Numerical Analysis for Real-time Nonlinear Model Predictive Control (NMPC) of Ethanol Steam Reformers

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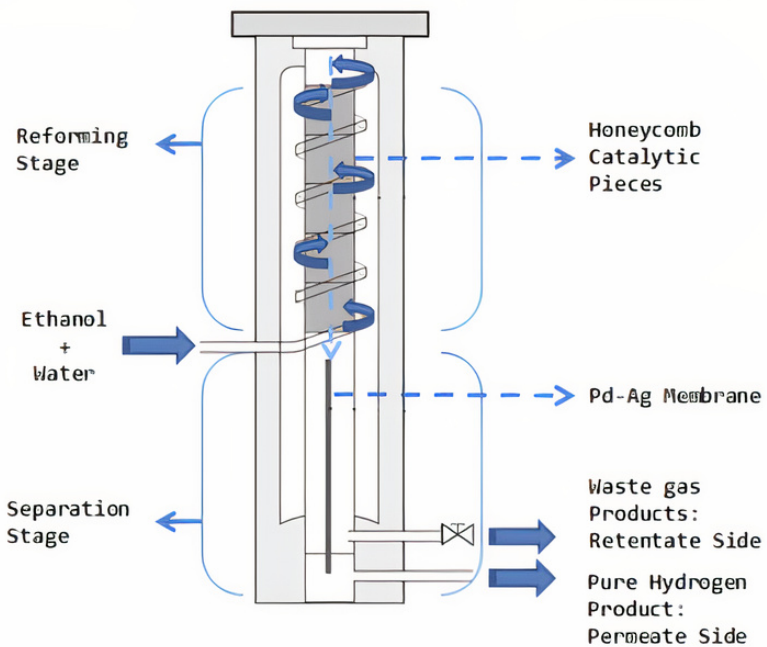
Outline

Motivation for Ethanol Steam Reformers (ESR)

- The use of **renewable energy** technology, notably hydrogen, has witnessed a surge in interest and spread over the world.
- Fluid fuels have far higher energy densities than batteries.
- Hydrogen **storage** and **transportation** has been a limitation
- **Ethanol Reforming** (Existing Technology and Infrastructure)



Chemical Equations

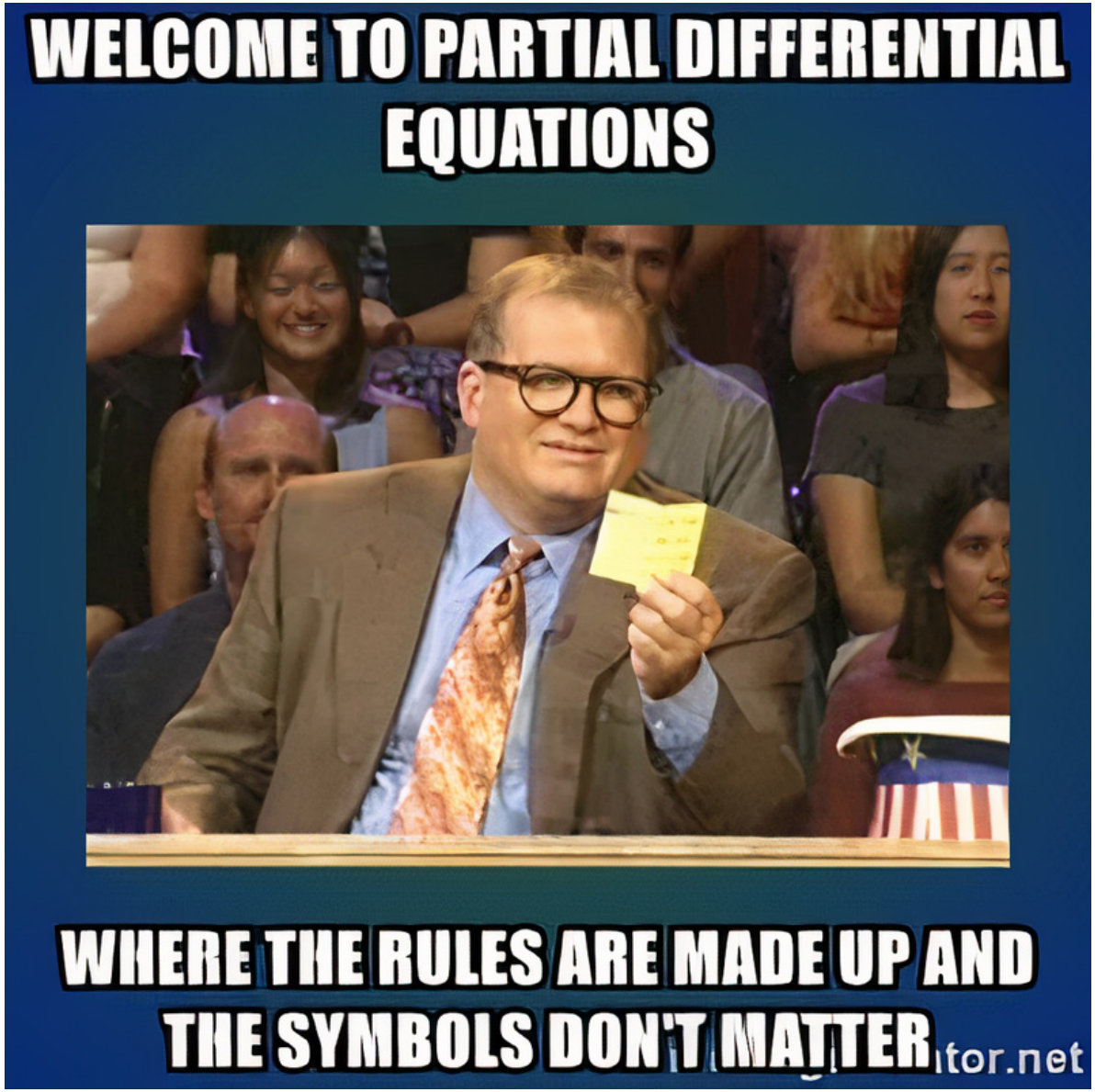
Chemical reactions take place in a tubular packed-bed reactor with a single intake and output. There are 4 primary chemical reactions over cobalt-based catalysts that take place in the staged membrane reactor are as follows

$$\mathrm{C_2H_5OH} \longrightarrow \mathrm{CH_3CHO} + \mathrm{H_2}, \tag{1a}$$

$$\mathrm{C_2H_5OH} \longrightarrow \mathrm{CO} + \mathrm{CH_4} + \mathrm{H_2}, \tag{1b}$$

$$\mathrm{CO} + \mathrm{H_2O} \rightleftharpoons \mathrm{CO_2} + \mathrm{H_2}, \tag{1c}$$

$$\mathrm{CH_3CHO} + 3\mathrm{H_2O} \longrightarrow 2\mathrm{CO_2} + 5\mathrm{H_2}. \tag{1d}$$


