DESCRIPTION OF A COMPREHENSIVE MATHEMATICAL MODEL: TOWARDS A COMPREHENSIVE BIOMASS PARTICLE GASIFICATION MODEL

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ABSTRACT: Biomass gasification is still a promising technology after over 30 years' research and development and has success only in a few niche markets. In this paper, a comprehensive mathematical model for biomass particle gasification is developed within a generic particle framework, assuming the feed is a woody biomass pellet. The particle is discretized into a number of cells along radical direction, on each of which the gas governing equations are numerically solved by using the finite volume method (FVM). FVM is used to solve transport equations of each process. All the key processes, e.g., moisture evaporation, pyrolysis, heterogeneous char reactions, intra-particle heat and mass transfer, and changes in thermos-physical properties and so on, are properly taken into account to update the densities of various solid/liquid components in each cell as well as to provide source terms to the relevant gas-phase governing equations. For the source term, different chemical reactions are assumed, the density, the species, the velocity, the temperature changes, the size of pellet, et al. will be studied and calculated in the end of the project.
Keywords: Biomass, Gasification, modelling.

1 INTRODUCTION

Biomass gasification has received unprecedented attention, as a renewable source of energy and chemicals. In biomass gasification, biomass pyrolysis is a key, inseparable initial sub-process, resulting in a complex system of intermediate products (or product groups: gases, tars and char). Then the gases and tar vapors undergo further gas-phase reactions (e.g., cracking, reforming, shift) whilst the char takes part in heterogeneous reactions, yielding the final gasification products. Partly due to government regulation and partly due to unsolved technological hurdles (e.g., lack of reliable modelling tool for biomass pyrolysis/gasification, lack of dominant design, fuel flexibility, efficiency, tar reduction, gas cleaning, scaling up), biomass gasification is still a promising technology after over 30 years' research and development (R&D) and has success only in a few niche markets [1].

On the other hand, with the fast consumption of fossil fuels, appealing from the public for looking for renewable resources is stronger and stronger. Biomass, as one of the readily available renewable resources, come to public view.

According to Danish Energy Agency (DEA) [2], the use of wood pellets and wood chips increase significantly this century. In Fig. I and Fig. II, we can see that solid biomass consumption is not only increase in Denmark, but also increase in the whole west Europe. In Fig. II, a prediction of the next ten years was given, we can see in 10 years, demand for wood pellets will be more than doubled, but demand for straw and biodegradable waste will keep almost the as the same stage as now.

Biomass has many benefits. Firstly, biomass is flexibly in technology [3], for biomass gasification on syngas generation, it has a wide option of choosing biomass species. Secondly, Biomass is clean renewable energy [4]. During the gasification, the CO_2, SO_2 and other harmful gas emissions are less compared to fossil fuel, the emissions are less even compared with the emission from producing photovoltaic cell materials [5]. Lastly but not least, the temperature for gasification is relative low, which is around 700℃- 900℃ [6]. Around that temperature, NO_x generation can be ignored.
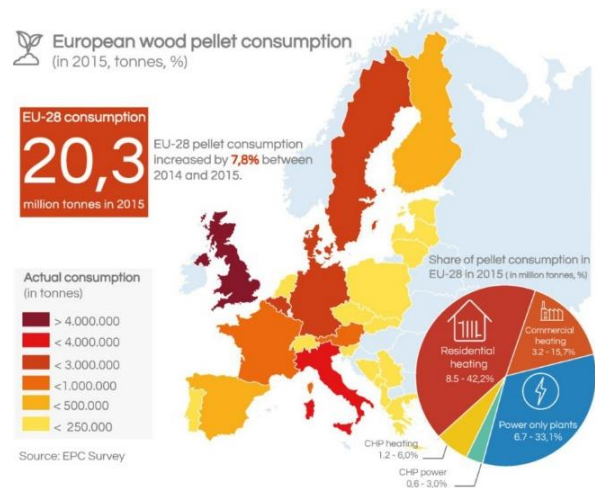

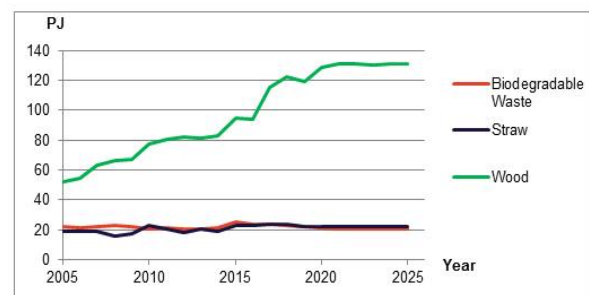**Figure I** Use of solid biomass in Europe in 2015 [7]

