



**MARMARA UNIVERSITY**  
**INSTITUTE FOR GRADUATE STUDIES**  
**IN PURE AND APPLIED SCIENCES**



**NUMERICAL ANALYSIS AND  
EXPERIMENTAL VALIDATION OF  
FLUIDIZED BED GASIFICATION SYSTEMS  
WITH LOW-RANK COAL FUELS**

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**Thesis Supervisor**

Doç. Dr. Barış YILMAZ

ISTANBUL, 2022



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## ÖZET

### ISIL DEĞERİ DÜŞÜK KÖMÜR KULLANILAN AKIŞKAN YATAKLI GAZLAŞTIRMA SİSTEMLERİNİN SAYISAL ANALİZİ VE DENEYSEL YÖNTEMLERLE İNCELENMESİ

Bu tez çalışmasında, düşük kalorili kömürlerin akışkan yatak sistemlerindeki gazlaştırma karakteristiklerinin incelenmesi amaçlanmıştır. Bu kapsamda, bir akışkan yatak gazlaştırma sistemi sayısal ve deneysel yöntemlerle incelenmiş ve bu teknolojinin endüstriyel ölçekte uygulaması araştırılmıştır. Çalışmada öncelikle, referans alınan kömür numunelerinin laboratuvar ölçekli bir kabarcıklı akışkan yatak gazlaştırma sistemindeki davranışları değerlendirilmiştir. Kullanılan deneysel yöntemlerle birlikte gazlaştırma prosesi sonucu çıkan sentetik gazın bileşenleri, alt ısı değeri ve prosesin termal verimi farklı hava yakıt oranları için elde edilmiştir. Gazlaştırma prosesi öncesinde kömür numunelerine uygulanan yıkama ve kurutma işlemlerinin etkisi de bu çalışma kapsamında değerlendirilmiştir. Ayrıca, elde edilen deneysel veriler Hesaplamalı Akışkanlar Dinamiği (HAD) yöntemiyle doğrulanmıştır. Yapılan sayısal analizlerde, hem kimyasal reaksiyon, ısı ve kütle geçişinin ihmal edildiği soğuk akış yaklaşımıyla, hem de reaktif termal çözümlemeler ile birlikte, deneysel yöntemlerle tespiti çok zor olan akışkan yataktaki parçacık davranışı incelenmiş, ayrık fazın hareketi ve akış alanına etkisi değerlendirilmiştir. Sonrasında, gazlaştırma reaksiyonları, devolatilizasyon ve enerji denklemlerinin uygulanması sonucunda gazlaştırma prosesinin verimlilik değerleri elde edilmiş, elde edilen bulgular deneysel sonuçlarla karşılaştırmalı olarak irdelenmiştir. Aynı kömür numuneleri referans alınarak endüstriyel ölçekli bir dolaşımli akışkan sistemi tasarlanmış, bu sistemin işletme parametreleri belirlenmiştir. Son adımda ise tasarlanan endüstriyel ölçekli dolaşımli akışkan yatak sisteminde termal verime ve karbon dönüşüm hızına doğrudan etki eden bir büyüklük olan katı sirkülasyon hızını etkileyen parametreler HAD yöntemleri ile incelenmiştir. Yüzeysel hız, hava fazlalık katsayısı ve basınç düşüşü gibi değişkenler ile birlikte akışkan yatak prosesi incelenerek endüstriyel ölçekli sistemlerin tasarımında gerekli parametrelerin belirlenmesi hedeflenmiştir.

## **ABSTRACT**

### **NUMERICAL ANALYSIS AND EXPERIMENTAL VALIDATION OF FLUIDIZED BED GASIFICATION SYSTEMS WITH LOW-RANK COAL FUELS**

In this thesis study, it was aimed to investigate the gasification characteristics of low-rank coals in fluidized bed systems. Within this scope, a fluidized bed gasification system was examined by numerical and experimental methods and the industrial-scale application of this technology was also evaluated. Initially, the gasification behavior of the reference coal samples in a laboratory-scale bubbling fluidized bed system was evaluated in the study. The composition of the synthetic gas resulting from the gasification process and thermal efficiency are obtained for different air-fuel ratios via experimental methods used. The effect of cleaning and drying processes applied to the coal samples before the gasification process was also evaluated within the scope of this study. Also, the experimental data are verified by using Computational Fluid Dynamics (CFD) methods. The behavior of the particles in the fluidized bed, which is very difficult to obtain by experimental methods, was examined with both reactive thermal analyses and cold flow analyses - in which chemical reaction, heat and mass transfer are neglected. The motion of the discrete phase and its effects on the flow field are evaluated. Moreover, the thermal efficiency of the gasification process was obtained as a result of the application of chemical reactions, devolatilization models and energy equations. The numerical results are also examined in comparison with the experimental results. An industrial-scale circulating fluidized bed system was designed and its operational conditions are determined by considering the same coal samples. Finally, the solid circulation rate of this industrial-scale circulating fluidized bed system is investigated by CFD methods, which is a deterministic parameter for thermal efficiency and carbon conversion rate in the gasification process. It was aimed to determine optimum values of the parameters such as superficial velocity, excess air coefficient, carbon conversion rate, and pressure drop in the design of industrial-scale systems by examining the fluidized bed gasification process.

## **CLAIM FOR ORIGINALITY**

I, Yalçın Yükselentürk hereby submit my thesis, entitled “Numerical Analysis and Experimental Validation of Fluidized Bed Gasification Systems with Low-Rank Coal Fuels” and truthfully declare that the paper is a product of my original research investigation. To the best of my knowledge, it involves no material previously published by another person nor does it include contents that are falsified or fabricated. I also sought permission from the copyright owner to use text, illustrations, and/or framework substantively used in the paper.



## LIST OF SYMBOLS

|           |                                                             |
|-----------|-------------------------------------------------------------|
| $a$       | Area / Surface area                                         |
| $A, A_0$  | Pre-exponential factor                                      |
| $Ar$      | Archimedes number                                           |
| $c$       | Model coefficient / Amount of char bridge                   |
| $C$       | Courant number                                              |
| $c_p$     | Specific heat                                               |
| $C_d$     | Drag coefficient                                            |
| $d_p$     | Particle diameter                                           |
| $D$       | Drag function                                               |
| $D_m$     | Mass diffusion coefficient                                  |
| $E, E_a$  | Activation energy                                           |
| $f$       | Particle distribution function                              |
| $F$       | Body force                                                  |
| $g$       | The gravitational acceleration                              |
| $G_f$     | Fluid mass flow rate in Recycle pipe                        |
| $G_p$     | Particle circulation rate                                   |
| $h$       | Enthalpy / Heat transfer coefficient / Height               |
| $h_m$     | Mass transfer coefficient                                   |
| $k$       | Thermal conductivity / kinetic rate / kinetic rate constant |
| $L$       | Cell size                                                   |
| $m$       | Mass                                                        |
| $\dot{m}$ | Mass flow rate                                              |
| $M$       | Moisture                                                    |
| $n$       | Model coefficient / Time step / Molar fraction coefficients |
| $N$       | Number of any quantity                                      |
| $p$       | Pressure                                                    |
| $p_s$     | Pressure constant                                           |
| $q$       | Heat source                                                 |
| $Pr$      | Prandtl number                                              |

|           |                                         |
|-----------|-----------------------------------------|
| $Q$       | Numerical quantity                      |
| $r$       | Reaction rate / particle radius         |
| $R$       | Universal gas constant                  |
| $Re$      | Reynolds number                         |
| $S_h$     | Energy transferred between phases       |
| $Sc$      | Schmidt number                          |
| $Sh$      | Sherwood number                         |
| $t$       | Time / Wall thickness                   |
| $T$       | Temperature                             |
| $u, U$    | Velocity                                |
| $v, V$    | Volume                                  |
| $\dot{V}$ | Volumetric flow rate                    |
| $X$       | Mass fraction                           |
| $W$       | Particle circulation rate per unit area |

## GREEK SYMBOLS

|               |                                                               |
|---------------|---------------------------------------------------------------|
| $\alpha$      | Mass fraction coefficient / Model constant                    |
| $\beta$       | Mass fraction coefficient / Model constant                    |
| $\delta$      | Mass ratio of an element in a molecule / Amount of side chain |
| $\varepsilon$ | Voidage / Model constant                                      |
| $\eta$        | Thermal efficiency                                            |
| $\phi$        | Flow direction indicator / Stoichiometric coefficient         |
| $\Phi$        | Turbulent dissipation / Weighting factor                      |
| $\Psi$        | Sphericity                                                    |
| $\zeta$       | Conversion of volatile matter                                 |
| $\xi$         | Amount of labile bridge                                       |
| $\lambda$     | Density of any quantity                                       |
| $\mu$         | Dynamic viscosity                                             |
| $\rho$        | Density                                                       |
| $\sigma$      | Pressure fluctuation                                          |
| $\theta$      | Volume fraction                                               |

|          |                  |
|----------|------------------|
| $\tau$   | Stress tensor    |
| $\omega$ | Weighting factor |

## SUBSCRIPTS

|                |                      |
|----------------|----------------------|
| <i>a</i>       | Air                  |
| <i>act</i>     | Actual               |
| <i>adj</i>     | Adjusted             |
| <i>b</i>       | Bridge               |
| <i>B</i>       | Boundary layer       |
| <i>c</i>       | Char / Core          |
| <i>cp</i>      | Close packed         |
| <i>d</i>       | Devolatilization     |
| <i>D</i>       | Diffusion            |
| <i>E</i>       | Ergun drag model     |
| <i>f</i>       | Fluid                |
| <i>g</i>       | Gas                  |
| <i>h</i>       | Nozzle hole          |
| <i>i, j, k</i> | Direction vectors    |
| <i>L</i>       | Local cell           |
| <i>mf</i>      | Minimum fluidization |
| <i>mr</i>      | Moisture release     |
| <i>N</i>       | Nozzle               |
| <i>p</i>       | Particle             |
| <i>R</i>       | Reactor              |
| <i>RC</i>      | Recycle chamber      |
| <i>s</i>       | Solid                |
| <i>SC</i>      | Supply chamber       |
| <i>sto</i>     | Stoichiometry        |
| <i>SU</i>      | Start-up             |
| <i>TO</i>      | Tar Oxidation        |
| <i>TC</i>      | Tar Cracking         |
| <i>WY</i>      | Wen-Yu drag model    |

## SUPERSCRIPTS

***a, b, c***      Exponential coefficients

***d***              Devolatilization

**0**                Initial

***m, n, p, t***    Exponential coefficients



## LIST OF ABBREVIATIONS

|                |                                                 |
|----------------|-------------------------------------------------|
| <b>AW</b>      | Atomic Weight                                   |
| <b>BFB</b>     | Bubbling Fluidized Bed                          |
| <b>CFB</b>     | Circulating Fluidized Bed                       |
| <b>CFD</b>     | Computational Fluid Dynamics                    |
| <b>CLC</b>     | Chemical Looping Combustor                      |
| <b>CPD</b>     | Chemical Percolation model for Devolatilization |
| <b>CPFD</b>    | Computational Particle Fluid Dynamics           |
| <b>DAE</b>     | Distributed Activation Energy                   |
| <b>DAF</b>     | Dry and Ash Free                                |
| <b>DTF</b>     | Dry and Tar Free                                |
| <b>DEM</b>     | Discrete Element Method                         |
| <b>DPC</b>     | Discrete Particle Chemistry                     |
| <b>DPM</b>     | Discrete Particle Method                        |
| <b>E-E TFM</b> | Eulerian - Eulerian Two Fluid Method            |
| <b>EMMS</b>    | Energy Minimization Multi-Scale                 |
| <b>ER</b>      | Equivalence Ratio                               |
| <b>FC</b>      | Fixed Carbon                                    |
| <b>GCV</b>     | Gross Calorific Value                           |
| <b>GPU</b>     | Graphics Processing unit                        |
| <b>HHV</b>     | Higher Heating Value                            |
| <b>IGCC</b>    | Integrated Gasification Combined Cycle          |
| <b>LAR</b>     | Loop-Seal Total Aeration Rate                   |
| <b>LES</b>     | Large-Eddy Simulation                           |
| <b>LS</b>      | Loop-Seal                                       |
| <b>LHV</b>     | Lower Heating Value                             |
| <b>MFR</b>     | Mass Flow Rate                                  |
| <b>MGAS</b>    | MTEC Gasifier Advanced Simulation               |
| <b>MTEC</b>    | The Munitions Engineering and Technology Center |
| <b>MP-PIC</b>  | Multiphase Particle-in Cell                     |
| <b>MW</b>      | Molecular Weight                                |

|              |                                                          |
|--------------|----------------------------------------------------------|
| <b>NMR</b>   | Nuclear Magnetic Resonance                               |
| <b>ODE</b>   | Ordinary Differential Equation                           |
| <b>PA</b>    | Proximate Analysis                                       |
| <b>PM</b>    | Particulate Matter                                       |
| <b>PDC</b>   | Partial Donor Cell                                       |
| <b>PID</b>   | Proportional-Integral-Derivative                         |
| <b>PSD</b>   | Particle Size Distribution                               |
| <b>RPM</b>   | Round per Minute                                         |
| <b>RS</b>    | Aeration rate ratio of recycle chamber to supply chamber |
| <b>SCR</b>   | Solid Circulation Rate                                   |
| <b>SP</b>    | Standpipe                                                |
| <b>TC</b>    | Thermocouple / Tar-Cracking                              |
| <b>TGA</b>   | Thermo-gravimetric Analysis                              |
| <b>UA</b>    | Ultimate Analysis                                        |
| <b>UDF</b>   | User-defined Function                                    |
| <b>VAC</b>   | Volume Average Chemistry                                 |
| <b>VFR</b>   | Volumetric Flow Rate                                     |
| <b>VM</b>    | Volatile Matter                                          |
| <b>2D</b>    | Two Dimensional                                          |
| <b>2P1SM</b> | Two Parameter - Single Stage Model                       |
| <b>3D</b>    | Three Dimensional                                        |

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# 1. INTRODUCTION

## 1.1. Background

### 1.1.1. Coal – A Primary Energy Source

In recent decades, many macroeconomic and political strategies are based on moves to meet the increasing level of energy demand by considering limited energy resources. While the worldwide energy consumption rate has increased by 16.2% in the last ten years, many countries have to import more fossil energy resources, notably natural gas and petroleum [1]. In Turkey, the Ministry of Energy and Natural Resources reported that the primary energy supply was raised by 35.7% and attained 143.7 mtep in 2018 compared to 2010 [2]. Only 24.3% of the total energy demand is supplied from domestic resources; the rest of the demand is supplied from imported energy resources [3]. Due to its availability, utilization of the local energy resources such as coal and biomass with efficient and clean technologies for energy production is a wise option to reduce energy dependency. At this juncture, development of the clean and efficient technologies is very important to utilize local energy resources.