WELCOME TO PARTIAL DIFFERENTIAL EQUATIONS
WHERE THE RULES ARE MADE UP AND THE SYMBOLS DON'T MATTER

Molar Conservation Equations

For the reforming stage, the molar conservation equations for the seven $j \in (1, \ldots, 7)$ species $(\mathrm{C_2H_5OH}, \mathrm{H_2O}, \mathrm{CH_4}, \mathrm{H_2}, \mathrm{CO}, \mathrm{CO_2}, \mathrm{CH_3CHO})$ are described by a set of seven nonlinear differential equations, initial conditions, and boundary conditions

$$
\begin{aligned}
& p\frac{\partial F_j}{\partial t} - p\frac{F_j}{\sum_i F_i}\sum_i \frac{\partial F_i}{\partial t} - \frac{p}{T}F_j\frac{\partial T}{\partial t} = RT\sum_i F_i\left(\sum_i \nu_{j,i} r_i\left(\{F_j\}\right) - \frac{1}{A}\frac{\partial F_j}{\partial z}\right) \\
& F_j(z,0) = F_{j,0}(z), \quad \forall z \in [0, \ell_1] \\
& F_j(0,t) = F_{j,\,\mathrm{in}}\,(t), \quad \forall t \geq 0
\end{aligned}
\tag{1}
$$

Energy Conservation Equation

The spatiotemporal dynamics of the ESR temperature are described by the energy conservation equation along with the initial and boundary conditions

$$\frac{p}{RT}\frac{\sum_j c_{v_j} F_j}{\sum_i F_i}\frac{\partial T}{\partial t} = U\beta\left(T_f - T\right) - \frac{RT}{A}\sum_j \frac{\partial F_j}{\partial z}$$

$$-\frac{1}{A}\left(\sum_j c_{p_j} F_j\right)\frac{\partial T}{\partial z} - \sum_j\sum_i r_i\left(\{F_j\}\right)\left(\Delta H_i - \nu_{j,i} RT\right)$$

$$T(z,0) = T_0(z), \quad z \in [0, \ell_1]$$
$$T(0,t) = T_{\text{in}}(t), \quad \forall t \geq 0$$

Conservation Law

For a given physical domain, conservation laws are frequently expressed in integral form. Assume we have a physical domain, Ω, with a domain boundary, $\partial\Omega$. Then, assuming that the physical domain is fixed, the canonical conservation equation is of the type

$$\frac{d}{dt}\int_{\Omega} U d\mathbf{x} + \int_{\partial\Omega} \mathbf{F}(U)\cdot\mathbf{n} ds = \int_{\Omega} S(U,t) d\mathbf{x}$$

where U is the conserved state, $\mathbf{F}$ is the flux of the conserved state, $\mathbf{n}$ is the outward pointing unit normal on the boundary of the domain, and S is a source term. Using Gauss's theorem, this conservation law may be represented as a partial differential equation(usually a hyperbolic system) as follows

$$\frac{\partial U}{\partial t} + \nabla\cdot\mathbf{F} = S$$

Molar Conservation Law

At any given point, the concentration of the j th species, C_j, and the flow's velocity, v, can be computed a

$$C_j = \frac{F_j}{\sum_i F_i} \frac{p}{RT}$$

$$v = \frac{1}{A} \sum_i F_i \frac{RT}{p}$$

Now notice we can transform the Molar Conservation Equation into a conservation equation by using C_j and v as defined above.

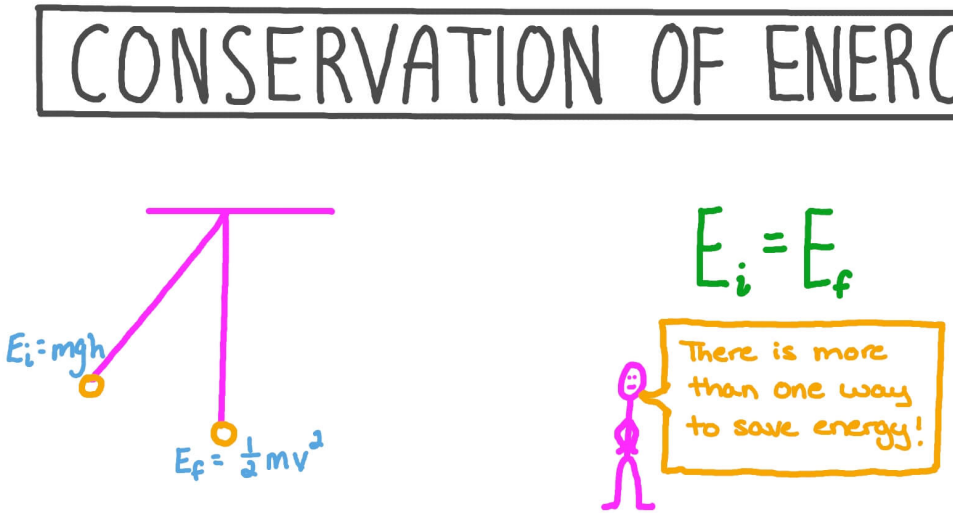
$$\frac{\partial \mathrm{C_j}}{\partial t} + \frac{\partial \left(v C_j\right)}{\partial z} = \sum_i v_{j,i} r_i,$$

$$\mathrm{C_j}(0, z) = \mathrm{C_{j,0}}(z), \quad \forall z \in [0, l_1]$$

$$\mathrm{C_j}(t, 0) = \mathrm{C_{j,\ in}}\ (t), \quad \forall t \geq 0$$

which is in a conservation form.

