze the effect of different operation and construction parameters on the efficiency of the gasification process.*

Keywords: *Thermochemical conversion; petroleum waste; oxidation; pyrolis*

1. INTRODUCTION

Oil sludge is one of the main wastes generated along the production chain of the petroleum industry. This residue represents risks to human health and the environment when mismanaged. Additionally, the sludge contains a high content of heavy hydrocarbons, which represent a significant potential for reuse as raw material in various applications, including thermochemical conversion processes. Thus, there is a growing demand for the improvement of oil sludge treatment methods, to mitigate its environmental impacts and to enable its reutilization.

The oil sludge itself is an oily residue composed of a mixture of oil, solids, and water, with occasional presence of other contaminants (Guimarães, 2008). Due to the recalcitrant nature of the sludge, achieving a balance between complying with strict environmental regulations and reducing treatment costs is challenging (Hu *et al.*, 2013). One promising approach to deal with the sludge is through gasification treatment, which involves the thermochemical conversion of the sludge into a gas called syngas (Andrade, 2007). This process offers low environmental impact and the possibility of reusing the syngas in high-value-added products, such as electricity generation and reuse as raw material in other processes within the petrochemical industry (Castillo Santiago *et al.*, 2022).

The objective of this article is to develop a mathematical model of the oil sludge gasification process, aiming to provide a qualitative guidance on input variables and on the influence of the design and operational parameters of a gasifier on the properties of the synthesis gas, considering mass balance, chemical kinetics of the reactions involved in the process, focusing on the Pyrolisis stage. A simplified version of the model is then computationally solved using the Wolfram Mathematica software.

2. PROBLEM FORMULATION

A heat and mass transfer model for the gasification process is herein developed. The model considers a downdraft gasifier, in which the fuel is supplied at the top of the reactor and the gasification agent is supplied in the oxidation zone, as seen in figure 1. A homogeneous mixture is considered, being composed of gas, liquid and solid phases, all of which

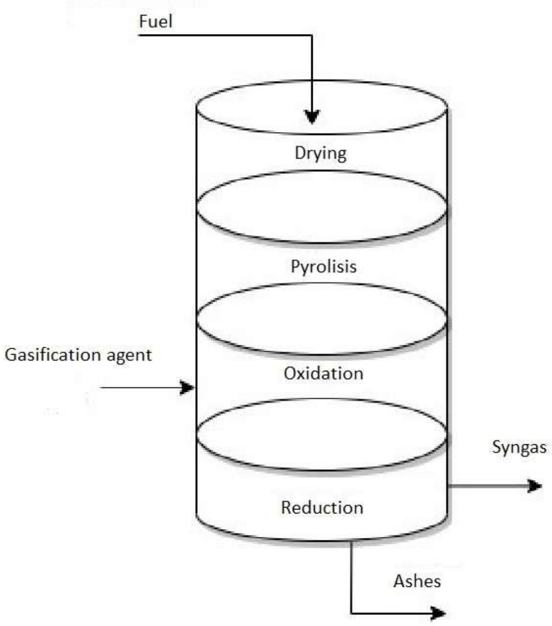

Figure 1. problem diagram

can co-exist within the reactor. Mass conservation for an arbitrary point within the gasifier yields:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho\, \boldsymbol{v}) = 0, \tag{1}$$

$$\frac{\partial C_i}{\partial t} + \nabla \cdot \big(C_i\, \boldsymbol{v} - \rho\, \mathcal{D}_i\, \nabla \omega_i)\big) = \dot{r}_i, \tag{2}$$

where $\boldsymbol{v}$ is the mixture velocity, $\mathcal{D}_i$ is the mass diffusivity of a particular species i, and $\dot{r}_i$ is the production rate for the same species. The mixture density ρ can be written in terms of the volumetric concentration of all species C_i, which is related to the mass concentration $\omega_i = C_i/\rho$, such that:

$$\rho = \sum_{i=1}^{i_{\max}} C_i, \qquad 1 = \sum_{i=1}^{i_{\max}} \omega_i, \tag{3}$$

Fick's Law mass diffusion coefficient is given by Barrozo and Benedetti (2016):

$$\mathcal{D}_i = \mathcal{D}_{i,0} \exp\left(\frac{-E_i}{R\,T}\right) \tag{4}$$

where E_i is the activation energy for a particular species and R is the gas constant.

