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Generalized model for predicting methane conversion to syngas in membrane reactor

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Abstract

Present work aims to provide a conceptual framework for predicting methane conversion efficiency and CO selectivity in a membrane reactor which may assist in selecting the type of membrane and minimizing the cost of syngas production. A comparative evaluation of the performance of ceramic (inorganic) membrane reactors was carried out with other types of synthetic membrane reactors. Data has been taken only from those research papers in which reactor is operating in a temperature range of 800 – 900ºC and the pressure range is 1-2 atm. Linear regression analysis was performed and a generalized equation was developed to predict the methane conversion and CO selectivity using different types of membranes. Simulations were performed using MATLAB ODE45 to investigate feasibility of the developed model. CO selectivity was observed better for ceramic membrane reactors since it achieves 90% saturation in 25 hours while for other synthetic membranes, the conversion is only 10% (approx). Overall, ceramic membrane reactors were found to be better than other synthetic membrane reactor due to effective CO selectivity and CH_4 conversion.

Keywords: Syngas, CH_4 conversion, CO selectivity, ceramic membrane

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1. Introduction

Rapid innovations in technologies and fast paced economic development across the globe are foretokens to edify the necessity of sustainable energy resources. Annual Energy Outlook 2012 projected the increase in world energy consumption by 47% from year 2010 to year 2035 (Energy Outlook, 2012). Global population is projected to reach approximately 8.8 billion people by year 2035 and consequently, world GDP is expected to more than double over the same time period (BP Outlook, 2016). Developing countries China and India are expected to drive the world economy by contributing together for almost 50% of the hike in global GDP (BP Outlook, 2016). Though, economic development and technology advancement are requisite for human welfare, it is pragmatically impossible to have economic growth without concomitant consumption of energy resources (Chauhan et al., 2013). Therefore, an urgent need is felt to look for sustainable energy resources which can be exploited as primary energy source.

Fossil fuels are being considered significant contributor in global energy supply, however tremendous increase in greenhouse gas emission and onset of climate change are prevalent issues associated with fossil fuel combustion. Several research efforts are being made worldwide to develop novel technologies for the efficient utilization of renewable energy sources and sequestration of emitted greenhouse gases. Natural gas is being considered the most economically accessible energy resource in present scenario. The ascertained reserves of natural gas amounted world-wide to over 6973 trillion cubic feet (International Energy Statistics, 2015). Call for natural gas is going up by 1.8% per annum which clearly indicates urgent necessity of this fossil fuel. Emerging economies (China and India) account for approximately 30% of the increase in natural gas demand for industrial utilization and power generation (BP Outlook, 2016).

Extensive research efforts have been riveted on up-gradation of natural gas to value-added products. Conversion of natural gas to liquid fuels (GTL) through a syngas intermediate (CO/H_2) is one of the major research streams in order to conserve the energy

resources. Three possible pathways (steam Reforming, dry reforming and partial oxidation of methane) may be adopted for the natural gas to liquid fuel conversion during GTL process. Steam reforming process (CH_4 + steam → CO + H_2), being an endothermic process, requires extensive energy supply. Also, poor selectivity for CO and high H_2/CO product ratio are the major limitations which make it unsuitable for industrial applications. Dry reforming process ($CH_4 + CO_2 \rightarrow CO + H_2$) utilizes the carbon dioxide gas and thus can reduce the greenhouse gas emission, however thermodynamic constraints associated with the process limit conversion efficiency and increases the economics of the process (Gallucci et al., 2008). Partial oxidation of methane (POM) ($CH_4 + O_2 \rightarrow CO + H_2$) over the supported transition metals is gaining attention these days due to higher reaction rate, more favourable H_2/CO ratio and mild exo-thermicity of the reaction (Wei et al., 2011). Several studies have been reported to identify novel catalysts for syngas production using POM pathway (Sharifnia and Karami, 2015; Wei et al., 2016), however, most of the catalysts for syngas synthesis are active at high temperatures (> 973 K). Secondly, consumption of significant quantities of expensive pure oxygen during POM and the cost associated with the production of pure O_2 using cryogenic air separators are added limitations which affect economics of the process.