Energy Convection Form



Sadly it was not possible: :(

Energy Convection Form

However it was transformed into a convection law by converting the following equation

$$\rho_g c_{p_g} \left(v \frac{\partial T}{\partial z} + \left(1 + \frac{\rho_s c_{p_s}}{\rho_g c_{p_g}} \right) \frac{\partial T}{\partial t} \right) = U a \left(T_f - T \right) + H_r$$

to

$$\frac{\partial g}{\partial t} + v \beta^{-1} \frac{\partial \left(g \right)}{\partial z} = \left(U a \left(g_f - \frac{g}{k} \right) + H_r \right) \frac{1}{\rho_g c_{p_g}} \frac{1}{k} \beta^{-1}$$

$$H_r = \sum_{j=1}^{N} -\Delta H_j r_j \quad j = 1, 2, \ldots, 4$$

$$g(0, z) = g_0(z) \quad z \in [0, L]$$

$$g(t, 0) = g_{in}(t) \quad \forall t > 0$$

Singular Partial Differential Equation System

The systems of singular partial differential equations of the ESR can be written in the form

$$Mf_t + Nf_z = g(t,z,f) \quad M = \begin{pmatrix} I - \frac{1}{\|F\|} F e^\top & -\frac{1}{T} F \\ 0^\top & \frac{C_v}{RT\|F\|} F \end{pmatrix}$$

$$f := \begin{pmatrix} F \\ T \end{pmatrix}, \quad F = \begin{pmatrix} F_1 \\ \vdots \\ F_7 \end{pmatrix} \quad N = \begin{pmatrix} \frac{RT}{Ap} I & 0 \\ \frac{RT}{Ap} e^\top & \frac{C_p}{Ap} F \end{pmatrix}$$

$$g = \begin{pmatrix} RT[F_1, F_1, \ldots F_1] F V_1 R' \\ RT[F_2, F_2, \ldots F_2] F V_2 R' \\ \vdots \\ RT[F_7, F_7, \ldots F_7] F V_7 R' \\ U\beta(T_f - T) - \sum_i F_i [R' H' - V_i R']^T \end{pmatrix}$$

Reformulation

The above system can also be reformulated by writing the vector f in terms of an orthonormal basis $\|F\| = \sqrt{7}u_7$

$$A = \begin{pmatrix} 1 & -\frac{u_1}{u_7} & -\frac{u_1}{T} \\ \ddots & \vdots & \vdots \\ 1 & -\frac{u_6}{u_7} & -\frac{u_6}{T} \\ & 0 & -\frac{u_7}{T} \\ & 0 & \frac{C_v^u U}{\sqrt{7}RTu_7} \end{pmatrix} \quad B = \begin{pmatrix} \frac{RT}{Ap} & 0 & 0 \\ \ddots & \vdots & \vdots \\ 0 & \frac{RT}{Ap} & 0 \\ 0 & \frac{\sqrt{7}RT}{Ap} & \frac{C_v^u U}{Ap} \end{pmatrix}$$

$$g = \begin{pmatrix} RT[u_1, u_1, \ldots u_1]UV_1R' \\ RT[u_2, u_2, \ldots u_2]UV_2R' \\ \vdots \\ RT[u_7, u_7, \ldots u_7]UV_7R' \\ U\beta(T_f - T) - \sum_i u_i[R'H' - V_iR']^T \end{pmatrix} \quad U = \begin{pmatrix} u_1 \\ \vdots \\ u_7 \end{pmatrix}$$

Method of Characteristics

The primary concept of the method of characteristics is to convert a partial differential equation to an ordinary differential equations using characteristic curves which enables us to then solve the PDE. The equations can be written as a singular system of quasi-linear hyperbolic equations

$$A(x, y, u)u_x + B(x, y, u)u_y = g(x, y, u)$$

Multiplying both sides of the equation by a row vector to be determined later gives

$$l^\top A u_x + l^\top B u_y = l^\top g$$

Assume that there exists an ordered pair $(x_{\omega'}, y_\omega)$ parameterized by a real scalar ω^*

$$\left(\frac{1}{x_\omega}\boldsymbol{l}^\top A\right) u_x x_\omega + \left(\frac{1}{y_\omega}\boldsymbol{l}^\top B\right) u_y y_\omega = \boldsymbol{l}^\top g$$

Method of Characteristics (cont'd)

If (x_ω, y_ω, l) can be found that satisfy $\frac{1}{x_\omega}\boldsymbol{l}^\top A = \frac{1}{y_\omega}\boldsymbol{l}^\top B$ then $\left(\frac{1}{x_\omega}\boldsymbol{l}^\top A\right)(u_x x_\omega + u_y y_\omega) = \boldsymbol{l}^\top g$. After some simplifications and by using the chain rule, these expressions can be rewritten as

$$l^\top A \frac{du}{dx} = l^\top g$$

$$l^\top B \frac{du}{dy} = l^\top g$$

In other words, u is related to x and y by ordinary differential equation which can be found if we find a way to satisfy $\frac{1}{x_\omega}\boldsymbol{l}^\top A = \frac{1}{y_\omega}\boldsymbol{l}^\top B$. Thus multiplying by x_ω gives $l^\top(A - \sigma B) = 0$ where $\sigma = \frac{x_\omega}{y_\omega} = \frac{dx}{dy}$

ODE system

Finally we have

$$\frac{\mathrm{d}x}{\mathrm{d}t} = \frac{1}{\sigma_k(x,t,u)}$$
$$\boldsymbol{l}_k(x,t,u)^\top A(x,y,u)\frac{\mathrm{d}x}{\mathrm{d}t} = \boldsymbol{l}_k(x,t,u)^\top g(x,t,u)$$
$$k = 1,2,\ldots,m$$

which is much easier to solve. Finding σ_k and l_k can be found by first finding the eigenvalues by finding the analytic solution to $\det(A-\sigma B)=0$ and then using the eigenvalues σ_k to find out the generalized eigenvectors l_k by solving $l^\top(A-\sigma B)=0$.

Generalized Eigenvalues and Eigenvectors of our System

In particular, the analytical solution to our reformulated system $\det(A - \sigma B) = 0$ is

$$\sigma_k = \frac{Ap}{RT}, \quad k = 1, \cdots, 6,$$

$$l_1 = \begin{pmatrix} 1 \\ 0 \\ \vdots \\ 0 \end{pmatrix}, \ldots, \boldsymbol{l}_6 = \begin{pmatrix} 0 \\ \vdots \\ 0 \\ 1 \\ 0 \\ 0 \end{pmatrix}$$

and

$$\sigma_7 = 0, \quad \sigma_8 = \frac{Ap}{RTC_p^u U}\left(\frac{C_v^u U}{\sqrt{7}u_7} - \sqrt{7}Ru_7\right)$$

Generalized Eigenvalues and Eigenvectors (cont'd)