The system given by equation (1) and the $i_{\max}$ equations (2) for $i = 1, \dots, i_{\max}$, are the governing equations that must be solved for determining the concentration of each species.

Table 1 lists all chemical species considered. The molecular mass for oil sludge M_{21} is calculated as a function of the composition coefficients v, w and x:

$$M_{21} = 12\, v + w + 16\, x. \tag{5}$$

Although sulphur and nitrogen are typically found in oil sludges, these are inert to the processes drying and pyrolisis, such that they are not included in the analysis.

Table 2 lists the chemical reactions involved in the gasifier. Note that each reaction receives a particular number denoted as j. The stoichiometry for each reaction is presented next. During the drying stage no particular reaction occurs, as there is only a phase change from liquid to gaseous water (vapor). For the pyrolysis zone, cracking reactions occur, for phenol and naphthalene, respectively:

$$\mathrm{C_6H_6O} \longrightarrow \mathrm{CO} + \frac{2}{5}\,\mathrm{C_{10}H_8} + \frac{3}{20}\,\mathrm{C_6H_6} + \frac{1}{10}\,\mathrm{CH_4} + \frac{3}{4}\,\mathrm{H_2} \tag{6a}$$

$$\mathrm{C_{10}H_8} \longrightarrow \frac{13}{2}\,\mathrm{C} + \frac{1}{2}\,\mathrm{C_6H_6} + \frac{1}{2}\,\mathrm{CH_4} + \frac{3}{2}\,\mathrm{H_2} \tag{6b}$$

Table 1. Chemical species

i	species	formula	M (g/mol)
1	hydrogen	H_2	2
2	methane	CH_4	16
3	steam	H_2O(v)	18
4	carbon monoxide	CO	28
5	nitrogen	N_2	28
6	oxygen	O_2	32
7	carbon dioxide	CO_2	44
8	pyrene	$C_{16}H_{10}$	202
9	bezene	C_6H_6	78
10	toluene	C_7H_8	92
11	phenol	C_6H_6O	94
12	chrisene	$C_{18}H_{12}$	228
13	xylene	C_8H_{10}	106
14	naphthalene	$C_{10}H_8$	128
15	anthracene	$C_{14}H_{10}$	178
16	phenanthrene	$C_{14}H_{10}$	178
17	calcium oxide	CaO	56
18	calcium carbonate	$CaCO_3$	100
19	char	C	12
20	water (liq.)	H_2O(l)	18
21	sludge	$\mathrm{C}_v\mathrm{H}_w\mathrm{O}_x$	M_{21}

Table 2. Chemical reactions: drying and pyrolisis

j	reaction	ΔH (kJ/mol)	zone
29	fuel drying	-44	drying
30	sludge devolatization	-420	pyrolisis
1	phenol cracking	51	pyrolisis
2	naphthalene cracking	-150	pyrolisis
3	reforming of benzene	-7.8	pyrolisis
4	reforming of toluene	-12	pyrolisis
5	reforming of xylene	90	pyrolisis
6	reforming of anthracene	94	pyrolisis
7	reforming of phenanthrene	79	pyrolisis
8	reforming of chrisene	81	pyrolisis
9	reforming of pyrene	38	pyrolisis

then, there are reforming reactions for benzene, toluene, xylene, anthracene, phenanthrene, chrisene and pyrene, respectively:

$$\mathrm{C_6H_6} + 2\,\mathrm{H_2O} \longrightarrow \frac{3}{2}\,\mathrm{C} + \frac{5}{2}\,\mathrm{CH_4} + 2\,\mathrm{CO} \tag{6c}$$

$$\mathrm{C_7H_8} + 2\,\mathrm{H_2O} \longrightarrow 2\,\mathrm{C} + 3\,\mathrm{CH_4} + 2\,\mathrm{CO} \tag{6d}$$

$$\mathrm{C_8H_{10}} + 2\,\mathrm{H_2O} \longrightarrow 2\,\mathrm{CO} + \frac{5}{9}\,\mathrm{C_6H_6} + \frac{8}{3}\,\mathrm{CH_4} \tag{6e}$$