Figure II Use of solid biomass in electricity and heat production 2005-2015, and projection to 2025 [3]

There are also some defects using biomass transformation technology. The first and foremost is biomass syngas is a low value fuel (aroud 6～10 MJ/Nm^3) [8]. For syngas, the tar and alkali inside the gas may ruin the device if it goes into gas turbine directly after gasification [9].

As a result of these reasons, there has been a compelling need for reliable, cost-effective modelling tools for the design of biomass pyrolysis/gasification processes and components. Using CFD simulation is one of the most popular methods.

Many scholars worked on this field and gain remarkable achievements. Bamford [10] started to use mathematic method for solving a single sheet of wood combustion problem at an early stage. After that, around eighties and nineties of last century and the beginning of this century, there is a great heat for kinetics study. Font[11][12], Di Blasi [13][14][15][16], Babu [17][18] are all active at working on that. Kinetic equations for mechanism were discussed, experimental data from different scholars using different technologies were compared. Until recently, most kinetic parameters used are from that time [19][20][21]. After the kinetic mechanism study, next step is to build a reliable mathematical model for the whole reaction process. As a young pioneer, Di Blasi started the exploration with using mathematical model [3], actually she stated this based on cellulose pyrolysis [16] and combustion of conceptual solid fuels [15]. She kept modifying this model and use it at different reactors [22]. Other researchers contribute to fertilize biomass gasification, combustion and pyrolysis under different assumptions. Bernhard [23] and Wu[24] studied their objects with solid phase and gas phase separated. An imaginary heat and mass transfer between phases were assumed. Haseli [25] compared his simulation results with former researchers according to different size. H.Fatehi [26] built his model based on one dimentional single large biomass particle combustion without particle size changes. Lu [21] [27][28] did a systematic study based one biomass pellet combustion with a fixed mass and heat transfer coefficient in ambient flow.

In order to make use of these advantages of biomass, as well as to decrease the defects as much as possible, a reliable mathematical model is still in need. This project try to combine the work of predecessors together, at the same time, to remove the defects and to consummate mathematical model. In this article, it mainly focus on consummating source term of reaction and revising transport equations. A brief case of using FVM to solve PDE is given in the end.

2 MODEL DESCRIPTION

2.1 Assumptions:

In order to make the best use of the theories and correlations, as well as to make the simulation results more credible, assumptions are necessary. In this paper, a one-dimensional mathematics model based on a single biomass pellet under gasification is assumed.

(1) Assuming the biomass pellet shape is nearly sphere.
(2) Assuming the biomass particle is in local thermal equilibrium.
(3) During the reaction, the biomass particle is considered as a porous structure media, and the gases generated inside the particle obey ideal gas law.
(4) The specific surface area changes during the whole gasification, thus shrinkage or swelling is also considered.
(5) The airflow inside the particle obeys Darcy law.
(6) Boundary layer thickness exist. And its outer surface properties equal to those in outer spaces, inner surface properties equal to those in the surface of particle.
(7) External forces are not considered.
(8) There is no fragmentation of the particle during the whole gasification.

Fig. III gives out the key issue to be solved in this project. There three key issues, which is very important during the calculation: the heat and mass transfer of boundary between gas phase and solid phase, the conversion process inside the particle itself, and the particle size changes during the whole reaction process.
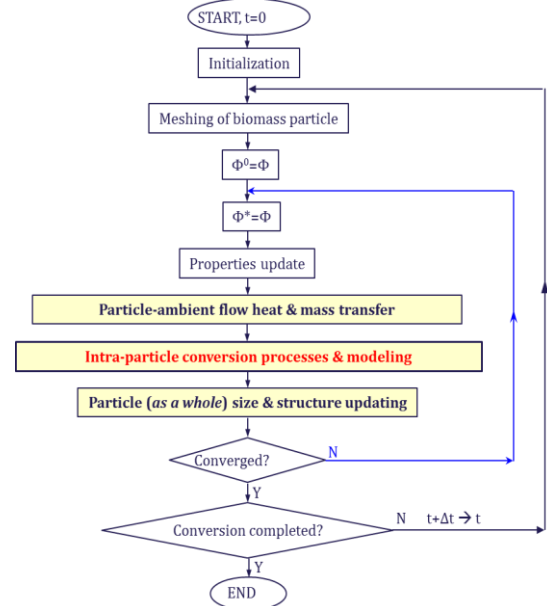