Closed form solutions of l_7 and l_8

$$\boldsymbol{l}_7 = \begin{pmatrix} \frac{u_1}{u_7} \\ \frac{u_2}{u_7} \\ \frac{u_3}{u_7} \\ \frac{u_4}{u_7} \\ \frac{u_5}{u_7} \\ \frac{u_6}{u_7} \\ 1 \\ 0 \end{pmatrix} \quad \boldsymbol{l}_8 = \begin{pmatrix} \frac{7u_1\left(-C_u^v C_u^p UU^\top u_7 + \sqrt{7}{C_u^p}^2 UU^\top u_7^2 - 7C_u^p RUu_7^3\right)}{T\left(C_u^v U + 7Ru_7^2\right)\left(\sqrt{7}C_u^v U - 7C_u^p Uu_7 + 7\sqrt{7}Ru_7^2\right)} \\ \frac{7u_2\left(-C_u^v C_u^p UU^\top u_7 + \sqrt{7}{C_u^p}^2 UU^\top u_7^2 - 7C_u^p RUu_7^3\right)}{T\left(C_u^v U + 7Ru_7^2\right)\left(\sqrt{7}C_u^v U - 7C_u^p Uu_7 + 7\sqrt{7}Ru_7^2\right)} \\ \frac{7u_3\left(-C_u^v C_u^p UU^\top u_7 + \sqrt{7}{C_u^p}^2 UU^\top u_7^2 - 7C_u^p RUu_7^3\right)}{T\left(C_u^v U + 7Ru_7^2\right)\left(\sqrt{7}C_u^v U - 7C_u^p Uu_7 + 7\sqrt{7}Ru_7^2\right)} \\ \frac{7u_4\left(-C_u^v C_u^p UU^\top u_7 + \sqrt{7}{C_u^p}^2 UU^\top u_7^2 - 7C_u^p RUu_7^3\right)}{T\left(C_u^v U + 7Ru_7^2\right)\left(\sqrt{7}C_u^v U - 7C_u^p Uu_7 + 7\sqrt{7}Ru_7^2\right)} \\ \frac{7u_5\left(-C_u^v C_u^p UU^\top u_7 + \sqrt{7}{C_u^p}^2 UU^\top u_7^2 - 7C_u^p RUu_7^3\right)}{T\left(C_u^v U + 7Ru_7^2\right)\left(\sqrt{7}C_u^v U - 7C_u^p Uu_7 + 7\sqrt{7}Ru_7^2\right)} \\ \frac{7u_6\left(-C_u^v C_u^p UU^\top u_7 + \sqrt{7}{C_u^p}^2 UU^\top u_7^2 - 7C_u^p RUu_7^3\right)}{T\left(C_u^v U + 7Ru_7^2\right)\left(\sqrt{7}C_u^v U - 7C_u^p Uu_7 + 7\sqrt{7}Ru_7^2\right)} \\ \frac{\sqrt{7}C_u^p Uu_7^2}{T\left(C_u^v U + 7Ru_7^2\right)} \\ 1 \end{pmatrix}$$

Uniqueness Theorem for First Order Quasi Linear Systems

The following uniqueness theorem remains valid for quasi-linear systems of first order

$$u_t + B(x,t,u)u_x + g(x,t,u) = 0 \tag{5}$$

even though the characteristic lines C_k of the above equation depend on the solution u. We assume here that the matrices B and g possess continuous derivatives with respect to x, t, and u in the region under consideration. Then as usual we consider the following equations

$$u_t + B(x,t,u)u_x + g(x,t,u) = 0$$
$$u(x,0) = \psi(x)$$

and

$$v_t + B(x,t,v)v_x + g(x,t,v) = 0$$
$$v(x,0) = \psi(x)$$

Uniqueness Theorem (cont'd)

Subtracting v from u and denoting this by z ie $(z = u - v)$ we get

$$z_t + B(v)z_x + [B(u) - B(v)]u_x + g(u) - g(v) = 0$$
$$z(x,t) = 0$$

Because both B and g are differentiable and continuous we may apply the mean value theorem

$$B(u) - B(v) = H(u,v)z; \quad g(u) - g(v) = K(u,v)z$$

where H, K are continuous functions. We now consider u, v, u_x as known expressions in x, t and substitute these expressions in H and K as well as in $B(v)$; thereby becomes a linear homogeneous differential equation for z of the form

$$z_t + bz_x + gz = 0$$

with initial values zero and the uniqueness theorem is proved for Linear systems (in the paper). Hence proved

Existence Theorem for First Order Quasi Linear Systems

The existence theorem is a bit more complicated to present here but the whole gist of it boils down to

- creating an iteration scheme and trying to show that the sequence u_n converges uniformly to the function u in a specific region G_h in the closed domain G.
- The solution then can be extended into the whole domain G as long as the coefficients retain their continuity properties throughout the whole domain.

Numerical Analysis

Now there are two directions on how we can proceed with the numerical analysis

- ODE System solvers - Euler Methods, Runge-Kutta Method, IDA (implicit backward differentiation formulas for DAEs) etc
- PDE System solvers - Finite Element Methods

ODE System solvers

We first try to consider a subsystem for simplicity ie we consider a 3 system equation with only 2 molar equations and the same temperature equation as follows (for simplicity) where $U = (u_1, u_2)$ and $u = (u_1, u_2, T)$

$$A = \begin{pmatrix} 1 & -\frac{u_1}{u_2} & -\frac{u_1}{T} \\ 0 & 0 & -\frac{u_2}{T} \\ 0 & 0 & \frac{C_v^u U}{\sqrt{2}RTu_2} \end{pmatrix} \quad B = \begin{pmatrix} \frac{RT}{Ap} & 0 & 0 \\ 0 & \frac{RT}{Ap} & 0 \\ 0 & \frac{\sqrt{2}RT}{Ap} & \frac{C_v^u U}{Ap} \end{pmatrix}$$

$$g = \begin{pmatrix} RTu_1 U V_1 R' \\ RTu_2 U V_2 R' \\ U\beta(T_f - T) - \sum_i u_i [R'H' - V_i R']^T \end{pmatrix}$$

Numerical Solution

Using Mathematica we code up the simplified system and the eigenvectors and eigenvalues of the system are as follows

$$\text{Out[6]=}\ \left\{0,\ \frac{A\,p}{R\,T[x]},\ \frac{A\,p\left(\frac{c\,U}{\sqrt{2}\,u2[x]}+\sqrt{2}\,R\,u2[x]\right)}{c1\,R\,U\,T[x]}\right\}$$

$$\text{Out[7]=}\ \left\{\left\{\frac{u1[x]}{u2[x]},\ 1,\ 0\right\},\ \{1,\ 0,\ 0\},\ \left\{\frac{2\,u1[x]\left(-c\,c1\,U^2\,u2[x]+\sqrt{2}\,c1^2\,U^2\,u2[x]^2-2\,c1\,R\,U\,u2[x]^3\right)}{T[x]\left(c\,U+2\,R\,u2[x]^2\right)\left(\sqrt{2}\,c\,U-2\,c1\,U\,u2[x]+2\sqrt{2}\,R\,u2[x]^2\right)},\ -\frac{\sqrt{2}\,c1\,U\,u2[x]^2}{T[x]\left(c\,U+2\,R\,u2[x]^2\right)},\ 1\right\}\right\}$$

Figure: Eigenvalues and Eigenvectors of the simplified system

where the generalized eigenvalue and eigenvectors have the same structure as our original reformulated system. Note the constants have been renamed to c, c_1.