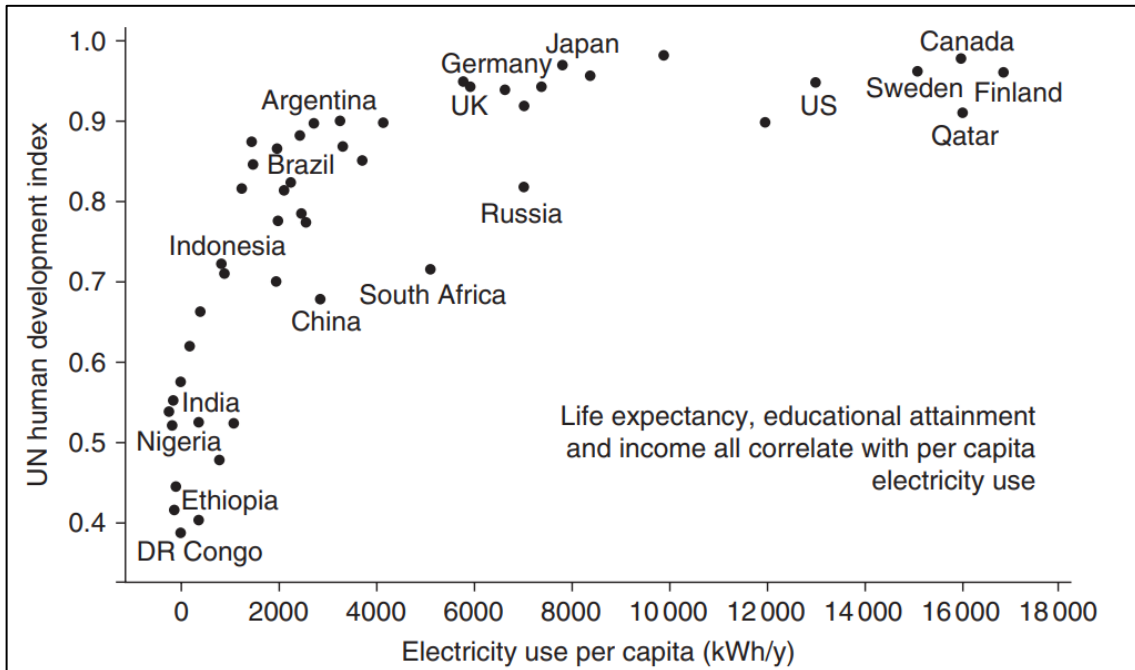


**Figure 1.1** Relationship between electricity usage and human development index [4]

Ease of access to energy resources determines the life quality of societies. Actually, UN Development Programme's Millennium Development Goals for 2015, such as poverty and hunger, education, child health and environmental sustainability, require access to electricity. In the literature, it was reported that higher electricity usage rates per capita improve human development (Figure 1.1). Moreover, it was stated that the annual electricity consumption rate per capita should be at least 4000 kWh to reach a reasonable Human Development Index level [4]. Hence, low-cost energy production is a vital topic for developing countries.

According to International Energy Agency (IEA) and World Coal Association's reports, 30% of energy production and 37% of electrical energy production are supplied from coal resources all over the world [1][5]. Figure 1.2 and Figure 1.3 show the consumption trend of different types of energy resources in the world and Turkey, respectively. Despite the increasing number of renewable energy applications, it is predicted that the use of coal resources will continue in the near future by increasing energy demand and improvements in clean coal technologies [6][7].

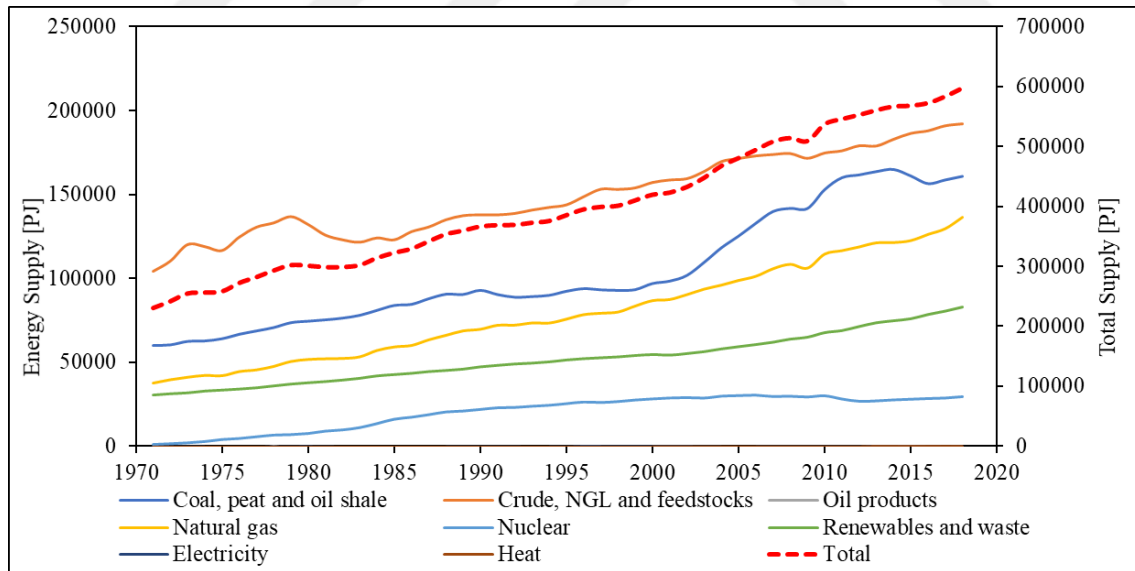


**Figure 1.2** Total primary energy supply in the world between 1971 and 2018, adapted from [1]

It is known that coal resources are the primary energy resource for energy production in Turkey. General Directorate of Turkish Coal (TKİ) reported that 15.8% of the primary energy supply is provided from local coal resources. The local coal supply consists of 16.4 Mtoe lignite, 1.3 Mtoe anthracite and 0.4 Mtoe asphaltite, and the overall magnitude

is 18.1 Mtoe. As a local energy source, the heating value of the lignite reserves in Turkey is at a very low level. The lower heating value is lower than 3000 kcal/kg for %90 of the local coal resources. However, it varies generally between 1000 kcal/kg and 4200 kcal/kg [3].

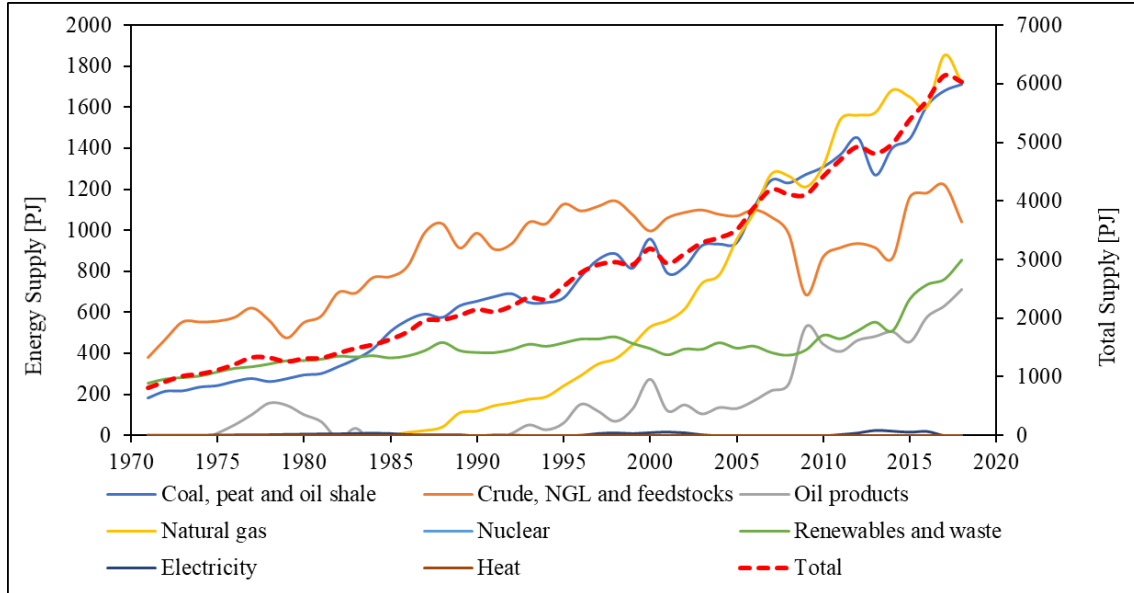


**Figure 1.3** Total primary energy supply in Turkey between 1971 and 2018, adapted from [1]

One of the most considerable challenges in utilization of these coal resources is related to the chemical composition of the low-rank coals. Most of them cannot be burned directly in thermal power plants due to their high sulphur content. It shows that the development of the efficient and clean energy production technologies become more significant for coal resources.

The coal rank increases from lignite to anthracite in the coalification process as seen in Table 1.1. As an indicator of the coal rank, Gross Calorific Value (GCV) is useful parameter in the market, because it is an easy and convenient way to describe coal quality basically. Although relatively small amount of anthracite is available, bituminous and sub-bituminous coals are widely used as a commercial coal resource. Also, low-grade coal resources such as lignite commonly takes a significant role in industrial applications due to its availability. However, it is not economically viable to transport low-rank coals over long distances; hence, most of the lignite resources are utilized close to sites where

it is mined. Low-rank lignite resources with high ash and high moisture content are taken into consideration in this study.

**Table 1.1** Classification of various coal resources by rank (moist, mineral-matter-free basis) [8][9][10]

| <b>Rank</b>                     | <b>GCV [Btu/lb]</b> | <b>GCV [MJ/kg]</b> |
|---------------------------------|---------------------|--------------------|
| Meta-Anthracite                 |                     |                    |
| Anthracite                      | 13760 - 14190       | 32-33              |
| Semi-Anthracite                 |                     |                    |
| High volatile A bituminous coal | >14000              | >32.56             |
| High volatile B bituminous coal | 13000-14000         | 30.23-32.56        |
| High volatile C bituminous coal | 11500-13000         | 26.74-30.23        |
| Sub-bituminous A coal           | 10500-11500         | 24.42-26.74        |
| Sub-bituminous B coal           | 9500-10500          | 22.09-24.42        |
| Sub-bituminous C coal           | 8300-9500           | 19.30-22.09        |
| Lignite A                       | 6300-8300           | 14.65-19.30        |
| Lignite B                       | <6300               | <14.65             |

### 1.1.2. Fluidized Bed Technology

A variety of industrial sectors involve many applications of fluidized bed because of numerous advantages coming with its technology. Fuel flexibility, high reaction/thermal efficiency, low NO<sub>x</sub> emissions, in-situ sulphur caption capability are some of well-known pros. Fluidized bed systems have also some disadvantages as described below [11]:

- Inefficient interaction may occur between fine particles in a bubbling fluidized bed application due to large fluctuations of a plug flow. It can be significant especially for the applications require high conversion rates.
- Erosion of the reactor and duct walls from abrasion by particles can cause serious problems during operating conditions.
- In the case of high bed temperatures, the agglomeration of fine particles can require a decrease in temperature, thus reaction rates and overall thermal efficiency can be reduced.

- The sudden mixing of particles may yield variable residence times in the bed region which affects the transient uniformity of syngas output and efficiency.

Fluidized bed technology is widely used in many industrial areas, despite all these disadvantages. Most common industrial applications are given in Figure 1.4 [12]. Only application of fluidized bed on gasification process as an energy conversion method is considered within the scope of this study.