Aforementioned limitations associated with available technologies elucidate the need of an innovative sustainable process for syngas production. Membrane reactors with oxygen semipermeable ionic (or mixed) conducting dense ceramic membranes have drawn considerable attention in last few decades due to their ability to carry out simultaneous reaction and separation (Tong et al., 2006; Silva et al., 2015; Rui et al., 2008). Broadly categorizing, the available membrane reactors can be divided under two categories i.e. ceramic (inorganic) membrane reactors and Organic/metallic/liquid membrane reactors. In the present study, all other synthetically prepared membranes except ceramic membranes are denoted as synthetic membrane reactor. Table 1 provides a brief review about the operating parameters and membrane properties employed in synthetic and ceramic membrane reactors.

Table 1 : Literature review on ceramic and synthetic membrane reactor

Reactor	Membrane	Operating Parameter	Efficiency	References
Ceramic Membrane Reactors				
Dual-phase composite membrane reactors.		960°C	Time dependence of oxygen permeation flux was observed during initial stages	Zhu et al., 2010
	Dense $BaCo_{0.7}Fe_{0.2}Ta_{0.1}O_{3-d}$ perovskite membrane	900°C	CH_4 conversion > 99% ; CO selectivity : 94%	Luo et al., 2010
	mixed conducting ceramic membrane reactors.	operating time : 2200 h Temp: 900°C Pressure: 1-1.2atm	CH_4 conversion > 96% ; CO selectivity > 95%	Tong et al., 2006
Hollow fibre catalytic reactor, Ni-based catalyst	perovskite mixed conducting membrane ($BaCo_xFe_yZr_zO_{3-d}$)	925°C	CO selectivity > 97% ; CH_4 conversion : 96%.	Wang et al., 2006
Membrane Reactor	ceramic composite of $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3\_d}$ (97.5 mol%) and Co_3O_4 (2.5 mol%).	T : 900°C Pressure : 1 atm	No significant change in CO selectivity with variation of CH_4 feeding rate	Chen et al., 2003
Tubular reactor (Catalyst : $LiLaNiO/-Al_2O_3$)	a perovskite material $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$, with electronic and ionic conductivity.	Temperature: 875°C	CO selectivity > 95% ; CH_4 Conversion > 94%	Wang et al., 2003
Catalytic membrane reactor, $LiLaNiO/\gamma$ -Al_2O_3 catalyst with 10wt.% nickel loading	zirconium-based dense membrane, mixed conducting perovskite ($BaCo_{0.4}Fe_{0.4}Zr_{0.2}O_{3-\delta}$)	Temperature : 850°C	CH_4 conversion : 96–98%; CO selectivity : 98–99%	Tong et al., 2002
Catalyst packed tubular reactor	Perovskite-type oxide $La_{0.6}Sr_{0.4}Co_{0.2}Fe_{0.8}O_{3-d}$	Temperature : 1098-1158 K	CH_4 conversion > 96% ; CO selectivity > 97% at low carbon space velocity	Jin et al., 2000
Other Synthetic Membrane Reactors				
Fixed bed reactor, Ni-based catalyst	Pd-Ag membrane	Temp : 823K, 873 K Pressure : 1.01×10^5 Pa	Hydrogen yield reached 21% at 823 K and 47% at 873K	Silva et al., 2015
Tubular membrane reactors	Pd–Ag alloy membrane	Temperature ranges 350 -450°C. 10^5 Pa	Maximum CO_2 conversion : 20.6% for the MR at 450°C, maximum CH_4 conversion: 17.41% for the TR at 450°C	Gallucci et al., 2008
Tubular reactor, Ni/γ-Al_2O_3 catalyst	oxygen-permeable $SrFeCo_{0.5}O_y$ membrane	Temperature : 900 °C; CH_4 feeding rate : 26.8 mL/min	Throughput conversion of CH_4 : 98%, CO selectivity : 98%, H_2/CO : 1.8, syngas generation rate : 16 $mL/min/cm^2$	Fend et al., 2004
a conventional reactor (CR) and a membrane reactor (MR)	50μm thick pinhole free Pd-Ag alloy membrane		Maximum CH_4 conversion : 69% at 450°C	Gallucci et al., 2004
a traditional reactor, Membrane reactor, Ni-based catalyst	composite ceramic palladium membrane reactors (CPMR), and a dense palladium membrane reactor (DPMR)	Temperature : 250–550°C.	DPMR showed highest methane conversion: 45.3% at 400°C and 56.2% at 450°C.	Basile and Paturzo, 2001
Traditional and membrane reactor	Palladium based membrane	Time factor of 4288 gcat min / mol, p = 1.2 bar	Maximum CH_4 conversion: 96% using palladium membrane reactor at 550°C	Basile et al., 2001