Figure III Key issue to be solved

It is also necessary to assume the right reaction during the gasification process. Thirteen chemical reactions happened inside the pellets are considered, as showing in table I. In table I, the reaction rate is also given. It is worth noting that:

$$K_i = A_i \exp(-E/RT)$$

K_i is the kinetic rate coefficients for all the reactions in table I. A is pre-exponential factor and E is activation energy. It is only dependent on temperature. The whole expression is called as Arrhenius expression.

Table I: Reactions

Reaction index	Chemical reactions	Rate expression	Ref.
1	biomass→gases	$r_1 = \partial \rho_B / \partial t = k_1 \rho_B$	[28]
2	biomass→tar	$r_2 = \partial \rho_B / \partial t = k_2 \rho_B$	[28]
3	biomass→char	$r_3 = \partial \rho_B / \partial t = k_3 \rho_B$	[28]
4	tar→gases	$r_4 = \partial \rho_g / \partial t = k_4 \rho_g Y_T$	
5	tar→char	$r_5 = \partial \rho_c / \partial t = k_5 \rho_g Y_T$	
6	H_2O (free)→ H_2O (g)	$r_6 = \partial \rho_{fw} / \partial t = s_a (\rho_{fw}/\rho_{fw}^0) h_{m,pore} (\rho_\vartheta^{sat} - Y_{V\rho g})$	[28]
7	H_2O(bound)→H_2O (g)	$r_7 = \partial \rho_{bw} / \partial t = k_7 \rho_{bw}$	[29]

8	C+1/2O$_2$→CO	$r_8 = \partial C_{O_2}/\partial t = s_{a,char}[\rho_C/(\rho_C + \rho_B + \rho_A)]k_8 C_{O_2}$	[30]
9	C+O$_2$→CO$_2$	$r_9 = \partial C_{O_2}/\partial t = s_{a,char}[\rho_C/(\rho_C + \rho_B + \rho_A)]k_9 C_{O_2}$	
10	C+CO$_2$→2CO	$r_{10} = \partial C_{CO_2}/\partial t = s_{a,char}[\rho_C/(\rho_C + \rho_B + \rho_A)]k_{10} C_{CO_2}$	
11	C+H$_2$O→CO+H$_2$	$r_{11} = \partial C_{H_2O}/\partial t = s_{a,char}[\rho_C/(\rho_C + \rho_B + \rho_A)]k_{11} C_{H_2O}$	[28]
12	C+2H$_2$→CH$_4$	$r_{12} = \partial C_{H_2}/\partial t = s_{a,char}[\rho_C/(\rho_C + \rho_B + \rho_A)]k_{12} C_{H_2}$	
13	CO+1/2O$_2$→CO$_2$	$r_{13} = -\partial C_{CO}/\partial t = -k_{13} C_{CO} C_{O_2}^{0.25} C_{H_2O}^{0.5}$	
14	H$_2$+1/2O$_2$→H$_2$O	$r_{14} = -\partial H_2/\partial t = -k_{14} C_{H_2} C_{O_2}^{0.5}$	
15	CH$_4$+2O$_2$↔CO$_2$+2H$_2$O	$r_{15} = -\partial CH_4/\partial t = -(k_{15} C_{CH_4} C_{O_2}^2 - \overleftarrow{K_{15}} C_{CO_2} C_{H_2O}^2)$	
16	CO+H$_2$O↔CO$_2$+H$_2$	$r_{16} = -\partial CO/\partial t = -(k_{16} C_{CO} C_{H_2O} - \overleftarrow{K_{16}} C_{CO_2} C_{H_2})$	
17	CO+H$_2$→1/2CO$_4$+1/2CO$_2$	$r_{17} = -\partial CO/\partial t = -k_{17} C_{H_2} C_{CO}$	
18	CO+3H$_2$↔CH$_4$+H$_2$O	$r_{18} = -\partial CO/\partial t = -(k_{18} C_{CO} C_{H_2}^3 - \overleftarrow{K_{18}} C_{CH_4} C_{H_2O})$	
19	CO+4H$_2$→CH$_4$+2H$_2$O	$r_{19} = -\partial CO/\partial t = -(k_{19} C_{H_2}^4 C_{CO} - \overleftarrow{K_{19}} C_{CH_4} C_{H_2O}^2)$	
20	CH$_4$+H$_2$O↔CO+3H$_2$	$r_{20} = -\partial CH_4/\partial t = -(k_{20} C_{CH_4} C_{H_2O} - \overleftarrow{K_{20}} C_{CO} C_{H_2}^3)$	
21	CH$_4$+0.5O$_2$→CO+2H$_2$	$r_{21} = -\partial CH_4/\partial t = -k_{21} C_{CH_4} C_{O_2}^{0.5}$	