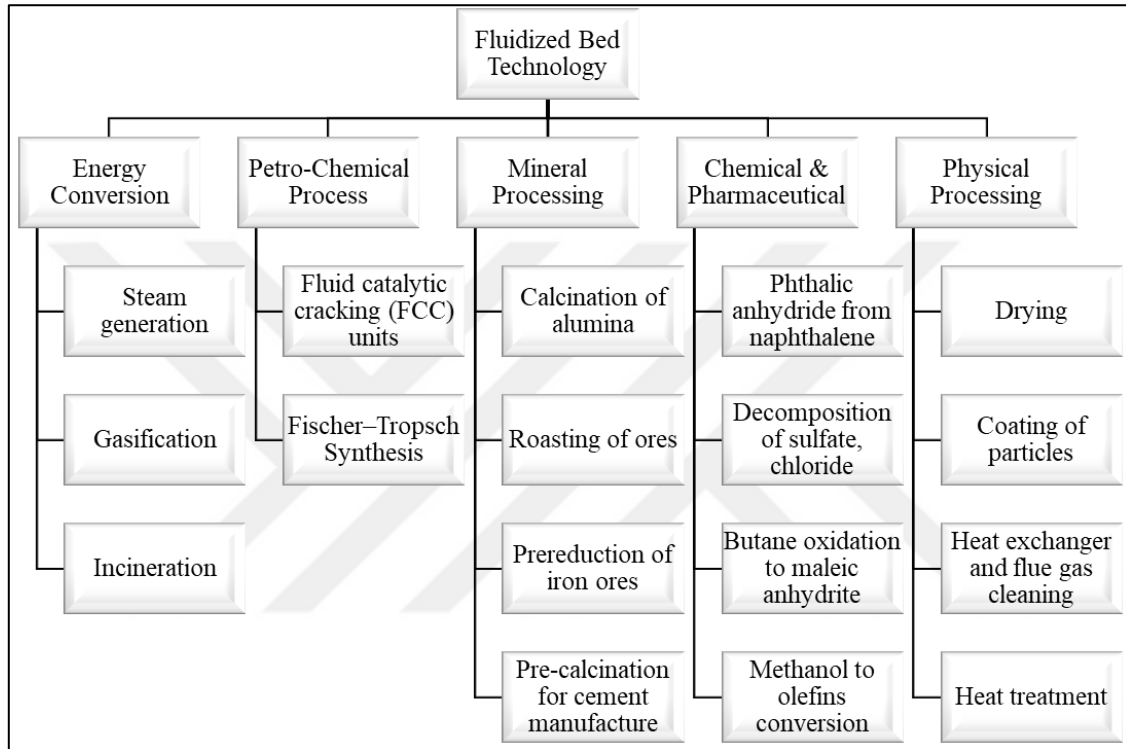


**Figure 1.4** Industrial applications of fluidized bed technology [12]

Fluidized bed gasification technology becomes prominent as a clean and efficient energy production method from coal and biomass in recent years [13]-[16]. This technology consists of subsystems some of which are subject to development and innovations. Especially for fluidized bed reactors, the design of the system is very difficult because of including complex physical phenomena and number of variables. The fluidized bed gasification system is a recent research area in many countries. There are numerous projects that supported by European 7<sup>th</sup> Framework and Horizon 2020 Programmes related to the fluidized bed technology [17]-[22].

The design and production of the fluidized bed technology require advanced engineering knowledge. Operating conditions such as mass flow rate of the fuel and fluidizer (air,

water vapor, oxygen, etc.), mean diameter of bed material, superficial velocity, bed regime, etc., affect directly all outputs and efficiency of the overall system. Since fluidization dynamics is based on many parameters related to the particle and flow properties, it is very arduous to predict main bed behavior and fluidization regime in a fluidized bed reactor. Change of the fluidization regime with increasing superficial gas velocity for a particular reactor with same gas phase properties and same solid particles is illustrated in Figure.1.5.

Geldart presented a convenient criterion to predict the fluidization structure depending on two main parameters: density and Sauter mean diameter of the particle [23]. Solid particles are classified into four Geldart groups - A, B, C, and D - as shown in Figure 1.6. Each group defines the main behavior of a particle type with certain physical properties in a multiphase flow field.

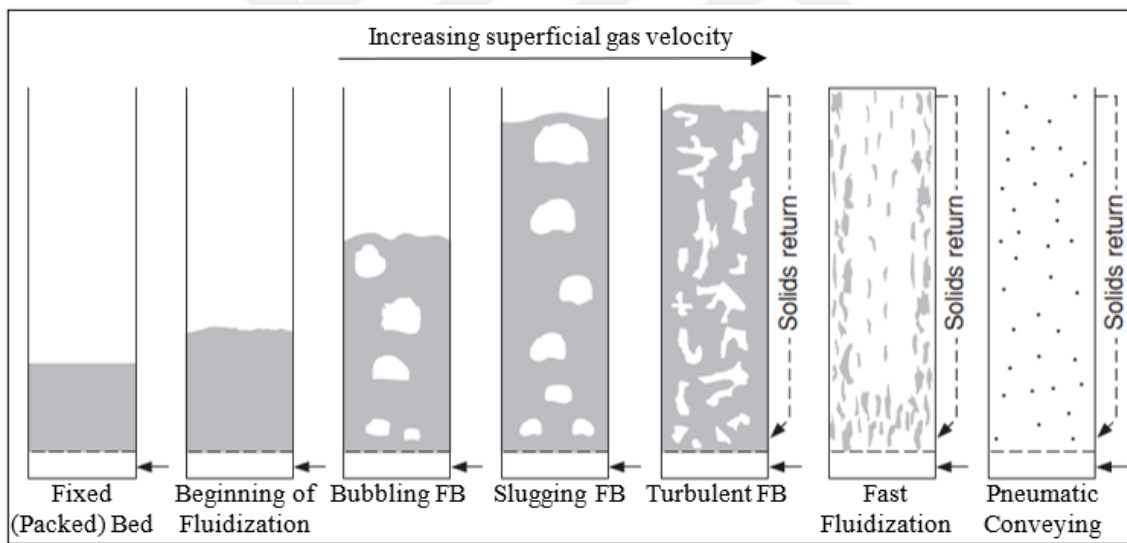


**Figure 1.5** Fluidized Bed Regimes, adapted from [24]

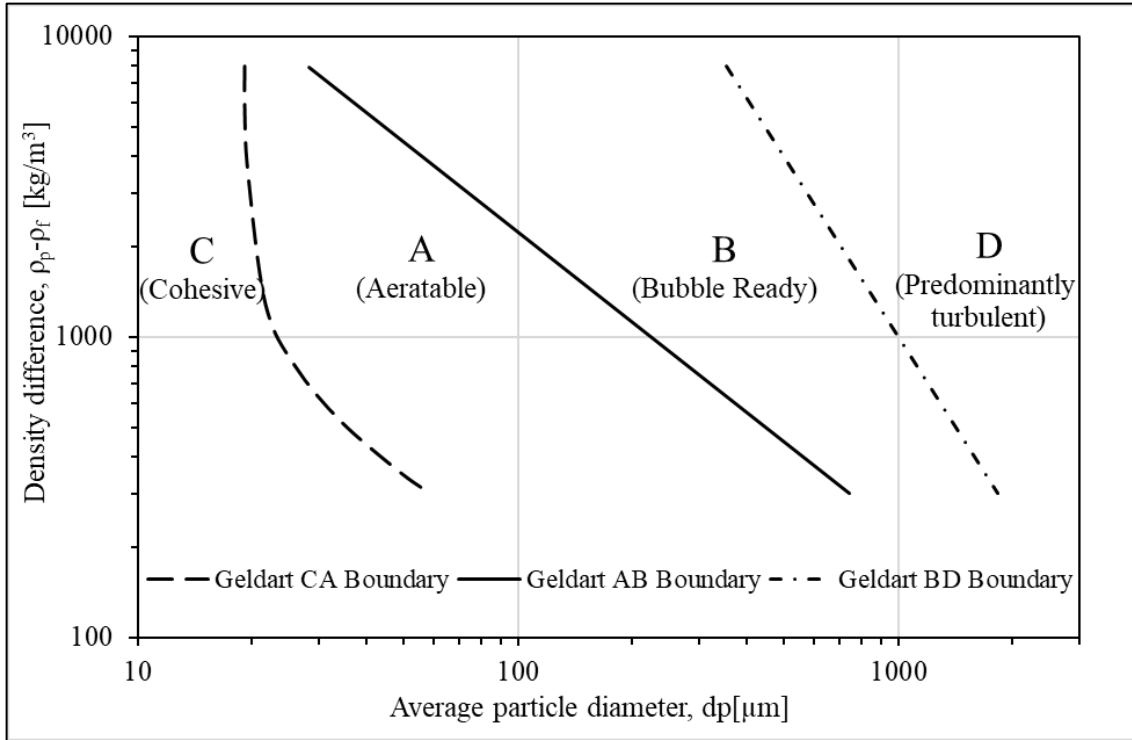


**Figure 1.6** Geldart Particle Classification Diagram, adapted from [25]

Determination of a fluidized bed regime for an application is the main design step of a fluidized bed system. Grace also proposed a diagram to select the working regime for a fluidized bed application by using dimensionless superficial velocity and dimensionless particle diameter as seen in Figure 1.7 [26]. Higher dimensionless particle diameters with lower superficial velocity values yield quasi-static beds with small motion. In order to obtain higher interactions between particles and stronger bed motion higher superficial gas velocity should be applied on smaller particle diameters.

Moreover, other flow characteristics and bed properties vary with different fluidization regimes. The comparison of the main parameters of different flow regimes for different applications is given in Table 1.2. As seen in this table there is no direct relationship between main properties such as superficial velocity, voidage and particle diameter on heat transfer coefficient.

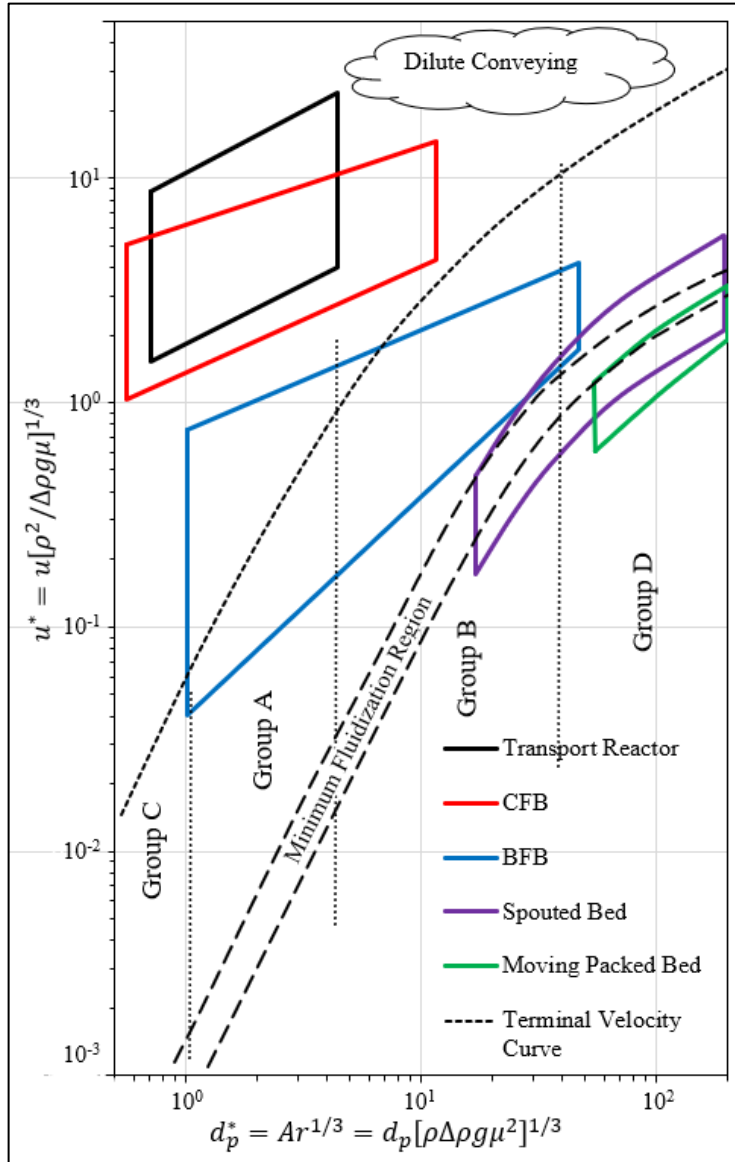


**Figure 1.7** Fluidization regime diagram with dimensionless superficial velocity and dimensionless particle diameter parameters, adapted from [26]

**Table 1.2** Comparison of physical properties of different fluidization regimes [26]

| Property                                 | Unit  | Packed Bed   | Fluidized | Fast Fluidization | Pneumatic Conveying |
|------------------------------------------|-------|--------------|-----------|-------------------|---------------------|
| Application in boilers                   |       | Stoker fired | Bubbling  | Circulating       | Pulverized          |
| Mean particle diameter                   | mm    | <300         | 0.03-3    | 0.05-0.5          | 0.02-0.08           |
| Superficial gas velocity                 | m/s   | 1-3          | 0.5-2.5   | 4-6               | 15-30               |
| Velocity ratio of phases                 | -     | 0.01         | 0.3       | 2                 | 40                  |
| Gas motion                               | -     | Static       | Up & down | Mostly up         | Up                  |
| Overall voidage                          | -     | 0.4-0.5      | 0.5-0.85  | 0.85-0.99         | 0.98-0.998          |
| Bed to surface heat transfer coefficient | W/m²K | 50-150       | 200-550   | 100-200           | 50-100              |

### 1.1.3. Gasification Technology and Applications

The gasification process aims to obtain useful gas as a result of the chemical reaction of organic solid particle molecules in a certain temperature and pressure environment by using gasification agents such as air, water vapor, oxygen and carbon monoxide. Gasification can be defined as a process in which organic materials are converted into a type of gaseous fuel, namely synthetic gas. The synthetic gas obtained as a result of the gasification process mainly includes carbon monoxide, carbon dioxide, water vapor, hydrogen, methane, tar and ash molecules. The efficiency of the gasification process depends on various parameters such as carbon and hydrogen content of the fuel, gasifier type and its operating conditions, reactor temperature and gasifier agents. Production of synthetic gas and liquid fuels can be utilized either in electricity and heat generation or in chemical production by different synthesizing methods such as Fischer-Tropsch, methanol, hydrogen, natural gas and ammonia.