Present study compares a variety of membranes for the conversion of CH_4 to syngas. An extensive literature survey has been done and it was inferred that limited reports are available which directly compare the performance of different types of membrane. Therefore, a comparative study for synthetic and ceramic membrane reactors has been performed. Preparation of various types of dense membranes is an added cost to the syngas production process; therefore efforts have been made in this study to develop a generalized model for predicting the conversion efficiency. Mathematical modeling of the syngas production process using membrane reactors can assist the engineer in ameliorating operation and performance of membranes. Better prediction of the syngas production and optimization of the corresponding retaliatory actions are certainly needed to increase the conversion efficiency of the process. Data was collected across each system studied to compare various parameters such as CH_4 conversion and CO selectivity. This study was conducted using scientific databases with search parameters restricted to scholarly articles and journals published between 2001 and 2015. A generalized equation was developed using linear regression analysis to predict the conversion of methane to syngas and CO selectivity using different types of membranes. Simulations were also performed in order to investigate the workability of the conceptual framework of deduced model.

2. Mathematical Model

In the present study, a mathematical model was developed for the comparative evaluation of the performance of ceramic and synthetic membrane reactor. Data was collected from the previous experimental studies reported in literature (Balachandran et al., 1995; Barbieri et al., 1997; Basile et al., 2001a, 2001b; Chen et al., 2003; Feng et al., 2004; Galucci et al., 2004; Jin et al., 2000; Silva et al., 2015; Smit et al., 2007; Tong et al., 2006; Wang et al., 2003) and regression analysis was performed using least square method. Based on the collected literature data, two equations were generated in the following forms as shown in equation (1) & (2):

$$na + b\sum y - \sum y' = 0 \tag{1}$$

$$a\sum y + b\sum y^2 - \sum yy' = 0 \tag{2}$$

where a and b are constants and y define the data points. y´ is calculated using equation (3):

$$y' = \frac{(y_{n+1} - y_{n-1})}{2\Delta t} \tag{3}$$

The above two equations were solved in order to get the values of constants a and b which were then used to generate an ordinary differential equation as shown in equation (4):

$$\frac{dy}{dt} = a + by \tag{4}$$

The developed ordinary differential equation was solved using 4th order Runge-Kutta method as mentioned in equation (5) – (7):

$$y_{n+1} = y_n + \left(\frac{1}{6}\right)(k_1 + 2k_2 + 2k_3 + k_4) \tag{5}$$

$$h = x_{n+1} - x_n \tag{6}$$

where

$$k_1 = hf(x_n, y_n)$$

$$k_2 = hf(x_n + h/2, y_n + k_1/2)$$

$$k_3 = hf(x_n + h/2, y_n + k_2/2)$$

$$k_4 = hf(x_n + h, y_n + k_3) \tag{7}$$

3. MATLAB Simulation

ODE45 solver was employed in the present study from the function library of MATLAB version 7.6.0.324 (R2008a). ODE45 function implements 4th order Runge-Kutta method with a variable time step for efficient computation in order to solve the first order differential equation. **ode45 function** may be invoked from the command line which is mentioned here in equation (8) :

$$[t, y] = ode45(fname, tspan, y_o, opts) \tag{8}$$

4. Results and Discussions

Linear regression analysis for the data collected from literature resulted into the linear equations of the type y = mx +c. Equations (9) and (10) demonstrate the linear equations obtained for CO selectivity using ceramic membrane and synthetic membranes respectively.