2.2 Biomass gasification process and meshing

Flow chart of biomass gasification is given in Fig. IV. The virgin biomass is put into the reactor, with the temperature growing up, different reactions happens. As we see from Fig. III, at low temperature, it is mainly vaporization happening, during which process, free water coming out first, when the temperature is higher than 100℃, hydrate water comes out. With temperature increasing, pyrolysis happens, during which process, biomass cracking into different species. These species keep cracking, finally small molecule gases and tar generate. During gasification process, which happens at much higher temperature, tar keep cracking into gases, and vapor may combine with these gas phase and solid phase to generate new species. Mainly the homogeneous and heterogeneous reactions happen in this stage.
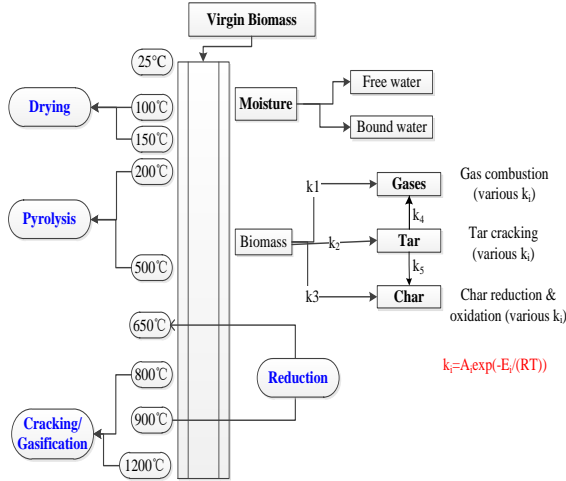

Figure IV Flow chart of biomass gasification

2.2 For boundary conditions

For all the N nodes we set in the control volume, there should be (N-1) partial differential equations for solving all the transport equations. Apparently, there is one equation missing for mass and energy equilibrium. Therefore the imaginary gas film Fig. V in between the solid and outside condition is crucial for deciding the heat and mass transport. Within each time step, the boundary conditions for mass and energy equation are shown in Table II.
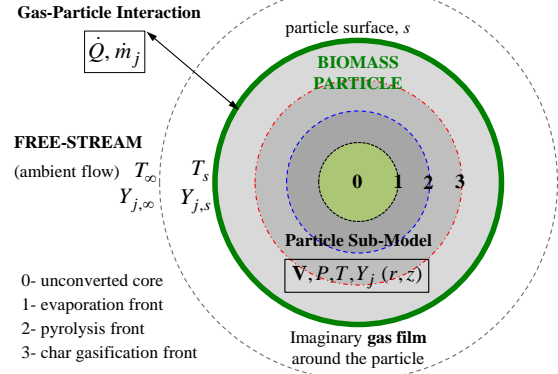

Figure V A conceptual view of the interaction of a large biomass particle with ambient flow and its conversion.

The h$_T$ and h$_M$ in the boundary condition is solved by Ranz-Marshall [31] model, with the following expression:
$$Nu \equiv \frac{h_T L_c}{k_g} = 2.0 + 0.6 Re^{1/2} Pr^{1/3}$$
$$Sh \equiv \frac{h_M L_c}{D_g} = 2.0 + 0.6 Re^{1/2} Sc^{1/3}$$

The reference temperature and species used in the boundary obey the one – third law [32]:
$$T_{ref} = T_s + 1/3\,(T_\infty - T_s)$$
$$Y_{j,ref} = T_{j,s} + 1/3\,(T_{j,\infty} - T_{j,s})$$

3 TRANSPORT EQUATIONS

3.1 Description of all transport equations

Partial differential equations (PDE) are used here to calculate all the result derived in the model. The general transport equations of different species are described as below:
$$\frac{\partial}{\partial t}(\varepsilon \rho \phi) + div(\rho \vec{u} \phi) = div(\rho \Gamma \nabla \phi) + S$$
ϕ can be any term we want to solve, for example temperature, pressure, density, species etc.