By using appropriate gasification techniques, many materials such as tree and forest residuals, low-rank coals, sewage sludge, tire wastes and plastic wastes can be effectively utilized and a sustainable energy ecosystem can be supported. These feedstocks are schematically classified by De et al. as shown in Figure 1.8 [27]. Commercial applications involving coal, wood, tire and sludge gasification technologies are given in Figure 1.9.a-d, respectively.

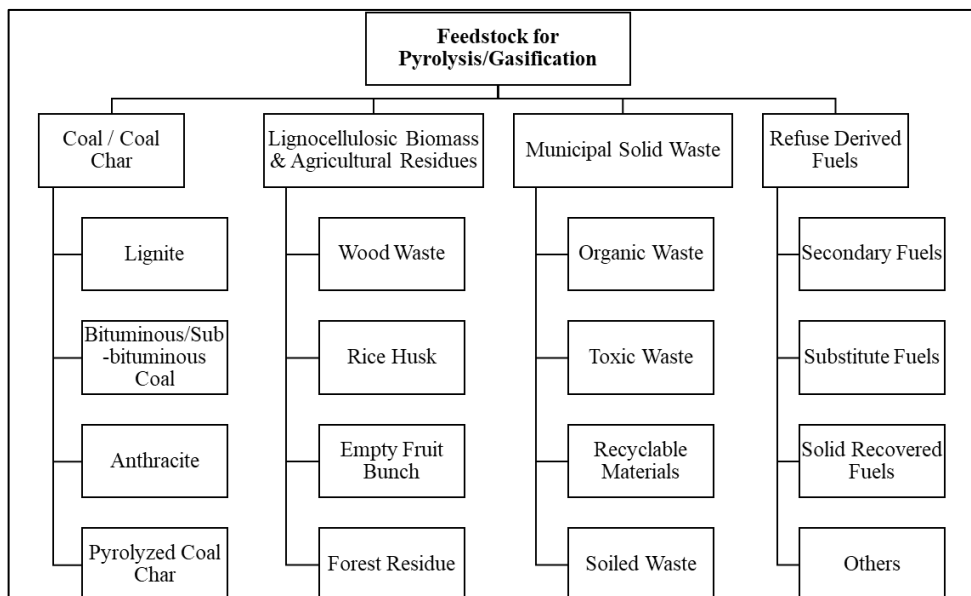


**Figure 1.8** Feedstock classification for gasification applications, adapted from [27]

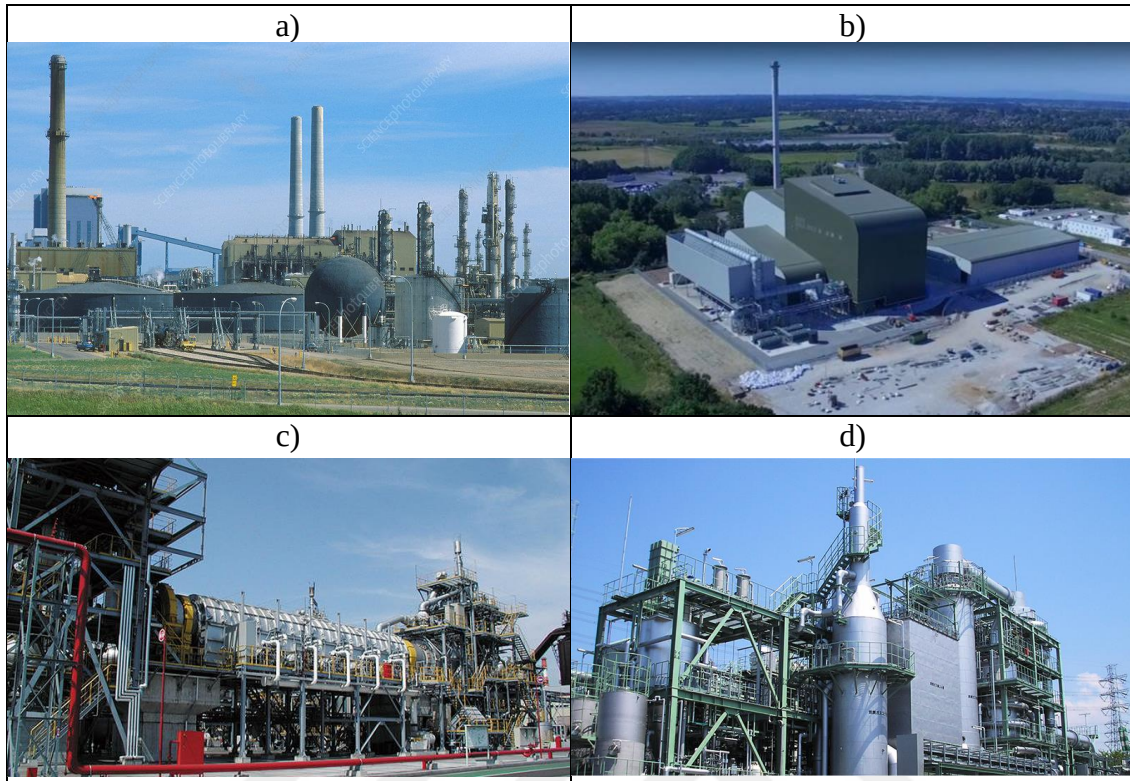


**Figure 1.9** a) Dakota Coal Gasification Plant [28], b) Wood Waste Gasification Plant in Cheshire [29], c) Waste Tire Gasification Recycling Facility in Himeji [31] d) Kiyose Water Reclamation Center in Tokyo [30]

In the case of usage of air as the gasification agent, a high rate of nitrogen gas is observed in the synthetic gas composition. However, toxic and hazardous molecules such as  $\text{NO}_x$ ,  $\text{H}_2\text{S}$ ,  $\text{NH}_3$ ,  $\text{HCl}$  and  $\text{HCN}$  may be seen as the outputs of this process. In order to utilize useful gases such as hydrogen, methane and carbon monoxide, synthetic gas must be purified from gaseous contaminants and solid particles with various gas cleaning technologies such as particle separation, scrubbing, fixed bed capturing, catalytic cracking etc [32]. Syngas cleaning and conditioning systems are vital components of commercial gasification plants as seen in Figure 1.10 [28]. Appropriate cleaning technologies ensure lower degree of pollutants such as  $\text{NO}_x$ ,  $\text{SO}_x$  and PM in the exhaust gases of the plants (Figure 1.11).

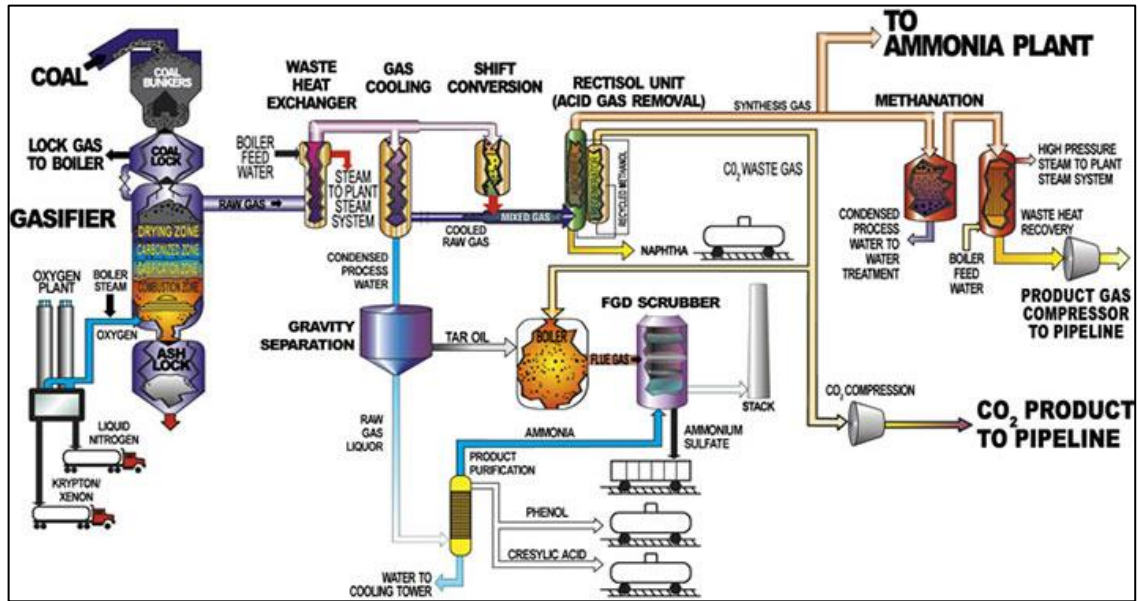


**Figure 1.10** Basic layout of the coal gasification plant in Dakota [28]

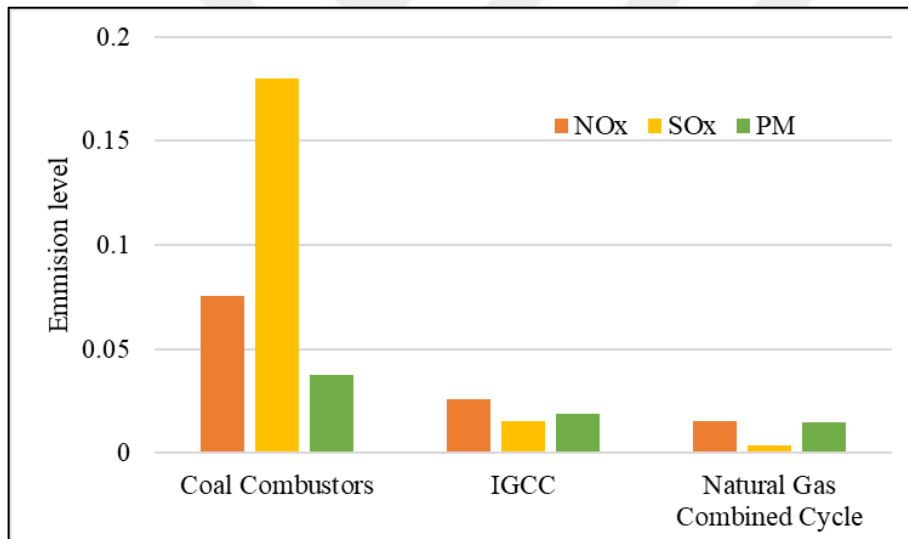


**Figure 1.11** Comparison of emission values in an arbitrary scale for different coal to energy conversion systems [33]

Gasification is widely known as an incomplete combustion process of a solid feedstock. In typical gasifiers, the following physical, thermal and chemical steps are realized at given temperatures [12]:

- Drying ( $T > 150^{\circ}\text{C}$ )
- Devolatilization & Pyrolysis ( $150^{\circ}\text{C} < T < 700^{\circ}\text{C}$ )
- Combustion ( $700^{\circ}\text{C} < T < 1500^{\circ}\text{C}$ )
- Gasification (Reduction) ( $800^{\circ}\text{C} < T < 1500^{\circ}\text{C}$ )

Each of these steps involves various reaction temperature ranges and occurs in the different time periods. Principle gasification steps are illustrated in Figure 1.12.