$$\mathrm{C_{14}H_{10}} + 2\,\mathrm{H_2O} \longrightarrow 2\,\mathrm{CO} + \frac{1}{10}\,\mathrm{C_6H_6} + \frac{2}{5}\,\mathrm{CH_4} + \frac{37}{10}\,\mathrm{H_2} + \frac{11}{20}\,\mathrm{C_{10}H_8} + \frac{11}{2}\,\mathrm{C} \tag{6f}$$

$$\mathrm{C_{14}H_{10}} + 2\,\mathrm{H_2O} \longrightarrow 2\,\mathrm{CO} + \frac{1}{10}\,\mathrm{C_6H_6} + \frac{2}{5}\,\mathrm{CH_4} + \frac{47}{10}\,\mathrm{H_2} + \frac{3}{10}\,\mathrm{C_{10}H_8} + 8\,\mathrm{C} \tag{6g}$$

$$\mathrm{C_{18}H_{12}} + 2\,\mathrm{H_2O} \longrightarrow 2\,\mathrm{CO} + \frac{13}{100}\,\mathrm{C_6H_6} + \frac{33}{100}\,\mathrm{CH_4} + \frac{443}{100}\,\mathrm{H_2} + \frac{63}{100}\,\mathrm{C_{10}H_8} + \frac{859}{100}\,\mathrm{C} \tag{6h}$$

$$\mathrm{C_{16}H_{10}} + 2\,\mathrm{H_2O} \longrightarrow 2\,\mathrm{CO} + \frac{1}{10}\,\mathrm{C_6H_6} + \frac{3}{10}\,\mathrm{CH_4} + \frac{57}{10}\,\mathrm{H_2} + \frac{1}{10}\,\mathrm{C_{10}H_8} + \frac{121}{10}\,\mathrm{C} \tag{6i}$$

and finally, the reaction of devolatization of the sludge:

$$\mathrm{C}_v\mathrm{H}_w\mathrm{O}_x \longrightarrow v_{\mathrm{C}}\,\mathrm{char} + v_{\mathrm{CO}}\,\mathrm{CO} + v_{\mathrm{CO_2}}\,\mathrm{CO_2} + v_{\mathrm{CH_4}}\,\mathrm{CH_4} + v_{\mathrm{H_2}}\,\mathrm{H_2} + v_{\mathrm{H_2O}}\,\mathrm{H_2O} + v_{\mathrm{C_6H_6O}}\,\mathrm{C_6H_6O} \tag{6j}$$

where, naturally, the values for v_i are obtained from:

$$v_{\mathrm{C}} + v_{\mathrm{CO}} + v_{\mathrm{CO_2}} + v_{\mathrm{CH_4}} + 6\,v_{\mathrm{C_6H_6O}} = v \tag{7}$$

$$4\,v_{\mathrm{CH_4}} + 2\,v_{\mathrm{H_2}} + 2\,v_{\mathrm{H_2O}} + 6\,v_{\mathrm{C_6H_6O}} = w \tag{8}$$

$$v_{\mathrm{CO}} + 2\,v_{\mathrm{CO_2}} + v_{\mathrm{H_2O}} + v_{\mathrm{C_6H_6O}} = x \tag{9}$$

considering that $v_{\mathrm{CO_2}} = v_{\mathrm{CO}} = 0$ and that $v_{\mathrm{C}} = v_{\mathrm{C_6H_6O}} = 1$.

Once the reaction equations have been presented, the rate of production for each species is presented. These are obtained from the velocity of the previously presented reactions.

The production rate for each component $\dot{r}_i$ is written in a general form in termos of a coefficient matrix, involving the stoichiometric coefficients $X_{i,j}$:

$$\dot{r}_i = M_i \sum_{j=1}^{29} X_{i,j}\hat{r}_j \tag{10}$$

for $i = 1, 2, \ldots, i_{\max}$. Its important noticing that the values $X_{i,j}$ are positive in reactions that involve the production of a certain species (products) and negative when these are consumed (reactants). The coefficients $\hat{r}_j$ represent the velocity of a given j reaction, which is calculated via:

$$\hat{r}_j = A_j \exp\left(\frac{-E_{a,j}}{RT}\right) [\mathrm{C}_v\mathrm{H}_w]^\alpha\,[\mathrm{H_2}]^\beta\,[\mathrm{H_2O}]^\delta \tag{11}$$

where the terms within square brackets represent the molar concentration of each species, that is:

$$[\mathrm{H_2O}] = C_3/M_3 \tag{12}$$

$$[\mathrm{H_2}] = C_1/M_1 \tag{13}$$

$$[\mathrm{CH_4}] = C_2/M_2 \tag{14}$$

$$[\mathrm{C_{16}H_{10}}] = C_8/M_8 \tag{15}$$

$$[\mathrm{C_6H_6}] = C_9/M_9 \tag{16}$$

$$[\mathrm{C_7H_8}] = C_{10}/M_{10} \tag{17}$$

$$[\mathrm{C_{18}H_{12}}] = C_{12}/M_{12} \tag{18}$$

$$[\mathrm{C_8H_{10}}] = C_{13}/M_{13} \tag{19}$$

$$[\mathrm{C_{10}H_8}] = C_{14}/M_{14} \tag{20}$$

$$[\mathrm{C_{14}H_{10}}](\text{anthracene}) = C_{15}/M_{15} \tag{21}$$

$$[\mathrm{C_{14}H_{10}}](\text{phenanthrene}) = C_{16}/M_{16} \tag{22}$$

For the processes of drying and oil sludge devolatization, the concentration of H_2O corresponds to liquid water (i.e. H_2O_l). Also for the sludge devolatization, $[C_vH_w]$ is equivalent to the fuel concentration $[C_vH_wO_xN_yS_z]$.

In order to determine the values of $X_{i,j}$ the rate of production of each species is written as

$$\mathrm{H_2}: \quad \frac{\dot{r}_1}{M_1} = 0.75\,\hat{r}_1 + 1.5\,\hat{r}_2 + 3.7\,\hat{r}_6 + 4.7\,\hat{r}_7 + 4.43\,\hat{r}_8 + 5.7\,\hat{r}_9 + v_{H2}\,\hat{r}_{30} \tag{23a}$$

$$\mathrm{CH_4}: \quad \frac{\dot{r}_2}{M_2} = 0.1\,\hat{r}_1 + 0.5\,\hat{r}_2 + 2.5\,\hat{r}_3 + 3\,\hat{r}_4 + 2.667\,\hat{r}_5 + 0.4\,\hat{r}_6 + 0.4\,\hat{r}_7 + 0.33\,\hat{r}_8 + 0.3\,\hat{r}_9 + v_{CH4}\,\hat{r}_{30}, \tag{23b}$$

$$\mathrm{H_2O}: \quad \frac{\dot{r}_3}{M_3} = -2\sum_{j=3}^{9}\hat{r}_j + \hat{r}_{29} + v_{H2O}\,\hat{r}_{30}, \tag{23c}$$

$$\mathrm{CO}: \quad \frac{\dot{r}_4}{M_4} = \hat{r}_1 + 2\sum_{j=3}^{9}\hat{r}_j + v_{CO}\,\hat{r}_{30}, \tag{23d}$$

$$\mathrm{CO_2}: \quad \frac{\dot{r}_7}{M_7} = v_{CO2}\,\hat{r}_{30} \tag{23e}$$

$$\mathrm{C_{16}H_{10}}: \quad \frac{\dot{r}_8}{M_8} = -\hat{r}_9 \tag{23f}$$

$$\mathrm{C_6H_6}: \quad \frac{\dot{r}_9}{M_9} = 0.15\,\hat{r}_1 + 0.5\,\hat{r}_2 - \hat{r}_3 + 0.556\,\hat{r}_5 + 0.1\,\hat{r}_6 + 0.1\,\hat{r}_7 + 0.13\,\hat{r}_8 + 0.1\,\hat{r}_9, \tag{23g}$$

$$\mathrm{C_7H_8}: \quad \frac{\dot{r}_{10}}{M_{10}} = -\hat{r}_4, \tag{23h}$$

$$\mathrm{C_6H_6O}: \quad \frac{\dot{r}_{11}}{M_{11}} = -\hat{r}_1 + v_{C6H6O}\,\hat{r}_{30} \tag{23i}$$

$$\mathrm{C_{18}H_{12}}: \quad \frac{\dot{r}_{12}}{M_{12}} = -\hat{r}_8, \tag{23j}$$

$$\mathrm{C_8H_{10}}: \quad \frac{\dot{r}_{13}}{M_{13}} = -\hat{r}_5, \tag{23k}$$