Numerical Solution (cont'd)

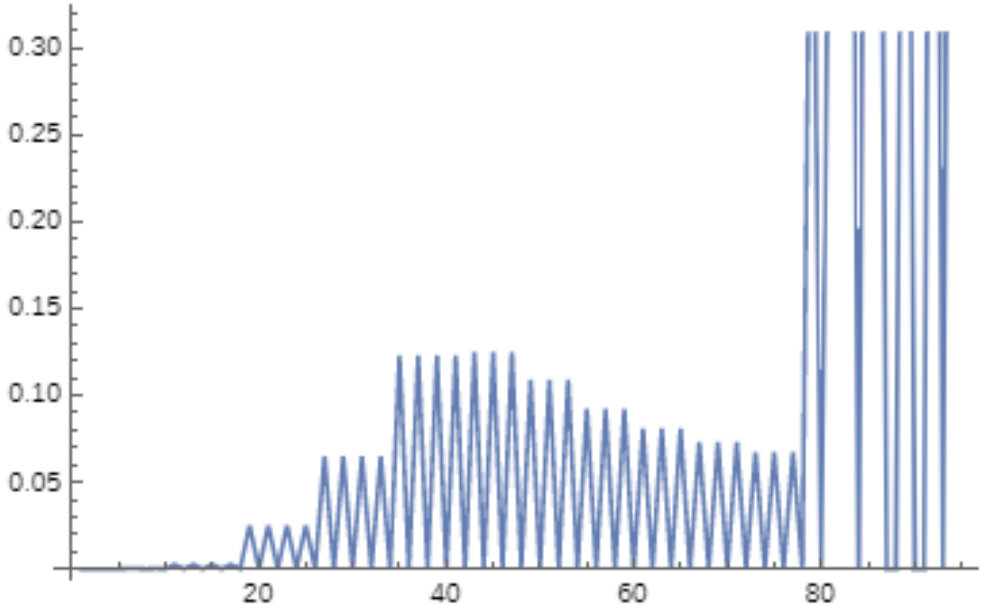
1. We now proceed to put the values for the constants (R, T_f, β, stoichiometric coefficients and etc) where some of the constants have the true values and others have been randomized.
2. We should also note that one of the eigenvalues in our system is 0 which is a point of singularity. This was replaced by $\epsilon = 0.00001$ just to analyse the system better.
3. Finally we use set all the boundary conditions and initial conditions to small but randomized values
4. We set the independent variable x to be solved in the range $[\epsilon, 10]$

Analysis

We use Mathematica's NDSolve feature and do not specify any explicit numerical method to solve the above system. We define MLR (Molar Flow Rate) and some of the results are as follows