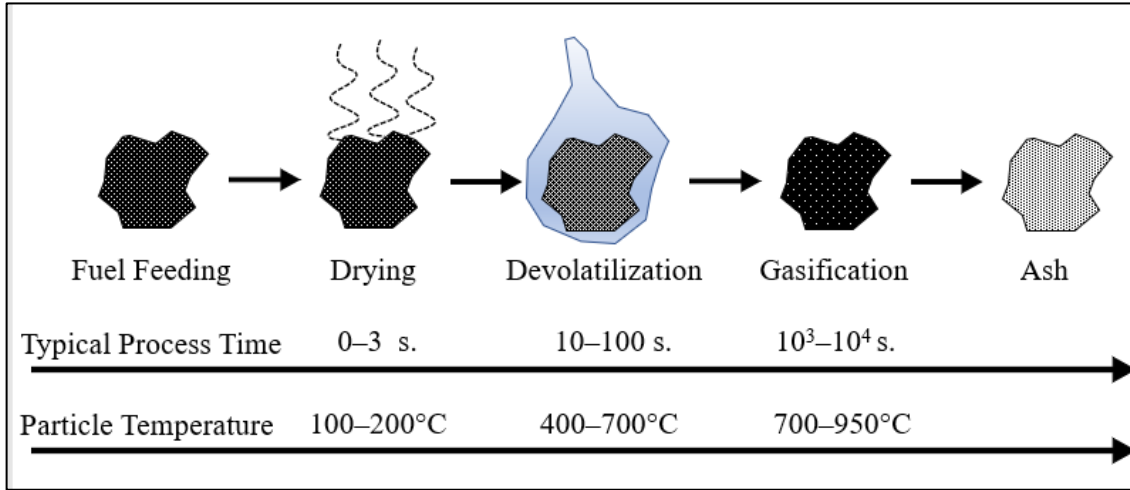
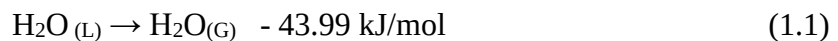


**Figure 1.12** Principal steps of gasification process, adapted from [34]

### Drying

Drying, pyrolysis and reduction processes consume some amount of heat energy generated as an output of the exothermic combustion reactions. When a feedstock particle enters a gasification reactor, the first step is the drying process. In the drying process, the moisture in the solid particle evaporates and releases into the gaseous phase. The phase transition of water can affect the overall gasification kinetics and efficiency of the process, especially in the case of gasification of high moisture content and low-grade coal. Phase change reaction for the drying process is defined as follows:

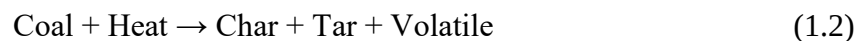


### Devolatilization & Pyrolysis

Next in the gasification process is the devolatilization/pyrolysis steps. A series of complex physical processes and chemical reactions occur during these steps which start approximately at 230°C. Some volatile may release at lower temperatures. Chemical bonds between carbon and oxygen, nitrogen, or sulfur start to break at this condition. The rate of these reactions increases with increasing temperature and it reaches very high levels around 700°C [12]. The pyrolysis process separates the water vapor, organic liquids and non-condensable gases from the char of solid fuel. As a result of the pyrolysis process, the output product can be categorized into three main groups:

- Light gases: Volatiles such as CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>, etc.
- Tar: Viscous, heavy and corrosive liquid compound
- Char particles: Solid particles mainly consist high-carbon ratio

At the initial stages of the pyrolysis process, unstable fragments such as tar and polyaromatic hydrocarbon molecules are formed. These molecules convert into relatively stable components at higher temperatures. The pyrolysis process can be simply represented by the following mechanism:



Mechanisms details and chemical modeling approach for this process are given next chapter.

### **Combustion**

After the pyrolysis process of the coal, intermediate products start to react with oxygen at higher temperatures between 800-1000°C before gasification reactions. These reactions are exothermic reactions that provide all heat energy needed for the endothermic reactions of the gasification process. The combustion stage involves partial oxidation of char particles, volatiles and other hydrocarbons in a gasifier. It is very important about these reactions that the mass flow rate of oxygen supplied with the pyrolysis products should be controlled at an appropriate amount in order to maintain the reactor temperature at the desired level and to avoid complete oxidation of the compounds.

### **Gasification (Reduction)**

Gasification involves a series of endothermic reactions provided by the heat generated in the combustion reactions. As the last step of the gasification process, char reacts mostly with carbon dioxide, hydrogen and steam by using external heat energy. Also, other intermediate gaseous products react with each other under similar conditions. The reduction process involves the interaction between carbon dioxide or water vapor molecules and hot charcoal particles in a gasifier. The carbon molecules in charcoal are highly reactive with oxygen; hence, there is a tendency to remove the oxygen atom from

water vapor and carbon dioxide at such high temperatures. The reduction process is completed when all available oxygen as single atoms is redistributed [35].

Since the gasification is an endothermic process, the activation energy for the reactions should be supplied to the gasifier to start the reactions and proceed them. If the necessary heat is supplied from an outer energy source, the method is called allothermal process. When the heat is provided by partial combustion of the coal in the reactor, then, it is called autothermal process. Gasification reactions typically occur at temperatures between 800°C and 1150°C for an atmospheric pressure gasifier. For the pressurized gasification process, reactor temperatures can reach up to 1400°C [36]. Reduction is the final stage in which the conversion rate, thermal efficiency and syngas quality are determined. There are four major gasification reactions: Boudouard, Water-Gas Shift, Carbon Monoxide Shift and Methanation reactions.

The principal chemical reactions involving carbon, carbon monoxide, carbon dioxide hydrogen, methane and steam during the gasification process of carbon-based feedstock in the form of coal, biomass or waste material particles, are given in Table 1.3. Enthalpy values of gasification reactions are given as well. The negative and positive enthalpy values indicate exothermic and endothermic reactions, respectively.

**Table 1.3** Main gasification reactions

| Reaction Type                  | Reaction                                  | Reaction Enthalpy [kJ/mol] |
|--------------------------------|-------------------------------------------|----------------------------|
| Oxidation reactions            | $C + 1/2 O_2 \rightarrow CO$              | -110.50                    |
|                                | $CO + 1/2 O_2 \rightarrow CO_2$           | -283.00                    |
|                                | $H_2 + 1/2 O_2 \rightarrow H_2O$          | -241.82                    |
|                                | $CH_4 + 0.5O_2 \rightarrow CO + H_2$      | +206.00                    |
| Boudouard reaction             | $C + CO_2 \rightleftharpoons 2 CO$        | +172.58                    |
| Char-Steam Gasification        | $C + H_2O \rightleftharpoons CO + H_2$    | +131.38                    |
| Methanation reaction           | $C + 2H_2 \rightleftharpoons CH_4$        | -74.90                     |
| Carbon monoxide shift reaction | $CO + H_2O \rightleftharpoons CO_2 + H_2$ | -41.98                     |

#### 1.1.4. Gasifier Types

A number of reactor types are available for different industrial applications of gasification process. These reactors differ from each other by their main structures and operational features such as fuel and gasification agent supply location, fuel particle size and chemical properties, ash removal method, directions of flow and solid particles etc. Simplified sketches of six common gasifiers are shown in Figure 1.13 [37].

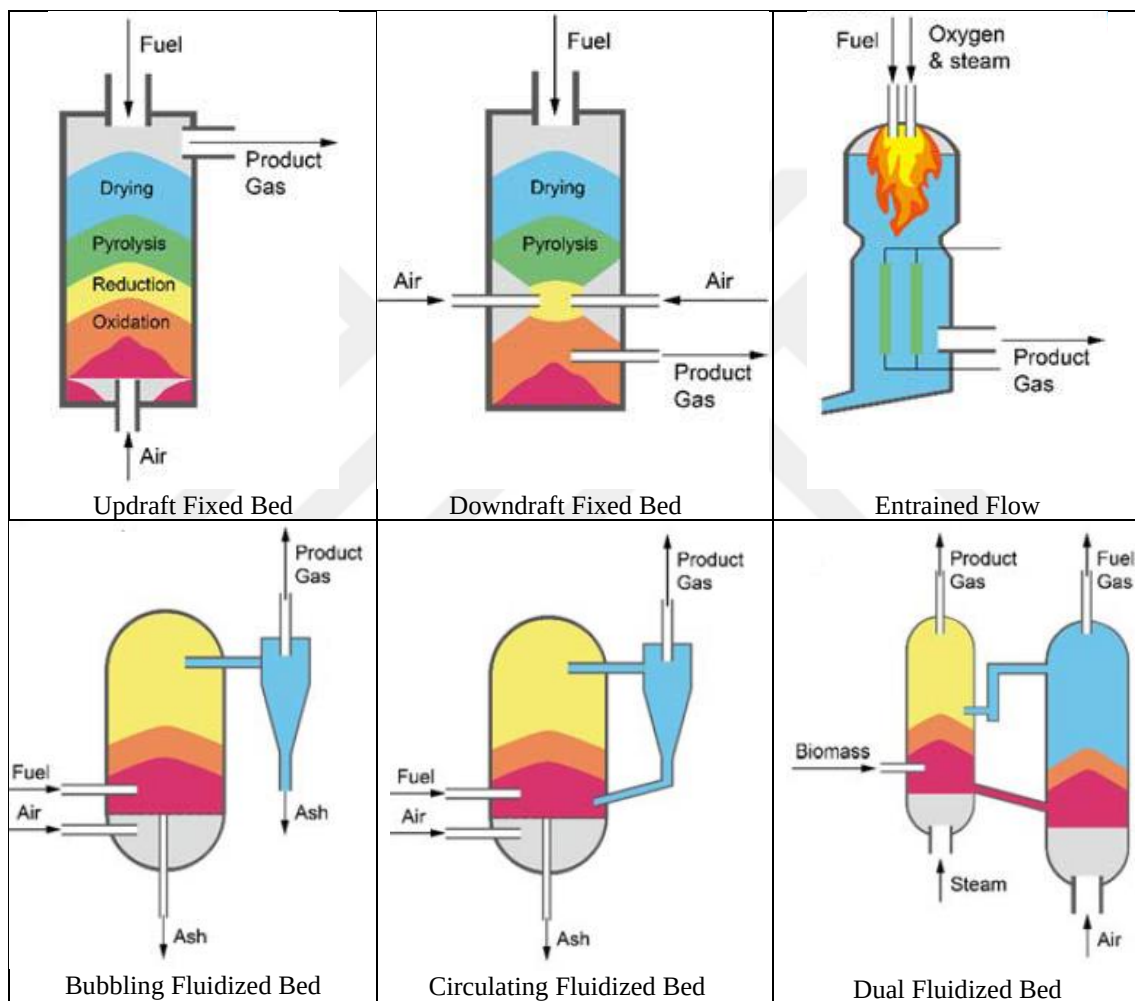


**Figure 1.13** Common types of gasification reactors in industry [37]

##### Updraft fixed bed gasifier

The fuel is fed in at the top of the reactor, and the intake of the gasification agent is at the bottom, hence the motion of fuel particles and gas flow are in opposite directions. Syngas leaves the reactor at the top. Ash and some unreacted char particles drop from the grate and residuals are collected at the bottom of the reactor. Simplicity in design and

operation, fuel flexibility and internal heat exchange capability are some benefits of updraft fixed bed gasifiers.

### **Downdraft fixed bed gasifier**

The fuel is fed in at the top same as updraft type, but the gasification agent is supplied also from the top or side walls of the reactor. Thus, the motion of fuel particles and gas flow are mainly in the same direction. Some of the fuel burns while passing through the inner throat of the reactor and provides enough energy for gasification reactions. Syngas leaves the reactor from the bottom and the ash and some unreacted char particles drop from the grate and residuals are collected at the bottom of the reactor. This method provides relatively high-quality syngas product. Updraft and downdraft fixed bed gasifiers are widely used in the gasification of biomass resources such as wood chips, barks, straws, rice husks, etc.

### **Entrained flow gasifier**

Pulverized fuel is fed into a gasifier with oxygen or steam at high pressure and temperature conditions. Some of the fuel burns at the top of the reactor and provides large provides enough energy for gasification reactions. Particle residence time is very low, so very high-quality syngas product is obtained at the outlet. The carbon conversion rate reaches up to 99% in this type of gasifier system. Due to high temperatures, the ash melts in the gasifier and is removed as molten slag. Entrained flow gasifiers are commonly used in integrated gasification combined cycle (IGCC) power plants for the utilization of high-rank coals. Coal feeding systems can be found in dry coal feed or coal water mixture form. Since the residence time of a fuel particle in a typical entrained flow reactor is very short and the usage of finer particle size (less than 75  $\mu\text{m}$ ) is required for the reactions to proceed completely, this method is rarely applied for biomass gasification [38].

### **Bubbling fluidized bed gasifier**

A particular amount of inert material such as sand is filled in the reactor bed region before start-up. The reactor bed is heated via a start-up burner that uses natural gas or fuel oil. At the same time gasification agent is supplied into the reactor with a velocity close to minimum fluidization to mix the bed material. Fuel is fed in from the side walls and the gasification agent is supplied from the bottom. Superficial gas velocities for bubbling

fluidized bed (BFB) systems usually vary between 1-2 m/s. Control of the bed temperature is very significant for the stable operation of a BFB reactor. Bed temperature is stabilized by adjusting the mass flow rate of the gasification agent to an optimum equivalence ratio.

### **Circulating fluidized bed gasifier**

The start-up procedure of a circulating fluidized bed (CFB) gasifier is the same as the bubbling fluidized bed type. Also, fuel is fed in from the side walls and a gasification agent is supplied from the bottom. The main difference between circulating and bubbling regimes is operating superficial velocity. Superficial gas velocity value in a CFB reactor is commonly selected between 4-7 m/s during the design stage. Due to higher velocity values, the significant amount of particle flow which includes unburnt char leaves the reactor with syngas. Thus, particles are separated by a mechanical separator (namely, cyclone) and returned to the reactor by a loop-seal or L-valve. Both circulating and bubbling fluidized bed gasifiers typically operate at temperatures between 750-900°C to avoid agglomeration issues caused by the formation of hot spots in the bed [39].

### **Dual fluidized bed gasifier**

A dual fluidized bed gasifier system involves two chemical reactors: a gasifier and a combustor. Fuel is fed into the fluidized bed gasification chamber from the side walls. Steam is fed as a gasification agent from the bottom. To provide enough heat for gasification reactions some of the fuel burns in a separate circulating fluidized bed combustion chamber and some of hot bed material is transferred to the gasification chamber. This type of gasifier also operates at temperatures between 750-900°C to avoid agglomeration in the gasification reactor, while higher temperature values are observed in a combustor.