The first term is transient term, which represent the changing rate for unsteady flow, the second term is convection term, the third one is diffusion term and the last one is source term.

Table II: Transport equations

Density

Biomasss

$$\partial \rho_B / \partial t = S_B$$
$$S_B = -(r_1 + r_2 + r_3)$$

Char

$$\partial \rho_C / \partial t = S_C$$
$$S_C = r_3 + r_5 - r_8 \frac{2M_C}{M_{O_2}} - r_9 \frac{M_C}{M_{O_2}} - r_{10} \frac{M_C}{M_{CO_2}} - r_{11} \frac{M_C}{M_{H_2O}} - r_{12} \frac{M_C}{2M_{H_2}}$$

Moisture

$$\partial \rho_w / \partial t = \frac{\partial}{\partial r} \left(D_{eff,water} \frac{\partial \rho_{water}}{\partial r} \right) + S_{water}$$
$$S_{water} = -r_6 - r_7$$

Ash

$$\partial \rho_A / \partial t = 0$$

Continuity equations

$$\frac{\partial}{\partial t} (\varepsilon \rho_g) + div(\rho_g \vec{u}) = s_g$$
$$s_g = r_1 + r_2 - r_5 + r_6 + r_7 + r_8 \frac{2M_C}{M_{O_2}} + r_9 \frac{M_C}{M_{O_2}} + r_{10} \frac{M_C}{M_{CO_2}} + r_{11} \frac{M_C}{M_{H_2O}} + r_{12} \frac{M_C}{2M_{H_2}}$$

Species transfer equations

$$\frac{\partial}{\partial t} (\epsilon \rho_g Y_j) + div(\rho_g \vec{u} Y_j) = div(\rho_g D_{j,m} \nabla Y_j) + S_{Y,j}$$

Source terms for gas species:

$$S_{CO_2} = r_9 \frac{M_{CO_2}}{M_{O_2}} - r_{10} + r_{13} \frac{M_{CO_2}}{M_{CO}} + r_{15} \frac{M_{CO_2}}{M_{CH_4}} + r_{16} \frac{M_{CO_2}}{M_{CO}} + r_{17} \frac{M_{CO_2}}{2M_{CO}} + (r_1 + r_4)\alpha_{CO_2}$$

$$S_{CO} = r_8 \frac{2M_{CO}}{M_{O_2}} + r_{10} \frac{2M_{CO}}{M_{CO_2}} + r_{11} \frac{M_{CO}}{M_{H_2O}} - r_{13} - r_{16} - r_{17} - r_{18} - r_{19} + r_{20} \frac{M_{CO}}{M_{CH_4}} + r_{21} \frac{M_{CO}}{M_{CH_4}} + (r_1 + r_4)\alpha_{CO}$$

$$S_{O_2} = -r_8 - r_9 - r_{14} \frac{M_{O_2}}{2M_{H_2}} - r_{15} \frac{2M_{O_2}}{M_{CH_4}} - r_{21} \frac{M_{O_2}}{M_{CH_4}}$$

$$S_{H_2} = (r_1 + r_4)\alpha_{H_2} + r_{11} \frac{M_{H_2}}{M_{H_2O}} - r_{12} - r_{14} \frac{M_{O_2}}{2M_{H_2}} + r_{16} \frac{M_{H_2}}{M_{CO}} - r_{17} \frac{M_{H_2}}{M_{CO}} - r_{18} \frac{3M_{H_2}}{M_{CO}} - r_{19} \frac{4M_{H_2}}{M_{CO}} + r_{20} \frac{3M_{H_2}}{M_{CO}} + r_{21} \frac{2M_{H_2}}{M_{CO}}$$