1. It takes around 96 evaluations for the solver to converge and stop

Plotting the distance between successive evaluations results in



Solution Plots

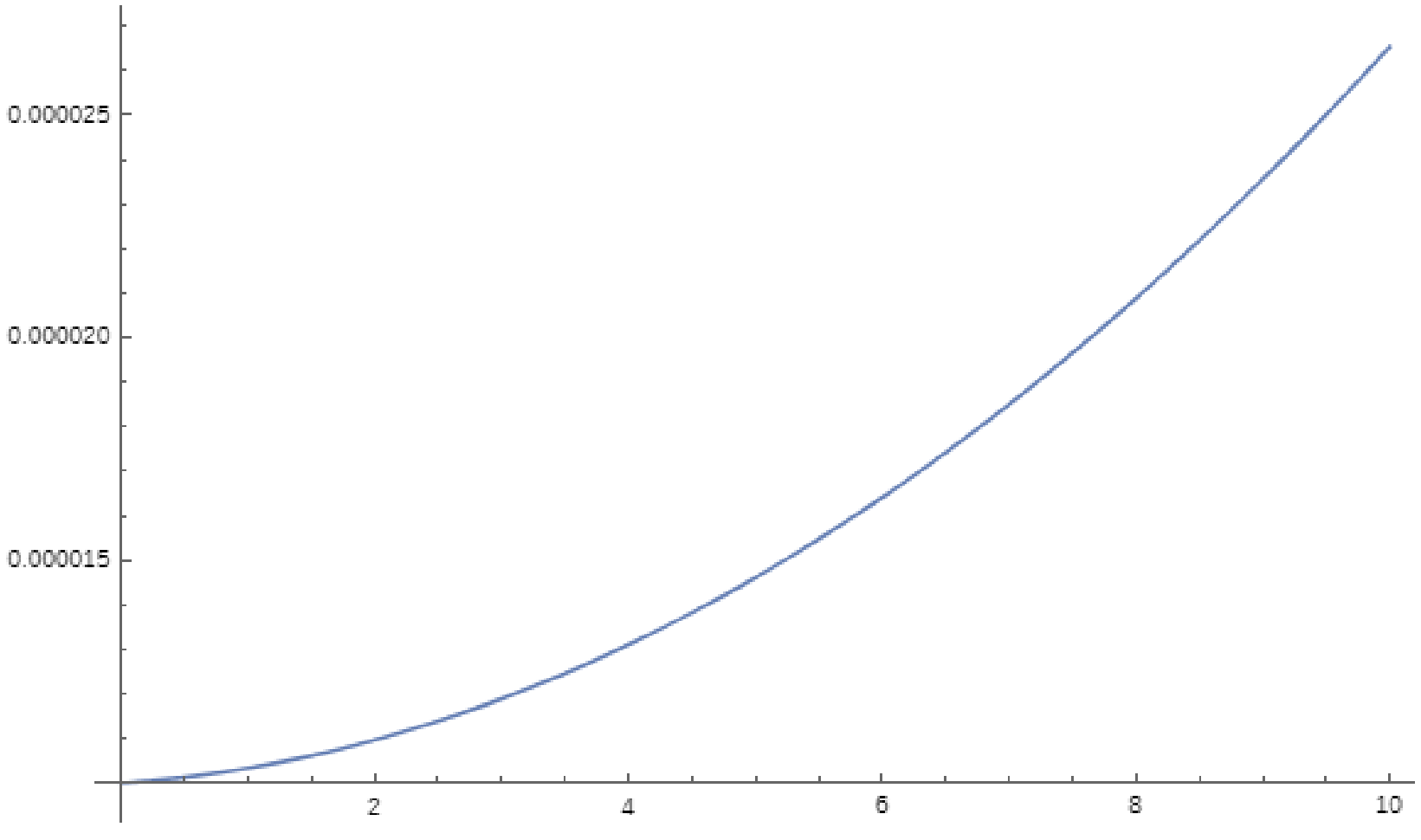


Figure: $dy/dx = 1/\sigma_1$

Solution Plots

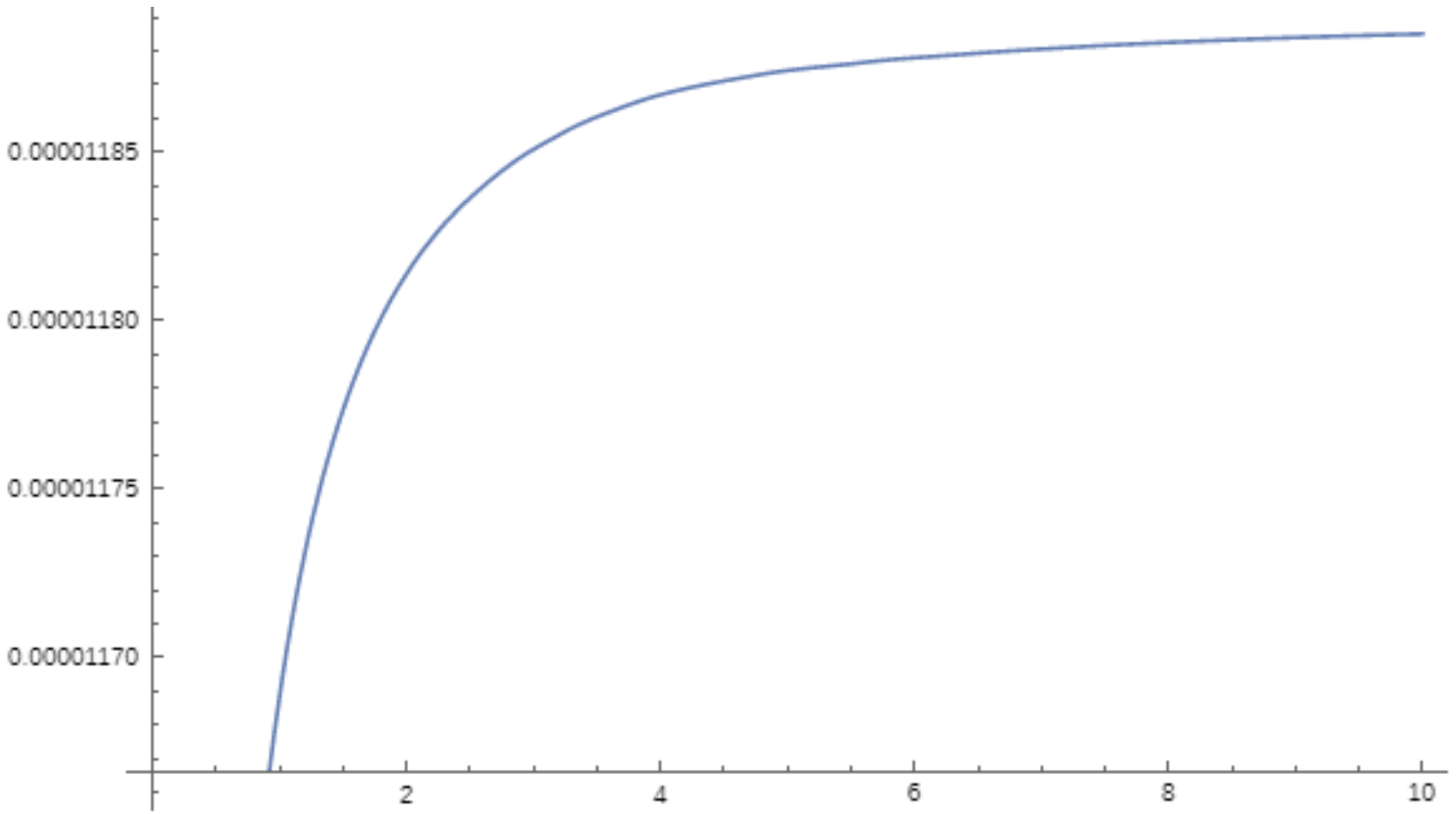


Figure: MLR vs $\boldsymbol{l}_1(x,y,u)^\top A(x,y,u)\frac{\mathrm{d}u}{\mathrm{d}x} = \boldsymbol{l}_1(x,y,u)^\top g(x,y,u)$

Final Plot

Plotting all the solutions results (when extrapolated backwards in the negative x axis) which shows the various discontinuities when $x < 0$