### **Other types of gasifiers**

#### Plasma Gasifier

Plasma is a high-temperature and ionized gas capable of conducting electrical current. The interaction of the gas with the high voltage electricity causes the conversion of the

gas molecules into electrons and ions which yields a high temperature increase, may reach up to 6,000°C in theory. In the plasma gasification process, the gasifier is heated by a plasma torch located at the bottom of the reactor. The fuel and hot air are fed into a vertical reactor from the side walls of the vessel with atmospheric pressure conditions. Additional air or steam can be supplied at different height levels of the gasifier to control the process. The product gas exit temperature in a typical plasma gasification reactor is higher than 1000°C; hence, tar formation is avoided at such high temperature values. Molten slag which is another outcome of high reactor temperature, is removed from the bottom of the reactor. This process is suitable for the utilization of high moisture content biomass such as sewage sludge. High calorific value gas output consists of high volumetric ratios of hydrogen and carbon monoxide, and low carbon dioxide and tar content are some advantages of this gasification method. A typical application of the plasma gasification method is illustrated in Figure 1.14-a.

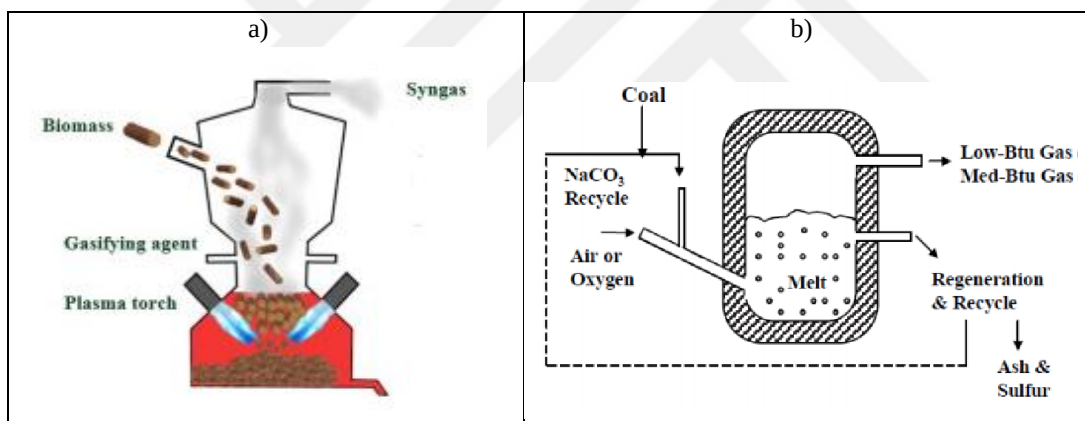


**Figure 1.14** a) Plasma Gasifier [40], b) Molten salt gasifier [41]

### Molten Salt Gasifier

The molten salt coal gasification process is an alternative approach to the other coal gasification techniques. This process involves a molten sodium carbonate bed to supply heat for gasification reactions under high-pressure conditions. High carbon conversion rate is one of the advantages of molten salt gasifiers. Both air and oxygen can be used as gasification agents in these systems. The capability of separation of SO<sub>x</sub> content from syngas products by using oxygen as gasification agent is another advantage of this type of gasifier. It is remarked in the literature that higher reactor pressure conditions improve the overall efficiency of molten salt gasifier systems [41].

#### **1.1.5. Numerical Analysis Approaches for Multiphase Flow in a Fluidized Bed Systems**

The application of gasification technology to low rank / high ash coals still needs more research studies to understand phenomena thoroughly and to design whole system and its components. The gasification process is complex since there are interactions of physical processes namely turbulence, combustion and particle dynamics. Due to the complexity of the system and the high cost of production, numerical modeling of these systems becomes compulsory. It's clear that analysis of the bed behavior with computational modeling of the fluidized bed and determination of the outputs from known inputs of the system before the production stage will decrease the cost of design and production.

Computational Fluid Dynamics (CFD) method is commonly used in fluidized bed gasification systems to investigate the physical behavior of the fuel and bed material and the chemical conversion of the fuel. Numerical modeling studies provide detailed information about the events that occur during the gasification process. Thus, pre-design of the industrial scale system can be done by using the data obtained from modeling of the lab scale systems.

Several mathematical models were developed in literature and industrial applications to calculate and improve thermal efficiency and the gas composition as a result of the gasification process of coal and biomass. In these studies, three primary methods are used generally. The first of these methods is the method that the Euler equations are solved for both continuous fluid phase and particle phase which represents the solid fuel particles in the physical domain (Eulerian - Eulerian Two Fluid Method / E-E TFM). The other one is the method that Euler equations are solved for continuous phase and Discrete Element Method (DEM) is applied for particle phase. The third method is that Euler equations are solved for continuous phase and Discrete Particle Method (DPM) is used for particle phase. On the other hand, some recent codes, which specialized in modeling the particle phase, become prominent.

DEM and DPM methods suppose particles as independent elements and solve equations for each particle individually. However, E-E TFM method assumes that the fluid and

particle phases are in interaction as two continuous media. Some general results were achieved from literature in which these computational methods are applied for the case of gasification/combustion of biomass or coal. It is shown that E-E TFM method is more suitable for fixed bed cases that contain higher particle phase density; the use of DEM/DPM methods is more convenient for fluidized bed cases that contain lower particle density to obtain accurate solutions. One of the major differences between the implementations of the E-E TFM, DEM and DPM methods for the investigation of fluidized bed applications is in the computational times. It is seen from numerical studies that DEM and DPM methods require much more computational time than E-E TFM approach. Moreover, it is understood that numerical analysis needs computational mesh with a huge number of elements due to the complexity of both chemical structure of fuel and the geometry. Hence, academic studies using DEM and DPM methods are generally carried out on the lab scale systems. Thereby, DPM and DEM methods are applied usually for lab-scale/pilot scale systems due to the high computational costs [42]. It is seen that high-performance GPU-based workstations and parallel computations are necessary for modeling industrial scale and 3-dimensional applications because of the huge number of particle contents in the physical domain.

Another output of the studies on the application of E-E TFM and DEM/DPM is that E-E TFM method gives less accurate results compared to experimental results, especially for emission values. Gerber et al. reported in their study published in 2011 that E-E TFM method gives less precise results, especially for H<sub>2</sub> and CO emissions than DPM method when it was compared with experimental results for gasification of the forest-based biomass [43]. Also, Myohanen and Hyppanen indicated in the study of coal gasification that E-E TFM method implemented by using Fluent software did not give accurate results for emission values [44].

Jeong et al. modeled an industrial coal gasification system with a two-stage entrained bed reactor through DPM method [45]. In this study, the effects of the different fuel particle sizes on reaction outputs were investigated and optimum fuel particle size was determined. Liu et al. examined the effect of the product distribution coefficient for char combustion in the 3-dimensional modeling of a circulating fluidized bed (CFB) system [46]. Numerical analysis was conducted by using three product distribution coefficients

for different air mass flow rates in this research. According to the findings, it is determined that the product distribution coefficient affects the gas composition even if it is a little.

Fluent software is widely used software in the literature. Nevertheless, in these studies, user-defined functions (UDF) were generated and implemented in Fluent in order to overcome the problems regarding insufficiencies of the software. Apart from that, there are other codes used in recent studies as CPFD Barracuda which uses MP-PIC (Multi-phase Particle in Cell) method [47] and MFIX (open-source code) [48]. It is reported that the latest studies provide more suitable approaches to the problems (application of drag force for different particle sizes and shapes, interaction with inter-particle static and dynamic friction, etc.) in modeling the physics of the particle phase. Chen et al. investigated the behavior of the Geldart-A particles in a circulating fluidized bed by using CPFD Barracuda which is MP-PIC based software [49]. This study showed that particle phase distribution in the control volume is more accurate than the results obtained from E-E TFM method. Similarly, Clark et al. stated in their article, which is about the use of sorbent to eradicate CO<sub>2</sub>, that numerical results of the sorbent particle distribution were consistent with numerical results [50].

It is seen from studies in the literature that,  $k-\epsilon$  turbulence model is mostly used for CFD modeling of the gasification process [42][45][46]. Additionally, in order to calculate the system efficiency more precisely, particle interactions must be modeled correctly due to the complex and multiphase structure of the gasification systems. CFD software like Fluent don't give satisfactory results yet for the numerical solution of particle dynamics unless appropriate UDF is used. On the other hand, it is argued that Barracuda software yields more precise results compared to other software for modeling the gasification systems in the literature [51].

In the design stage of the industrial scale fluidized bed system, the data obtained from numerical modeling of the lab scale system is the basis of the scale-up process. Therefore, the lab scale validation of the numerical models and the use of validated models in the scale-up stage is very significant for design studies of the industrial scale system. However, it was stated in the literature that some changes must be taken into consideration

for the models in the scale-up process to obtain acceptable computational times [44]. There is a big difference between industrial and lab scale systems, especially in terms of the number of particles in the bed. The flow model must be revised appropriately to represent these particles in the numerical model. The industrial-scale system should be modeled again by using the data obtained from the lab-scale system.

## **1.2. Aim of the Study**

The main purpose of this study is to investigate fluidized bed gasification systems by numerical and experimental methods to utilize local low-rank coal resources. Within the frame of the research studies, chemical composition of chosen lignite samples extracted from coal seams in Edirköy-Tekirdağ site is determined by characterization techniques. A lab scale fluidized bed system in TÜBİTAK MAM (The Scientific and Technological Research Council of Turkey – Marmara Research Center) is used in order to investigate gasification behavior of low-rank coal resources experimentally. System outputs and operating conditions and such as pressure drop, bed temperature, syngas composition etc., are examined by modeling the system numerically through a CFD code. The numerical results are validated by the experimental data measured from experimental rig.

The validated CFD model is used for scale up calculations in which industrial scale was modelled numerically. Design parameters of the industrial scale fluidized bed system are determined by evaluation of the fluidized bed behavior for different variables as superficial velocity, equivalence ratio, etc. Based on results of the numerical model validated from lab scale, the industrial scale system is examined for different case studies in which it is aimed to investigate pressure, velocity and particle distributions inside the gasification chamber, and maximize the solid circulation rate indicating the quality of the syngas produced from gasification process and overall thermal efficiency of the system.

Another objective of the thesis study is to determine the optimum operating condition parameters by considering various physical conditions, with the aim of increasing the efficiency of the fluidized bed system and decreasing the mass flow rate of the undesired product gases. Thus, it will be a contribution to the development of the fluidized bed gasification systems which make possible to utilize local coal resources with low calorific

value by advanced technologies and benefit them more efficiently. It is also thought that the systems improved by using outputs of this thesis will contribute to decreasing the energy dependency of our country to the imported resources.



## 2. MATERIALS AND METHODOLOGY

### 2.1. Coal Sample Preparation and Characterization

High ash content low-rank lignite samples are received from Edirköy/Tekirdağ coal site for experimental studies (Figure 2.1). Various sample characteristics are obtained with different pre-process operations on coal samples for the gasification experiments. Firstly, some of the samples are dried and/or washed. Then, coal samples are milled and screened to provide chosen particle size range of 0.5 – 1.0 mm. At the end of all processes, four different samples are obtained:

- Sample-1: As received conditions
- Sample-2: Dried
- Sample-3: Washed
- Sample-4: Washed & dried



**Figure 2.1** Lignite samples – As received

Thermogravimetric Analysis (TGA) measures the change in mass of a sample material as a function of temperature or time under a controlled environment. The composition and thermal characteristics of the sample for any kind of chemical process at specified conditions can be determined by this method. A typical TGA is performed for a sample by gradually increasing the temperature in a furnace. In TGA, mass loss is observed due to thermal degradation which involves the release of a volatile component or gaseous product of a chemical reaction such as combustion. While chemical reactions cause mass losses in the solid phase, there is not any change observed in physical properties such as melting. The weight of the sample is plotted against temperature or time to evaluate the

thermal effects on the material structure and to determine the thermal transitions. TGA is a fast and easy technique for observing the thermo-kinetic behavior and investigating the composition of solid fuels.

In this study, Leco TGA 701 is used as a thermogravimetry analyzer to perform proximate analysis of the coal samples and determine moisture, volatile, fix carbon and ash content (Figure 2.2-a). The calorific content of the samples (LHV and HHV) is evaluated by using Leco AC600 isoperibol calorimeter (Figure 2.2-b). Additionally, Leco TruSpec Micro device is used for ultimate analysis (Figure 2.2-c). This experimental tool directly measures Carbon, Hydrogen, Nitrogen and Sulphur content in the coal particles. Oxygen ratio is obtained by subtracting the sum of the measured component from 100%.

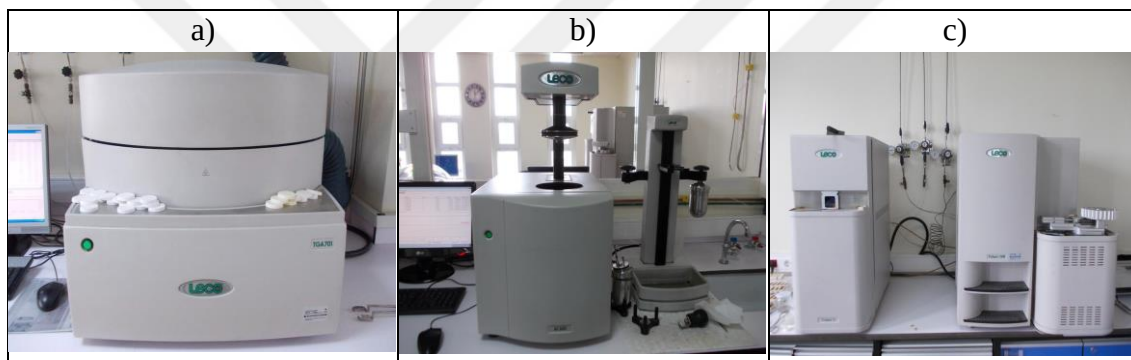


**Figure 2.2** Instruments used for coal characterization: a) Leco TGA701, b) Leco AC600, c) Leco TruSpec Micro

Approximate and ultimate analyses are carried out for each of these samples according to ASTM standards. ASTM D 2013 standard is used for sample preparation. ASTM D 7582-12 standard is considered in order to determine the moisture, volatile matter and ash content of the samples. ASTM 3172-13 standard is used to obtain fixed carbon content. The ultimate analysis is applied according to standard ASTM D 5373-13, ASTM D 3176-09 and ASTM 4239-13 standards. Table 2.1, Table 2.2, Table 2.3 and Table 2.4 show the proximate analysis, ultimate analysis and calorific value of Sample-1, Sample-2, Sample-3 and Sample-4, respectively, on both as received (original) and dry basis.