$$S_{H_2O} = (r_1 + r_4)\alpha_{H_2O} + r_6 + r_7 - r_{11} + r_{14} \frac{M_{H_2O}}{M_{H_2}} + r_{15} \frac{2M_{H_2O}}{M_{CH_4}} - r_{16} \frac{M_{H_2O}}{M_{CO}} + r_{18} \frac{M_{H_2O}}{M_{CO}} + r_{19} \frac{2M_{H_2O}}{M_{CO}} - r_{20} \frac{M_{H_2O}}{M_{CH_4}}$$

$$S_{CH_4} = r_2 + (r_1 + r_4)\alpha_{CH_4} + r_{12} \frac{M_{CH_4}}{2M_{H_2}} - r_{15} \frac{M_{CH_4}}{2M_{O_2}} + r_{17} \frac{M_{CH_4}}{2M_{CO}} + r_{18} \frac{M_{CH_4}}{M_{CO}} + r_{19} \frac{M_{CH_4}}{M_{CO}} - r_{20} \frac{M_{CH_4}}{M_{H_2O}} - r_{21} \frac{2M_{CH_4}}{M_{O_2}}$$

$$S_{N_2} = 0$$

Energy equations

$$\frac{\partial}{\partial t} \left(\sum_{s,i} \rho_i h_i + \sum_{l,k} \rho_k h_k + \rho_g h_{mix} \right) + div(\rho_g \vec{u} h) = div(k_{eff} \nabla T) + div(\sum_j h_j \rho_g D_{j,m} \nabla Y_j) + S_h$$
$$h \triangleq \sum_j h_j Y_j$$
$$h_j = \int_{T_{ref}}^{T} C_{p,j}(T) dT$$
$$S_h = -\sum_{k=1}^{N} \Delta h_{f,k}^0 \dot{w}_k = \sum_{k=1}^{N} \Delta h_{R,k} \dot{w}_k$$

Darcy law
Ideal gas law

$$u = -\frac{k}{\mu} \frac{\partial p}{\partial x}$$
$$p = \frac{\rho_g RT}{W_g}$$

Boundary conditions

Outer face:
$$D_{j,m} A_s \left. \frac{\partial Y_{i(j,k)}}{\partial r} \right|_{r=r_p} = h_m A_s \left(Y_{i(j,k),\infty} - Y_{i(j,k),s} \right)$$

$$k_{eff} A_s \left. \frac{\partial T}{\partial r} \right|_{r=r_p} = h_T A_s (T_f - T) + A_s \omega \sigma (T_w^4 - T^4)$$

Inner face:
$$\left. \frac{\partial \emptyset}{\partial r} \right|_{r=0} = 0 , u = 0, P = P_{atm,} T = 298K, Y_j = 0$$

Initial conditions

Outer face:
$$T_\infty = T_f, u_\infty = u_f, Y_{O_2,\infty} = 0.23, Y_{N_2,\infty} = 0.77, P_\infty = P_{atm}$$

3.2 Discretized form of PDEs

Integrate all these partial differential equations and then discrete it into discretized equations which can be easily solved by Tri-diagonal matrix algorithm (TDMA). Geometry of a simple case using central differential method for discretion is shown in Fig. VI. P is the central

point of each node, while W and E represent the west face and east face. w and e represent the centers of west and east face. W and E are located in the center of previous point and the next point separately. In central differential method, the distance between each point is the same. For the transient term, a weighting parameter θ is chosen as zero during the integration, as a typical form of fully implicit scheme.

A typical discretized form PDEs using central differential method and fully implicit scheme is as shown below.

$$a_p \emptyset_p = a_w \emptyset_w + a_e \emptyset_e + a_P^0 \emptyset_P^0 + b$$

4. UPDATING STRUCTURE

Shrinking or swelling happens during gasification process. In this project, empirical parameters were used to calculate the size change. The shrinking factor θ has following expression:

$$\theta = V_P/V_{p,0} = (d_p)^3/(d_{p,0})^3 = 1 + (1 - \theta_m)\left(\frac{\rho_m}{\rho_{m,0}} - 1\right) + \theta_m(1 - \theta_v)\left(\frac{\rho_m}{\rho_{m,0}} - 1\right) + \theta_m\theta_v(1 - \theta_c)\left(\frac{\rho_c}{\rho_{c,0}} - 1\right)$$

θ_m, θ_v, θ_c represent three empirical shrinking factors of moisture, volatile and char.