Ceramic Membranes $$y' = 16.44383 - 0.167770\,y \quad (9)$$

Synthetic Membranes $$y' = 0.0527918 - 0.0053867\,y \quad (10)$$

The derived equation for CO selectivity (equation 9) of ceramic membrane has a negative slope indicating a decrease by 0.1678 in time-rate change of CO selectivity with 1% improvement in CO selectivity and will show a 16.4% selectivity with zero improvement in CO selectivity. The respective equation (equation 10) for synthetic membrane having a positive slope indicates an increase by 0.0054 in time-rate change of CO selectivity with 1% improvement in CO selectivity and will show a 0.053% selectivity with zero improvement in CO selectivity.

Similarly, linear equations were derived for methane conversion using ceramic and synthetic membranes. Equations (11) and (12) depict the methane conversion in case of ceramic and synthetic membrane reactor respectively.

Ceramic Membranes $$y' = 45.74085 - 0.459155\,y \quad (11)$$

Synthetic Membranes $$y' = 13.43801 - 0.072750\,y \quad (12)$$

The regression equations for CH_4 conversion for both the membranes have negative slopes and positive intercepts describing an increase by 0.46 and 0.072 in time-rate change of CH_4 conversion with 1% improvement in CH_4 conversion efficiency for ceramic and synthetic membranes respectively. Nearly 45.7% and 13.4% CH_4 conversion ability was observed for ceramic and synthetic membranes respectively if zero improvement is made. The equations obtained by linear regression predict that the CO selectivity of synthetic membrane will follow linear growth with respect to time while the CO selectivity of ceramic membrane will increase with time. The high intercept value of the equations derived for CH_4 conversion indicate that higher start up time is required by ceramic membrane reactors than synthetic membrane reactors.

All the derived equations (9) - (12) are ordinary differential equation of first order which were solved using OE45 solver of MATLAB®. It was demonstrated in Fig.1 that (%) CO selectivity for both synthetic and ceramic membrane increased with increase in operation time of the reactor. The (%) CO selectivity for ceramic membrane followed a concave down curve reaching a saturation selectivity of 98% within 30 h of operation while the plot was linear for synthetic membrane and showed only 5% selectivity up till 50 h of operation as shown in Fig.1.

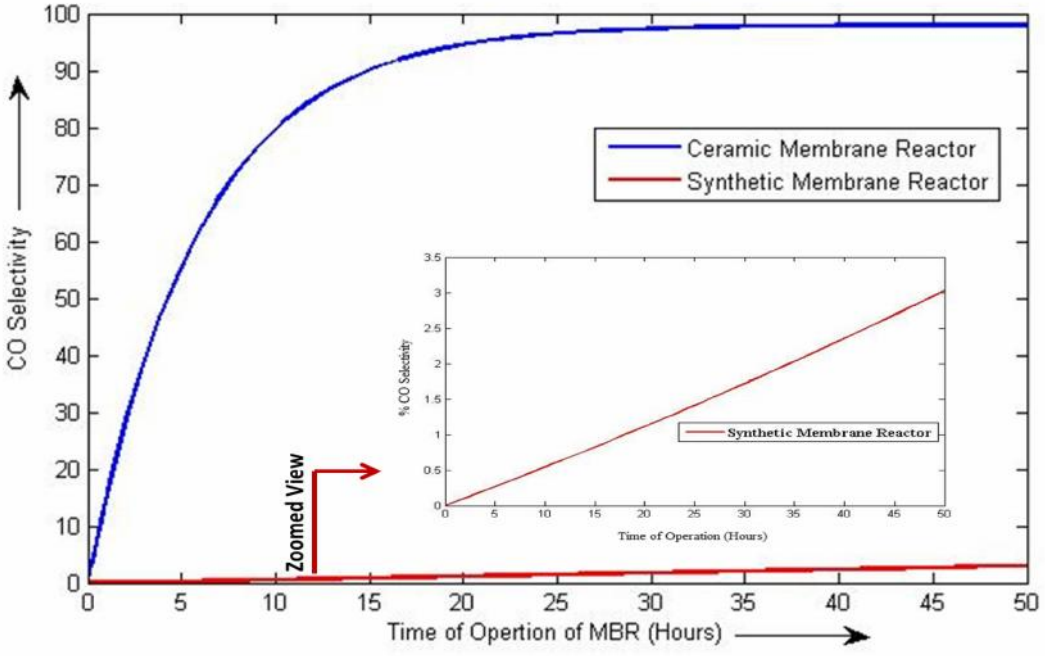

Fig. 1 : CO selectivity of Ceramic & Synthetic Membrane Reactors

Similarly, the plot of regression equations using ODE45 for CH_4 conversion was observed to be concave down curves for both types of membranes with a 99% conversion within 10 h of operation for ceramic membrane while the curve for synthetic membrane had not achieved saturation up till 50 h of operation.