**Table 2.1** Coal Characteristics: Sample-1

| Content  | Unit  | Original | Dry B. | Content | Unit | Original | Dry B. |
|----------|-------|----------|--------|---------|------|----------|--------|
| Moisture | %     | 34.24    | -      | C       | %    | 16.21    | 24.65  |
| Volatile | %     | 21.06    | 32.03  | H       | %    | 2.10     | 3.2    |
| Ash      | %     | 36.36    | 55.3   | N       | %    | 0.43     | 0.65   |
| Fixed C  | %     | 8.34     | 12.67  | S       | %    | 2.85     | 4.34   |
| LHV      | cal/g | 1028     | 1850   | O       | %    | 7.80     | 11.86  |
| HHV      | cal/g | 1293     | 1966   |         |      |          |        |

**Table 2.2** Coal Characteristics: Sample-2

| Content  | Unit  | Original | Dry B. | Content | Unit | Original | Dry B. |
|----------|-------|----------|--------|---------|------|----------|--------|
| Moisture | %     | 18.28    | -      | C       | %    | 20.14    | 24.65  |
| Volatile | %     | 26.18    | 32.03  | H       | %    | 2.62     | 3.2    |
| Ash      | %     | 45.19    | 55.3   | N       | %    | 0.53     | 0.65   |
| Fixed C  | %     | 10.35    | 12.67  | S       | %    | 3.55     | 4.34   |
| LHV      | cal/g | 1411     | 1850   | O       | %    | 9.69     | 11.86  |
| HHV      | cal/g | 1607     | 1966   |         |      |          |        |

**Table 2.3** Coal Characteristics: Sample-3

| Content  | Unit  | Original | Dry B. | Content | Unit | Original | Dry B. |
|----------|-------|----------|--------|---------|------|----------|--------|
| Moisture | %     | 20.95    | -      | C       | %    | 47.68    | 60.32  |
| Volatile | %     | 32.08    | 40.59  | H       | %    | 4.10     | 5.19   |
| Ash      | %     | 16.29    | 20.61  | N       | %    | 0.90     | 1.14   |
| Fixed C  | %     | 30.68    | 38.81  | S       | %    | 3.55     | 4.49   |
| LHV      | cal/g | 3957     | 5151   | O       | %    | 6.52     | 8.25   |
| HHV      | cal/g | 4241     | 5364   |         |      |          |        |

**Table 2.4** Coal Characteristics: Sample-4

| Content  | Unit  | Original | Dry B. | Content | Unit | Original | Dry B. |
|----------|-------|----------|--------|---------|------|----------|--------|
| Moisture | %     | 9.35     | -      | C       | %    | 54.68    | 60.32  |
| Volatile | %     | 36.79    | 40.59  | H       | %    | 4.70     | 5.19   |
| Ash      | %     | 18.68    | 20.61  | N       | %    | 1.03     | 1.14   |
| Fixed C  | %     | 35.18    | 38.81  | S       | %    | 4.07     | 4.49   |
| LHV      | cal/g | 4618     | 5151   | O       | %    | 7.48     | 8.25   |
| HHV      | cal/g | 4863     | 5364   |         |      |          |        |

## 2.2. Experimental Setup

A BFB gasifier in TÜBİTAK Marmara Research Center premises is used in order to investigate the gasification characteristics of low-rank lignite samples in a lab-scale environment. The gasification reactor has a 20 kWth thermal input power capacity and a cylindrical section with a diameter of 82 mm. The height of the reactor is 2.09 m. A distributor plate exists at the bottom of the system to supply air into the reactor. A cyclone is located at the exit of the gasifier to hold ash particles. Both of gasification reactor shell and cyclone are made of corrosion-resistive stainless steel (AISI 310S).

Although this BFB gasifier has an autothermal structure, an additional external energy source is needed to reach steady-state operation temperature. Thus, two electric furnaces each has 650 mm height, are installed on the reactor walls to keep the operational temperature constant. There is an unheated region with a 270 mm height between two heaters. Each heater has a separate PID controller fed by data obtained from thermocouples inside the reactor. The main stream of the product gas goes to the chimney while a small stream is vacuumed by the gas analyzer to examine the composition as shown in Figure 2.3.

The fuel feeder system has two-stage screw feeders. Two screws having variable speed controller electric motors with 0,55 kW power are installed for both stages. In the first stage, coal is taken from the hopper, and coal mass flow rate is adjusted to the desired level with appropriate RPM values. At the end of the first screw, N<sub>2</sub> is supplied to the system. Coal particles and N<sub>2</sub> are mixed in the second stage to make the feeding process easier and ensure transient fuel uniformity at the gasifier inlet. The mass flow rate of supplied N<sub>2</sub> is measured by a rotameter before feeding the system. The fuel inlet of the gasifier has a circular section with an inner diameter of 28 mm and a height of 200 mm from the distributor plate.

Air is fed by using a compressor with a maximum flow rate of 70 l/min. A moisture trap is used after the compressor exit to ensure dry air is fed into the reactor. There is a flow controller to adjust the mass flow rate of air to chosen equivalence ratio (ER) values. The mass flow rate of air is also measured by a rotameter before feeding the system.

The distributor plate involves 180 air feeding holes with a diameter of 2 mm and an axis angle of  $45^\circ$  to ensure homogenous fluidization. The thickness of the distributor plate is 15 mm. The holes are equally-spaced at 5 different distances from the center: 14 mm, 30 mm, 46 mm, 62 mm and 78 mm. The distribution of the holes in these circles is 11, 24, 36, 48, and 61, respectively.

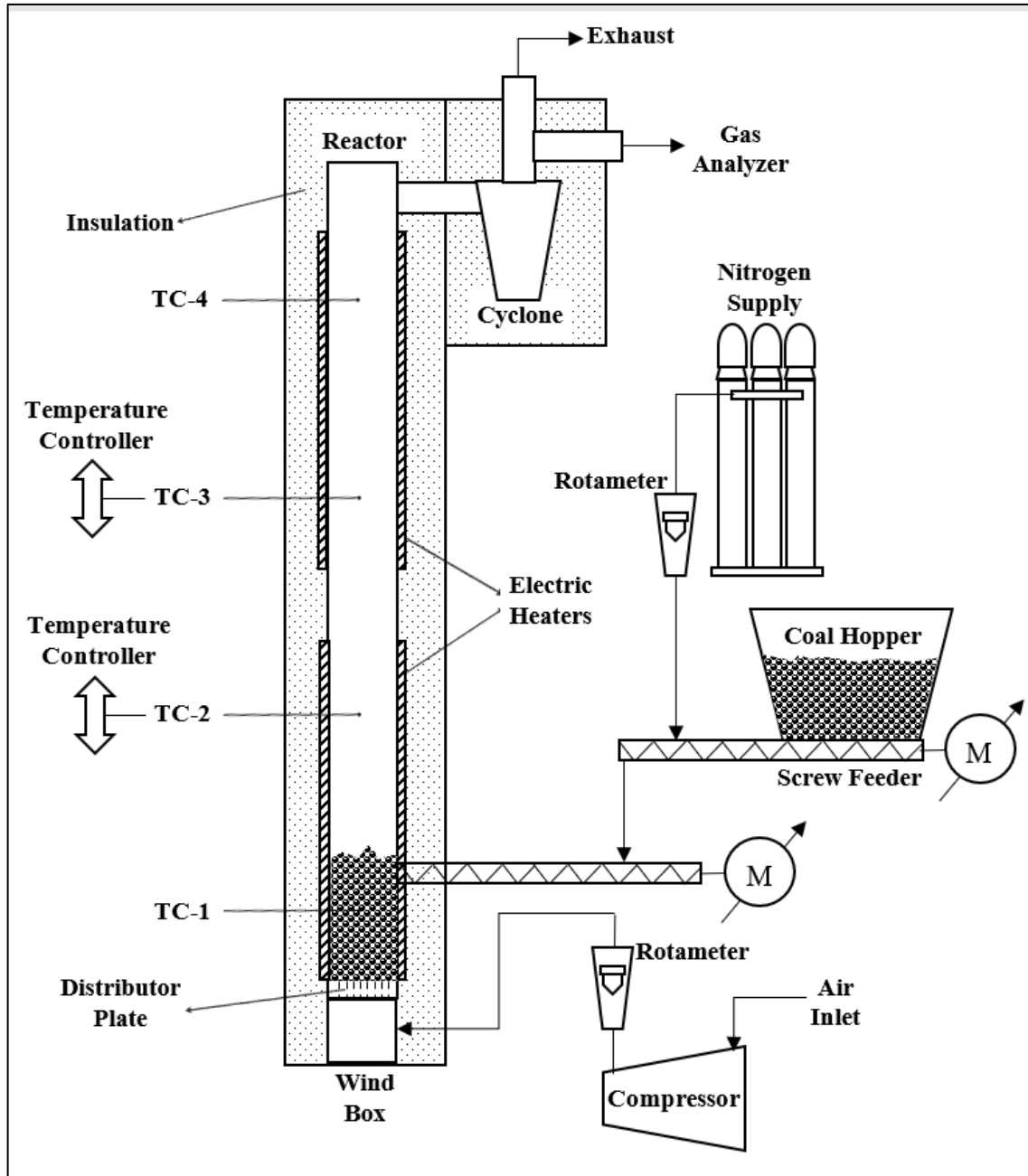


**Figure 2.3** Schematic of the experimental setup

Silica sand with an average particle diameter of 450 microns is used as inert bed material in lab-scale gasification experiments to stabilize the gasification process within the scope of the study. Silica sand consists of 99.2%  $\text{SiO}_2$ , 0.5%  $\text{Al}_2\text{O}_3$  and 0.1%  $\text{Na}_2\text{O}$ . The density of the sand particle is  $2500 \text{ kg/m}^3$ . The volume of initial bed material used in the experiments is 750 ml and the bed height is approximately 150 mm during all experiments. An image of the experimental setup is given in Figure 2.4.



**Figure 2.4** Experimental setup

Air mass flow rate is adjusted by considering the equivalence ratio, while the other main parameters such as the bed temperature, coal mass flow rate and nitrogen volumetric flow rate in the experiments are kept constant. Coal is fed to the reactor with Nitrogen at a constant temperature of  $20^\circ\text{C}$ .

Air is fed during bed material feeding in order to prevent blockage inside the gasifier. Moreover, a water-cooling system is used to cool the fuel feeding system and avoid the tar formation around the feeding screws and gasifier inlet. The PID controller of the

electrical heaters is set to an operating temperature of 825°C which is constant for all experiments in the scope of this study. Then, heaters are activated and air feeding is continued during heating to preserve heaters. When the bed temperature reaches 500°C, fuel feeding is started. After a transient period taking a couple of hours, the gasification system reaches a steady-state condition and an amount of gas is sucked from the exit of the cyclone by the vacuum pump of the gas analyzing system. In order to determine the composition of the synthetic gas, a sampling system is installed in the experimental setup. In the sampling system, syngas sample is directed to a filter including perlite particles, a silica gel bed and a glass wool bed in the interest of separating particulate matter. Hot syngas stream is cooled to 5°C before the inlet of the gas analyzer by same water-cooling system used for the fuel feeding system with a separate line. During the cooling process, most of the steam and tar content condenses and these species are captured in the filters. ABB AO2040 online gas analyzer is used for gas analysis. This gas analyzer is capable of measuring the volumetric ratio of CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub> and O<sub>2</sub> species and determining the composition of syngas on dry basis. The volumetric ratio of N<sub>2</sub> is calculated from the difference.

The experimental procedure of lab-scale gasification experiments is given in Figure 2.5.