$$\mathrm{C_{10}H_8}: \quad \frac{\dot{r}_{14}}{M_{14}} = 0.4\,\hat{r}_1 - \hat{r}_2 + 0.55\,\hat{r}_6 + 0.3\,\hat{r}_7 + 0.63\,\hat{r}_8 + 0.1\,\hat{r}_9, \tag{23l}$$

$$\mathrm{C_{14}H_{10}}: \quad \frac{\dot{r}_{15}}{M_{15}} = -\hat{r}_6, \tag{23m}$$

$$\mathrm{C_{14}H_{10}}: \quad \frac{\dot{r}_{16}}{M_{16}} = -\hat{r}_7, \tag{23n}$$

$$\mathrm{C}: \quad \frac{\dot{r}_{19}}{M_{19}} = 6.5\,\hat{r}_2 + 1.5\,\hat{r}_3 + 2\,\hat{r}_4 + 5.5\,\hat{r}_6 + 8\,\hat{r}_7 + 8.59\,\hat{r}_8 + 12.1\,\hat{r}_9 + v_{char}\,\hat{r}_{30}, \tag{23o}$$

$$\mathrm{H_2O}(l): \quad \frac{\dot{r}_{20}}{M_{20}} = -\hat{r}_{29} \tag{23p}$$

$$C_vH_wO_xN_yS_z: \quad \frac{\dot{r}_{21}}{M_{21}} = -\hat{r}_{30} \tag{23q}$$

Finally, table 3 presents the kinetic parameters for the previously presented reactions.

Table 3. Kinetic parameters for chemical reactions

j	A_j^a	$E_{a,j}$, kJ/kmol	α	β	δ	Ref.
1	1.0×10^7	1.0×10^5	1.0	0	0	Morf *et al.* (2002)
2	1.0×10^{14}	3.5×10^5	1.6	-0.5	0	Sreejith *et al.* (2015)
3	2.0×10^{16}	4.43×10^5	1.3	-0.4	0.2	Ji *et al.* (2009)
4	3.3×10^{10}	2.47×10^5	1.0	0.5	0	Jess (1996)
5	3.3×10^{10}	2.47×10^5	1.0	0.5	0	Sreejith *et al.* (2015)
6	1.7×10^{14}	3.5×10^5	1.6	-0.5	0	Sreejith *et al.* (2015)
7	1.7×10^{14}	3.5×10^5	1.6	-0.5	0	Sreejith *et al.* (2015)
8	1.7×10^{14}	3.5×10^5	1.6	-0.5	0	Jess (1996)
9	1.7×10^{14}	3.5×10^5	1.6	-0.5	0	Jess (1996)
29	5.13×10^{10}	88	0	0	1.0	Gómez-Barea and Leckner (2010)
30	1.52×10^3	105	1.0	0	0	Patra *et al.* (2016)

3. Computational solution

For this paper a simplified version of the model is considered in the computational implementation. Firstly, a one-dimensional steady formulation is considered; then, the effects of mass diffusion are considered negligible compared to those of the advective transport. Hence, the governing equations are reduced to:

$$\frac{\partial}{\partial z}(v_z\,\rho) \;=\; 0 \tag{24}$$

$$\frac{\partial}{\partial z}(v_z\,C_i) \;=\; \dot{r}_i, \quad \text{for} \quad i = 1, \ldots, 20, \tag{25}$$

and the momentum equation leads to:

$$v_z\,\frac{\partial v_z}{\partial z} \;=\; 0 \tag{26}$$

the mixture velocity is constant, such that

$$v_z \frac{\mathrm{d}\rho}{\mathrm{d}z} = 0 \tag{27}$$

$$v_z \frac{\mathrm{d}C_i}{\mathrm{d}z} = \dot{r}_i, \quad \text{for} \quad i = 1, \ldots, 20, \tag{28}$$

which implies that the mixture density ρ must also be a constant and only equations (28) require a solution. Despite the simplicity of equations (28), they constitute a nonlinear system of ODEs that require a numerical solution. This is achieved using the *Mathematica* function **NDSolve**, after imposing the following inlet conditions (at $z = H$):

$$C_i(H) = 0, \quad \text{for} \quad i = 1, \ldots, 20, \tag{29}$$

$$v_z \, \mathcal{A}_z \, C_{21}(H) = \dot{m}_{in,21} \tag{30}$$

where $\mathcal{A}_z$ is the inlet area, $\dot{m}_{in,21}$ is the sludge inlet flowrate and C_{21} is directly obtained from its density, as only sludge is supplied at the inlet.