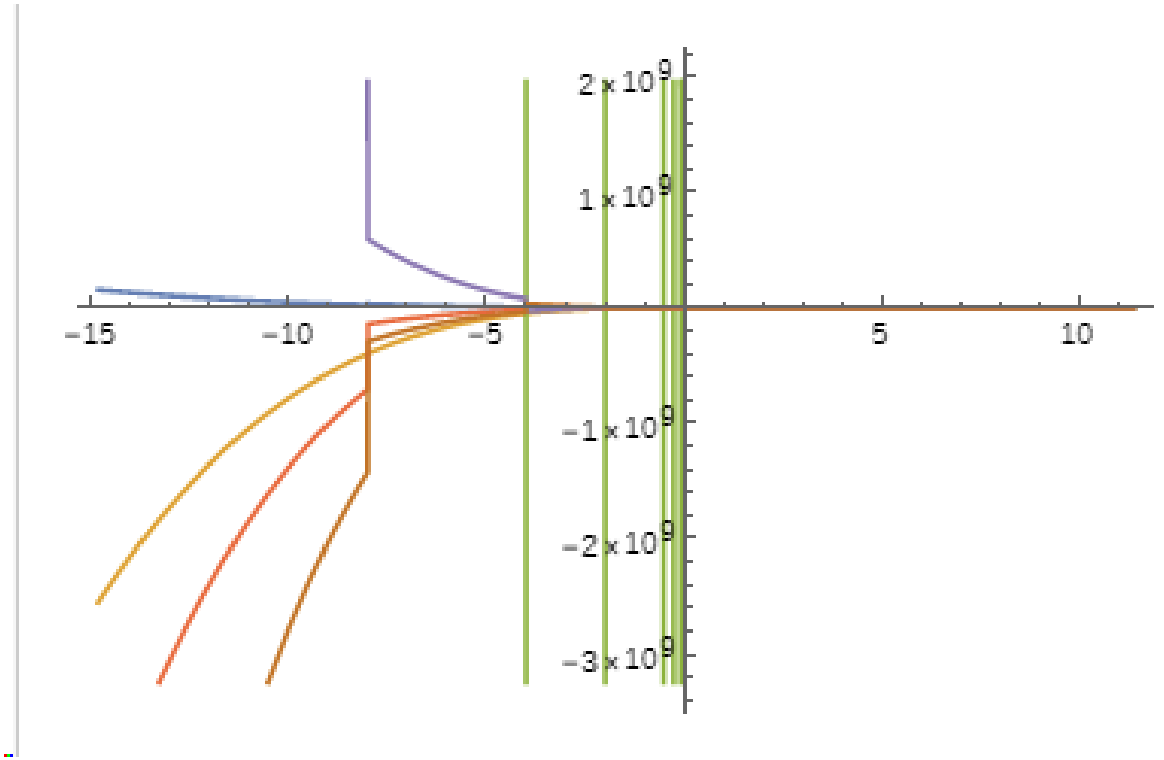


Figure: All solutions $x < 0$

Comparison

When we only consider from the positive x - axis we get the following plot

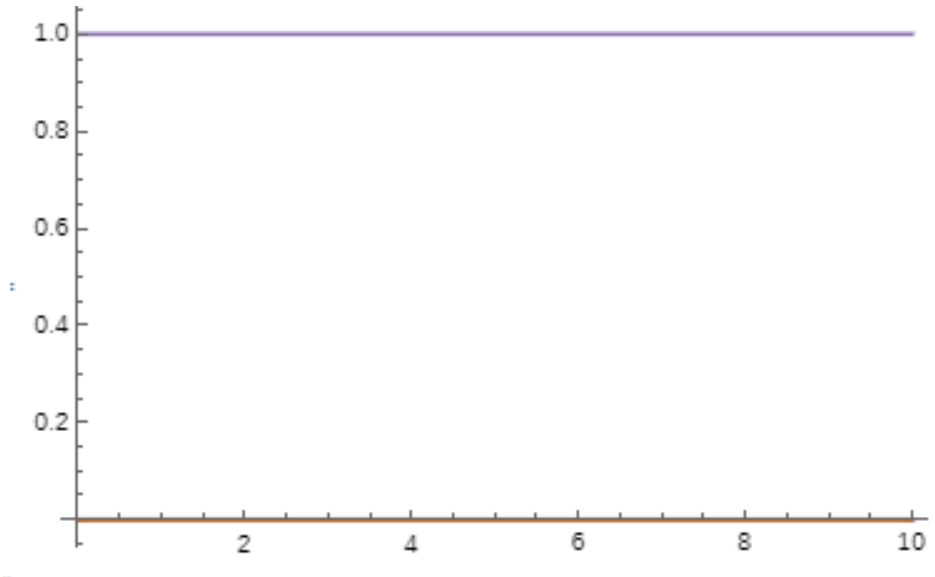
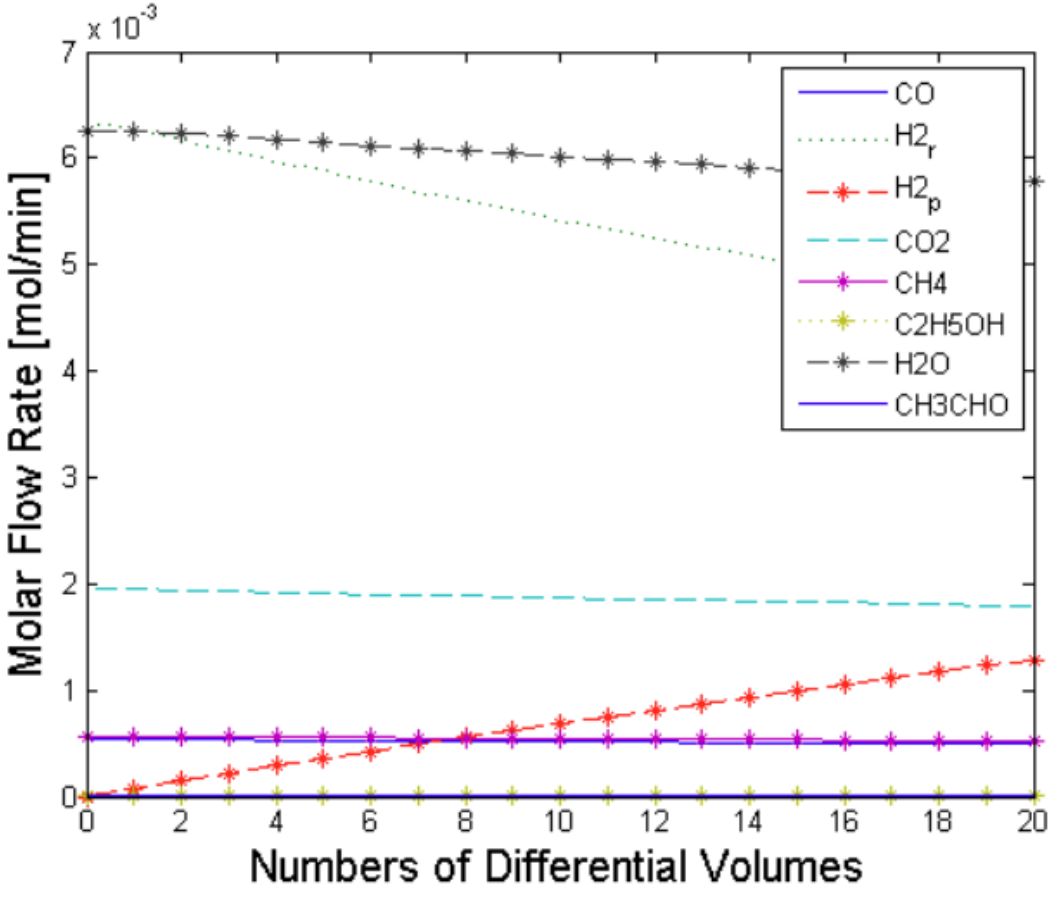


Figure: All solutions $x > 0$

which is consistent when only considering the sub system of equations ie $j = 1, 2$