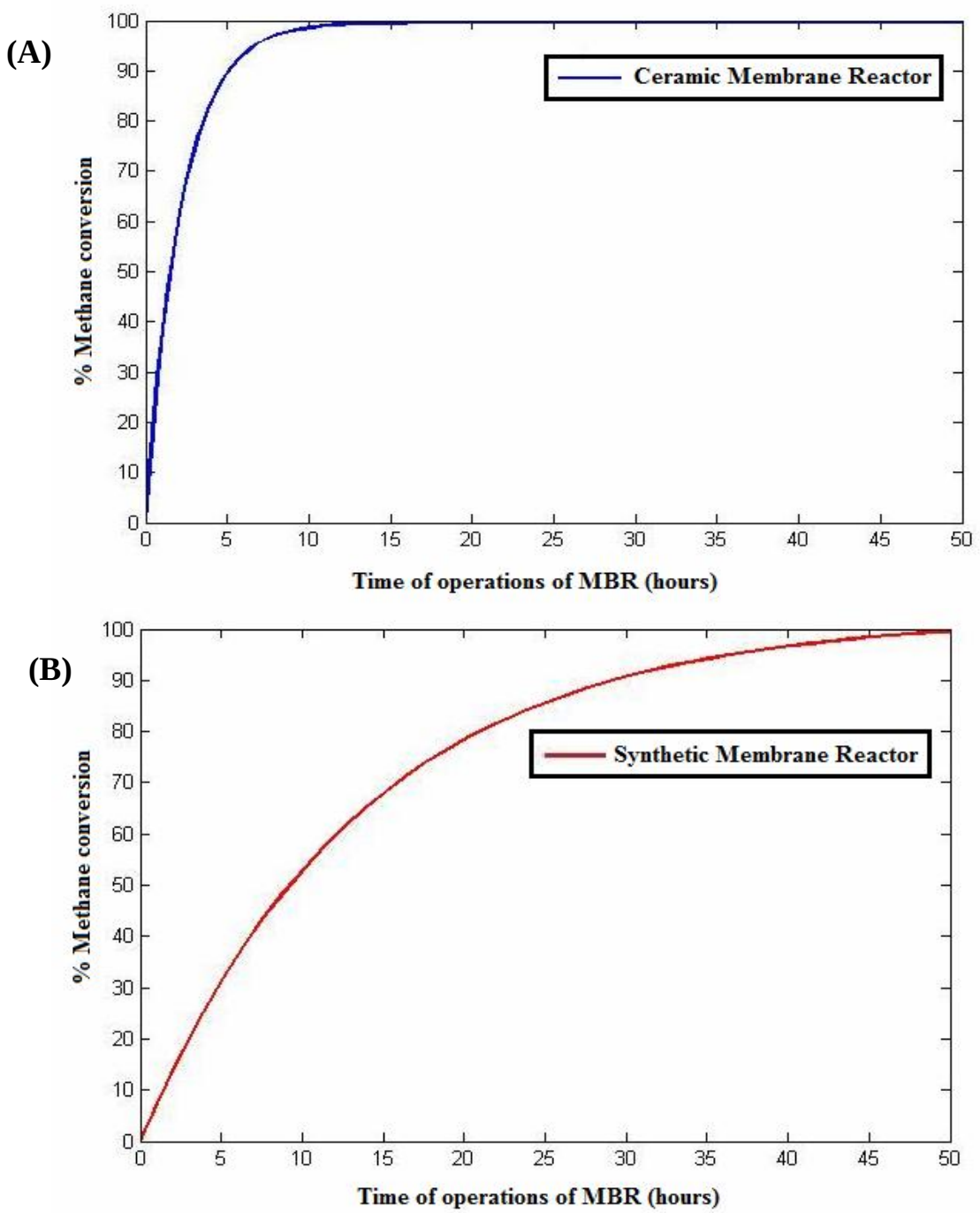

Fig. 2 : CH_4 Conversion for (A) Ceramic Membrane Reactor (B) Synthetic Membrane Reactors