Particle porosity ε is connected to size change as below:

$$\frac{\partial \varepsilon}{\partial t} = \frac{1}{\rho_{true}}\sum_k \vartheta_{c,k}\omega_k - \frac{\varepsilon}{\theta}\frac{\partial \theta}{\partial t}$$

ω_k is reaction rate of char reaction k,
$\vartheta_{c,k}$ is the stoichiometric coefficient of char in reaction k,
ρ_{true} is solid density without porous.

5. A SIMPLE CASE OF USING FVM

Given the complexity of solving such a highly coupled mathematical equations in this model. At this stage, a simple example is stated here about how to use central differential scheme to solve a transient convection diffusion equation with a source term.

The followings are the main descriptions of this problem.

$t > 0; a = -200, b = 100; x_1 = 0.6, x_2 = 0.2;$

$T = 0, x = 0; \frac{\partial T}{\partial x} = 0, x = L;$

$L = 1.5, u = 2.0, \rho = 1.0, \Gamma = 0.03$

Following are the numerical description:

$$\frac{\partial(\rho T)}{\partial t} + \frac{\partial(\rho uT)}{\partial x} = \frac{\partial}{\partial x}\left(\Gamma\frac{\partial T}{\partial x}\right) + S$$

For S, there are:

$0 < x \leq 0.6, S = 100 - 200x;$
$0.6 < x \leq 0.8, S = 100x - 80;$
$0.8 < x \leq 1.5, S = 0$

The discretized form of transport equations are:

$AE[i] = D - F/2$
$AW[i] = D + \frac{F}{2}$
$AP[i] = AW[i] + AE[i] + ap\_0$
$dx = \frac{Lmax - Lmin}{NR}$
$D = (\Gamma)/dx$
$F = \rho * U$
$ap\_0 = \rho * dx/dt$

At boundary nodes, the coefficients of equations are:
At the first node:
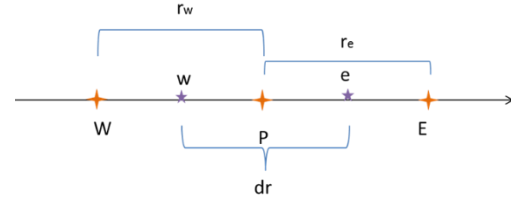$AW[0] = 0$

Here are all the PDEs, and there discretized form:

Figure. VI Geometry of a simple example

$AP[0] = AW[0] + AE[0] + ap\_0 + F + 2 * D$
At the last node:
$AE[NR - 1] = 0$
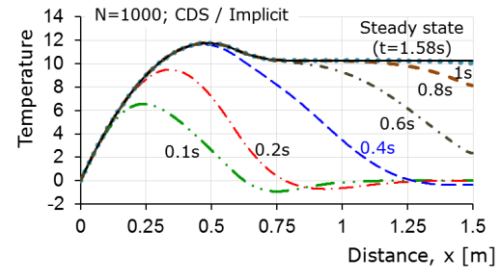$AP[NR - 1] = AW[NR - 1] + AE[NR - 1] + ap_0$

Figure VI Comparison of analytical solution and numerical solution based on a simple case

Fig. VI shows the final results from analytical and numerical calculation. A discretized grids N=1000 is taken. The solid black line is the result from analytical solution. The different dash lines show the result calculated from numerical solution with time from t=0.1s, 0.2s, 0.4s, 0.6s, 0.8s, 1s and 1.58s. Compared the blue dash line with the solid line, it is easy to find that the results from using FVM is almost the same with using analytical solution.

In another way, since using FVM can solve a fully general transport equation. It should be capable of solving a sequence of highly coupled similar transport equations. And with the best expectation, a similar overlap should be found between numerical calculation and experimental results.

6. CONCLUSIONS

Biomass gasification is processing technology in the future. Although a lot of work both in simulation and in experiments has been done in this area. There are still many issues waiting ahead. The key conclusions from this project are summarized below:
(1) Briefly summarizing the development of modelling of biomass gasification in state of arts.
(2) A relatively integral mathematical model is given in this paper. All the twenty one chemical reactions and their reaction rates are given. The key transport equations of a single biomass pellet during gasification is presented.
(3) Analytical solution and numerical solutions from a fully general transport equations are compared. Highly overlapping between an analytical solution and a stable state of numerical solution certify the

possibility of using central differential scheme to solve complete mathematical questions.

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