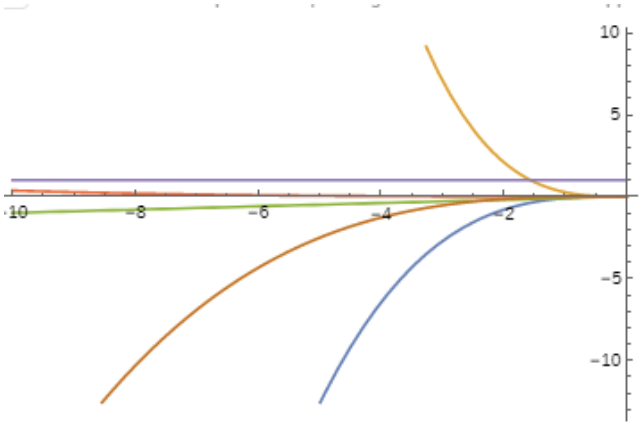
Comparison (cont'd)

This is consistent with the results from the previous paper by observing the Molar Flow Rate of C_2H_5OH and CH_4

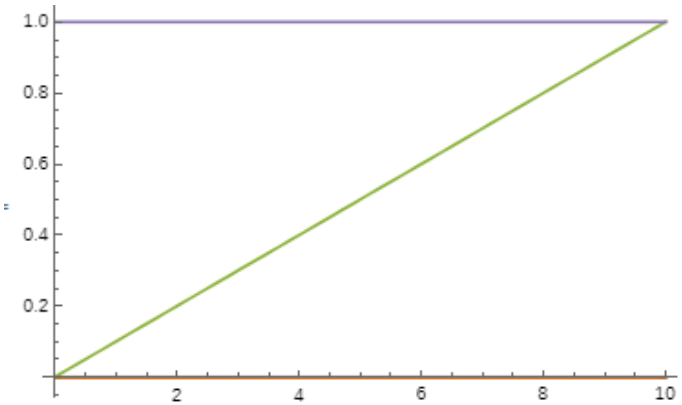


Epsilon value change

Changing the value of $\epsilon > 1$ resulted in



(a) $x < 0$



(b) $x > 0$

Symbolic Solution

Tried to use the symbolic solver to find a closed form solution of the simplified system (with $\epsilon = 0.000001$)

```
In[•]:= qns = {eqns1, eqns2, eqns3, eqns4, eqns5, eqns6, eqns7, eqns8, eqns9, eqns10, eqns11, eqns12, eqns13, eqns14, eqns15, eqns16, y1[0] == 0, y2[0] == 0,
    y3[eps] == 0, y4[eps] == eps, y5[eps] == eps, y6[eps] == eps, y7[0] == 0, y8[0] == 0, u1[eps] == 1, u2[eps] == eps, u3[eps] == eps, u4[eps] == eps,
    u5[0] == eps, u6[0] == eps, u7[eps] == eps, T[0] == eps};
  sol = NDSolve[qns, {y1, y2, y3, y4, y5, y6, y7, y8, u1, u2, u3, u4, u5, u6, u7, T}, {x, eps, 1000}];

  ••• NDSolve : Cannot solve to find an explicit formula for the derivatives. Consider using the option Method->{"EquationSimplification"->"Residual"}.
```

Upon debugging the stack trace message showed this

> In some versions of the Wolfram Language, an error occurs because this differential equation does not have a solution:

It was worth the try, I also considered just a 2 by 2 system and a closed form solution was found (Maybe due to the fact that there was no singularity in a 2 by 2 system)

Limitations

There were certain limitations and assumptions that were made due to lack of time, some of them were

- The whole 16 equations were programmed, but the solver wasn't able to figure out even a numerical solution to the whole system (Mainly due to the randomized values that were set for the boundary and initial conditions)
- Could not completely code up the any of the PDE solvers
- Our system has a singularity point due to the fact that $\det(A) = 0$, and so we need to do more theoretical analysis on how it behaves.
- All the constants were randomized, need a more controlled setting to set out the constants as accurately as possible relating to the reactants

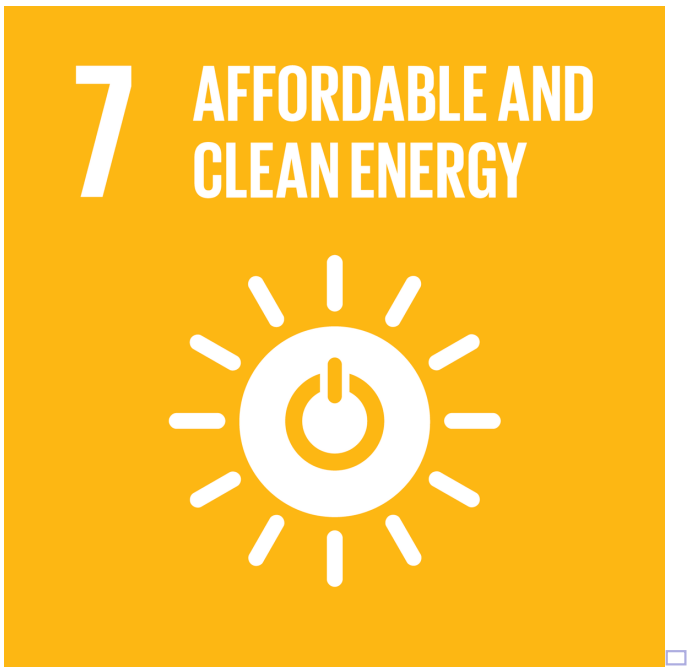
Future work

Some of the possible future work include

- An indepth study of the system (more theoretical Analysis)
- More research on the various numerical methods which can help us to solve this system faster as well as more numerical analysis.
- Generalize this approach by incorporating these ode solvers into Nonlinear Model Predictive control algorithms which can be used for other Nonlinear systems.

Applications

- To use the above ODE solvers which can be fed into a NMPC which could enable a mechanistic model to be used in **real-time** control calculations with **minimal** online computational cost.
- This would allow us to **manufacture hydrogen safely** from ESR, which could be utilized as green energy.



Conclusions

To conclude, some of the results are as follows

- Try to convert the equations into some form of conservation law (The energy conservation equation could not be converted but rather put into a convection form)
- Solution of the system
- Uniqueness and Existence Theorems for Linear/Quasi-Linear First Order PDE Systems.
- Closed form solutions of all the eigenvalues and eigenvectors of both the reformulated and original system
- Numerical Analysis of a simplified system in hopes of extending it to the whole system

References

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Thanking remarks