In order to carry on the computational solution properties and process data are required. The selected input parameters used in the simulations are presented in table 4. The sludge composition is obtained from the elementar analysis performed in Santiago *et al.* (2023), whose results are summarized in table 5.

Table 4. Input data

Quantity	value	unit
Inlet temperature	25	°C
inlet sludge density	1010	kg/m³
sluge inlet flowrate	5	kg/h
reactor height	0.6	m
reactor diameter	0.1	m

Table 5. Oil sludge ultimate analysis

Element	symbol	mass (%)	kmol
carbon	C	85.84	7.1533
hydrogen	H	10.70	10.6151
oxygen	O	1.99	0.1244
nitrogen	N	1.19	0.0850
sulfur	S	0.28	0.00873

4. RESULTS AND DISCUSSION

Once the simplified model and numerical solution scheme have been discussed, preliminary results of the numerical implementation are presented. A isothermal operation is considered for simplification purposes, at the current stage of development of this work. Figure 2 displays the concentration distribution of methane, while figure 3 display the similar results for other components. As can be seen, for the simulated conditions, the reactions occur very neat the entrance of the reactor (i.e. $z = 0.6$m). Also, a much larger methane concentration is seen when compared with the other species. The overlapping curves are all at zero concentration due to the adopted simplifications in the reactions.

5. SUMARY AND CONCLUSIONS

Oil sludge is one of the most generated residues in the oil industry. Its treatment is challenging due to the content of heavy materials and petroleum hydrocarbons in the residue. In this work, computational modeling of the gasification process that includes the chemical equilibrium and heat and mass transfer mechanisms was developed. The study was carried out considering mass balance and the chemical kinetics of the reactions involved in the process, in a downdraft gasifier. For the computational solution was considered a simplified version of the model, with a one dimensional steady formulation assuming negligible effects of mass diffusion, which was solved using the Wolfram Mathematica software. Finally, it can be concluded that gasification represents a promising technological alternative for the management and treatment of oil sludge since it produces a gas that can be used in different applications, with emphasis on electricity generation, and at the same time considerably reduces the environmental impact associated with this oily residue.

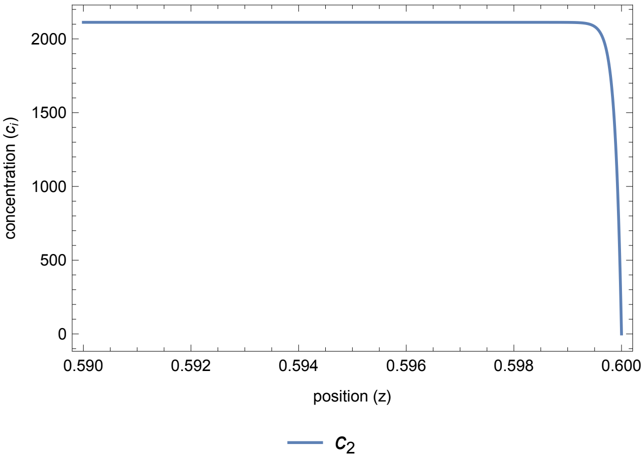

Figure 2. Methane concentration distribution (g/l)

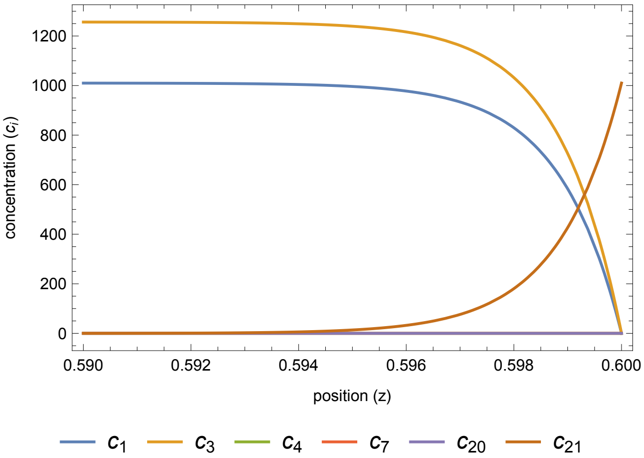

Figure 3. Concentration distribution for other species (g/l.

6. ACKNOWLEDGEMENTS

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