Due to the reaction between methane and the oxygen permeating through the membrane, H_2 and CO are generated in presence of reforming catalyst. Direct contact between the tube and the reducing gases may cause the oxide phase to lose its lattice oxygen. When a portion of the oxygen is lost, the lattice would collapse and the material would decompose and break apart thus making CH4 conversion stable (Balachandran et al., 1995) in ceramic membrane reactor. Synthetic membrane showed approximately 80% higher conversion-ability compared to ceramic membrane as shown in Fig. 2. It was reported in literature (Wang et al., 2003) that POM reaction in the tubular membrane reactor follows the Combustion and reforming mechanism (CRR) i.e. all the permeated oxygen is used up by the complete combustion of methane therefore there is a sharp increase in the CH_4 conversion of ceramic

membrane (Wang et al., 2003) while in the case of synthetic membrane CH_4 conversion increases with increasing temperature due to endothermicity of the reaction system. The general trend is smoothly decreasing with time due to carbon deposition and correspondingly reduced catalyst activity (Gallucci et al. 2008).

Thus it can be concluded that CO selectivity is better for Ceramic membrane reactors since it achieves 90% saturation in 25 hours while for synthetic membrane the conversion is only 10% (approx). Overall, ceramic membrane reactors were found to be better than synthetic membrane reactor due to effective CO selectivity and CH_4 conversion. Synthetic membrane reactors show poor or negligible selectivity for the observed period of operation. The predicted results were found to be in good agreement with literature information (Gallucci et al., 2008; Tong et al., 2006; Tong et al., 2002).

5. Summary and Outlook

A mathematical model was proposed in the present study to predict conversion of methane to syngas using different types of membrane reactor and simulated using MATLAB©. The model fits quite well with the experimental data at certain operating conditions i.e. temperature range between 800 -900°C for some experimental conditions. Ceramic membrane reactors require a initiation time of about 25 hrs which is in agreement with the literature results. The CO selectivity of synthetic membrane reactors are dependent on temperature and thus have little or negligible effect with respect to time of operation. It is proposed on the basis of derived equations and the simulation results, a membrane reactor setup can be used which has CH_4 conversion ability of synthetic membrane and the CO selectivity of ceramic membrane. The setup can be assembled either by setting a synthetic membrane reactor in series with the ceramic membrane reactor or by making a membrane by making alloy of the synthetic and the ceramic material to use in the reactor for partial oxidation of methane. The mathematical model would act as a helping hand in the further works related to membrane reactors since it would provide an idea regarding the expected results thus saving time and energy.

Supporting Information:
MATLAB Coding :

(A) CO selectivity:

```
%Function file for selectivity of synthetic MRs
function dcdt = synselectivity(t,c)
A= 0.0527291811;
B= 0.0053867742*c;
dcdt= A+B

%Function file for selectivity of Ceramic MRs
function dcdt = selectivity(t,c)
A= 16.44383286;
B= 0.167771*c;
dcdt= A-B
```

Program:

```
tspan = [0 50];
c0= 0;
fname= 'selectivity';
[t,c]= ODE45(fname, tspan, c0);
plot(t,c);
hold on
tspan=[0 50];
c0=0;
fname='synselectivity';
[t,c]=ODE45(fname,tspan,c0);
plot (t,c, 'r');
```

(B) CH_4 Conversion:

```
%Function File for CH4 conversion in Synthetic MRs
function dmdt = syntheticconversion(t,m)
A=13.43801212;
B=0.0727502319*m;
dmdt=A-B

%Function File for CH4 conversion in Ceramic MRs
function dmdt = ceramicconversion(t,m)
A=45.74085065;
B=0.4591554759*m;
dmdt=A-B;
```

Program:

```
tspan = [0 50];
m0 = 0;
fname = 'ceramicconversion';
[t,m] = ODE45(fname,tspan,m0);
plot (t,m);
hold on
tspan=[0 50];
m0=0;
fname='syntheticconversion';
[t,m]=ODE45(fname,tspan,m0);
plot (t,m, 'r');
```

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