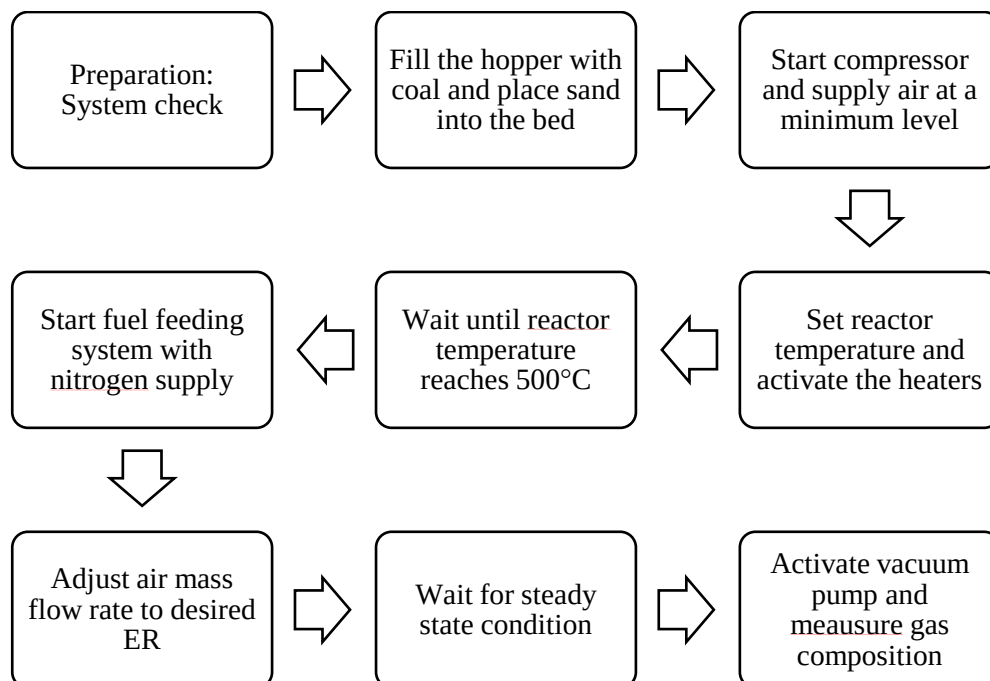


**Figure 2.5** Procedure of gasification experiments

The most important parameter for the experimental investigation of coal gasification process is the equivalence ratio which is a dimensionless variable that refers to the ratio of the actual mass flow rate of the gasification agent to the air mass flow rate needed for stoichiometric combustion condition.

$$ER = \dot{m}_{a,act} / \dot{m}_{a,sto} \quad (2.1)$$

In the case of stoichiometry, the air mass flow rate is calculated in units of kg-air/kg-fuel by using original basis elemental analysis results of the fuel as follows [12]:

$$\dot{m}_{a,sto} = \left[ 11.53C + 34.34 \left( H - \frac{O}{8} \right) + 4.34S \right] \quad (2.2)$$

$ER = 1$  yields stoichiometric condition where all the oxidant is consumed in combustion reactions and there is not any unreacted fuel remaining. Since the gasification is assumed to be a partial-combustion process, all ER values are smaller than one in the experiments. Five different ER values are used for Sample-1&Sample-2 while four different ER values are used for Sample-3&Sample-4 in this study. Air mass flow rate is adjusted to satisfy these values during experiments.

The lower heating value of the syngas produced during the gasification process can be obtained in units of MJ/Nm<sup>3</sup> by using volumetric fractions of the species through the equation below:

$$LHV = \frac{282.99xCO + 802.34xCH_4 + 241.83xH_2}{22.4} \quad (2.3)$$

Thermal efficiency of the gasification process can be defined in two ways: cold gas efficiency and hot gas efficiency. The difference between two definitions is that hot gas efficiency involves the contribution of sensible heat. Gasification efficiency can be expressed as follows:

$$\eta_{cold} = \frac{\rho_g \dot{V}_g LHV}{\dot{m}_{coal} LHV_{coal}} \quad (2.4)$$

$$\eta_{hot} = \frac{\rho_g \dot{V}_g (LHV + c_{p,g}(T_g - T_a))}{\dot{m}_{coal} LHV_{coal}} \quad (2.5)$$

where,  $c_{p,g}(T_g - T_a)$  denotes sensible heat term.

## 2.3. Numerical Approach

In the present work, an extensive reactive model for the coal gasification process is implemented into the MP-PIC method on a CPFD code Barracuda Virtual Reactor, which is known as a very efficient tool in modeling reactive multiphase flows in fluidized bed gasifiers and combustors. According to MP-PIC method, the discrete particles are tracked in a Lagrangian framework while the continuous flow phase is calculated under the Eulerian framework. Partial Donor Cell (PDC) method involves a finite volume approach is used for the discretization of the governing equations. The velocity and pressure variables of continuous phase are coupled by a semi-implicit pressure equation. The momentum, energy, and pressure equations are solved by using a conjugate gradient solver. Moreover, the turbulence in the multiphase flow is solved with a LES model. For the discrete phase, the drag coefficient of the particle is obtained by Wen-Yu/Ergun Drag Model. Particle-particle and particle-wall interactions are also taken into account and calculated with a particle stress function and momentum retention coefficients, respectively. This technique avoids intense computation effort for particle interactions and reduces the computational time significantly, especially for the reactive multiphase flow cases that involve very small time steps in this study. Additionally, differential equations that calculate species concentrations are solved using a stiff Ordinary Differential Equation (ODE) solver. Details of the numerical approaches are given in the sub-sections below.

### 2.3.1. Governing Equations for Continuous Phase

The 3-D unsteady equations for continuity, momentum and energy are solved to obtain the continuous phase characteristics of the flow. Continuity and momentum equations for a flow with discrete phase can be defined as follows [52]:

$$\text{Continuity: } \frac{\partial(\theta_f \rho_f)}{\partial t} + \nabla \cdot (\theta_f \rho_f u_f) = \delta \dot{m}_p \quad (2.6)$$

$$\text{Momentum: } \frac{\partial(\theta_f \rho_f u_f)}{\partial t} + \nabla \cdot (\theta_f \rho_f u_f u_f) = -\nabla p + F + \theta_f \rho_f g + \nabla \cdot (\theta_f \tau_f) \quad (2.7)$$

where  $\theta_f$  is the fluid volume fraction,  $\rho_f$  is the density of the fluid,  $u_f$  is the velocity vector,  $p$  is the pressure,  $F$  is the body force,  $g$  is the gravitational acceleration and  $\tau_f$  is

the viscous stress term.  $\delta \dot{m}_p$  is the mass transfer rate from the discrete phase to the continuous phase. Viscous stress term can be written by the following equation:

$$\tau_{f,ij} = \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \mu \delta_{ij} \frac{\partial u_k}{\partial x_k} \quad (2.8)$$

where  $\mu$  is the dynamic viscosity of the fluid.

The energy equation can be written as follows:

$$\frac{\partial(\theta_f \rho_f h_f)}{\partial t} + \nabla \cdot (\theta_f \rho_f h_f u_f) = \theta_f \left( \frac{\partial p}{\partial t} + u_f \cdot \nabla p \right) + \Phi - \nabla \cdot (\theta_f q) + S_h + \dot{q}_D \quad (2.9)$$

where  $h_f$  is the enthalpy,  $\Phi$  is the turbulent dissipation term,  $q$  is the heat flux,  $S_h$  is the energy exchange between the two phases and  $\dot{q}_D$  is the enthalpy diffusion term.

### 2.3.2. Governing Equations for Discrete Phase

The MP-PIC method which solves a transport equation for the particle distribution function,  $f$ , is used to compute the discrete phase field. In this method, the governing equations can be represented by the following expressions:

Fluid mass source:

$$\delta \dot{m}_p = - \iiint f \frac{dm_p}{dt} dm_p du_p dT_p \quad (2.10)$$

Velocity change on a particle:

$$\frac{du_p}{dt} = D_p(u_f - u_p) - \frac{1}{\rho_p} \nabla p - \frac{1}{\theta_p \rho_p} \nabla \tau_p + g + \iiint f \frac{dm_p}{dt} dm_p du_p dT_p \quad (2.11)$$

where  $m_p$  is the particle mass,  $u_p$  the velocity vector,  $T_p$  the temperature,  $\rho_p$  the density and  $\theta_p$  is the volume fraction of the particle.  $D_p$  is the drag function and  $\tau_p$  is the stress tensor. Particle forces on a control volume can be written as:

$$F = - \iiint f \left\{ m_p \left[ D_p(u_f - u_p) - \frac{\nabla p}{\rho_p} \right] - u_p \frac{dm_p}{dt} \right\} dm_p du_p dT_p \quad (2.12)$$

The energy equation for a particle can be written as:

$$C_v \frac{dT_p}{dt} = \frac{1}{m_p} \frac{k_f Nu_{f,p}}{d_p} A_p (T_f - T_p) \quad (2.13)$$

where  $C_v$  is the specific heat,  $d_p$  is the particle diameter and  $A_p$  is the surface area of the particle.  $Nu_{f,p}$  is the Nusselt number between the two phases. The energy exchange between the two phases can be defined as a source term:

$$S_h = \iiint f \left\{ m_p \left[ D_p(u_p - u_f)^2 - C_v \frac{dT_p}{dt} \right] - \frac{dm_p}{dt} \left[ h_p + \frac{1}{2} (u_p - u_f)^2 \right] \right\} dm_p du_p dT_p \quad (2.14)$$

### 2.3.3. Drag Model

In order to calculate the drag force on solid particles, there are several models available in the literature. Studies on the comparison of drag models with CPFD method for Geldart-B group particles showed that Wen-Yu/Ergun drag model resulted in more reliable agreement with experimental data (for example, Chen et al. [49]; Wang et al. [53]). Therefore, Wen-Yu/Ergun drag model proposed by Gidaspow is used in the simulations [54]. This model is a combination of Wen-Yu [55] and Ergun [56] models. The model utilizes the Reynolds number and particle volume fractions.

Wen-Yu drag model is expressed as:

$$D_{WY} = 0.75 C_d \frac{\rho_f}{\rho_p} \frac{|u_f - u_p|}{d_p} \quad (2.15)$$

where drag coefficient  $C_d$  is defined as follows:

$$C_d = \begin{cases} \frac{24}{Re} \theta_f^{-2.65} & Re < 0.5 \\ \frac{24}{Re} (1 + 0.15 Re^{0.687}) \theta_f^{-2.65} & 0.5 \leq Re \leq 1000 \\ 0.44 \theta_f^{-2.65} & Re > 1000 \end{cases} \quad (2.16)$$

It can be seen from the drag coefficient equation that, Wen-Yu model uses different correlations for different Reynolds regions. Stokes drag correlation ( $24/Re$ ) is used for low-Re numbers, while Schiller-Naumann drag correlation is applied for the transition region. High Reynolds regions are calculated by an initial drag coefficient, as well. Also, the drag force can be adjusted by a multiplier specified in this model.

On the other hand, Ergun drag model is expressed as:

$$D_E = \left( \frac{1800}{Re} \frac{\theta_p}{\theta_f} + 2 \right) \frac{\rho_f}{\rho_p} \frac{|u_f - u_p|}{d_p} \quad (2.17)$$

$$Re = \frac{\rho_f d_p |u_f - u_p|}{\mu_f} \quad (2.18)$$

Drag function for Wen-Yu/Ergun model is expressed as follows:

$$D_p = \begin{cases} D_{WY} & \theta_p < 0.75\theta_{cp} \\ \frac{\theta_p - 0.85\theta_{cp}}{0.85\theta_{cp} - 0.75\theta_{cp}} (D_E - D_{WY}) + D_E & 0.75\theta_{cp} < \theta_p < 0.85\theta_{cp} \\ D_E & \theta_p > 0.85\theta_{cp} \end{cases} \quad (2.19)$$

Sphericity of a particle can be defined as follows:

$$\psi = \frac{\text{Surface area to volume ratio of a sphere}}{\text{Surface area to volume ratio of the particle}} \quad (2.20)$$

The particles are considered as perfect spherical-shaped materials according to Wen-Yu/Ergun drag model. Hence, the sphericity of the particles is assumed to be equal to 1 in the calculations.

#### 2.3.4. Heat transfer coefficients

The local wall heat transfer coefficient,  $h_w$ , comprising two main terms [57]: the heat transfer coefficient of the continuous phase ( $h_f$ ), and the weighted contribution of the particles on heat transfer coefficient ( $h_p$ ) by a function of the particle volume fraction at

the wall ( $f_p$ ). In the numerical model, there is not any direct heat transfer between particles and walls. Heat is firstly transferred between the walls and continuous fluid phase, then it is transferred between fluid and the particles as the discrete phase and vice versa. Also, it can be clearly seen in the particle energy equation that there is not any term indicates the wall temperature, which also shows the heat transfer from or to wall and from the wall to the gas is governed by the heat transfer coefficient  $h_w$ .

$$h_w = h_f + f_p h_p \quad (2.21)$$

where,  $f_p$ ,  $h_f$  and  $h_p$  can be expressed as follows:

$$f_p = 1 - e^{-10 (\theta_p / \theta_{cp})} \quad (2.22)$$

$$h_f = (c_0 Re_L^{n_1} Pr^{n_2} + c_1) \frac{k_f}{L} + c_2 \quad (2.23)$$

$$h_p = (c_3 Re_p^{n_3} Pr^{n_2} + c_1) \frac{k_f}{d_p} \quad (2.24)$$

In equations between 2.16 and 2.18,  $L$  is the cell size,  $c_0$ ,  $c_1$ ,  $c_2$ ,  $c_3$ ,  $n_1$ ,  $n_2$  and  $n_3$  are input coefficients of the model.  $c_0$ ,  $c_1$ ,  $c_2$  and  $c_3$  have a value of 0.46, 3.66, 0 and 0.525, respectively. Additionally,  $n_1$ ,  $n_2$  and  $n_3$  are equal to 0.5, 0.33 and 0.75, respectively. These coefficients are the default values of CPFD software based on the correlations proposed by Leva and Grummer [58] and Douglas and Churchill [59]. The Reynolds numbers for each phase can be defined as follows:

$$Re_L = \frac{\rho_f u_f L}{\mu} \quad (2.25)$$

$$Re_p = \frac{\rho_p u_p L}{\mu} \quad (2.26)$$

where  $u_f$  is the local fluid velocity and it is calculated by CPFD-Barracuda during computations for each cell. The fluid-to-particle heat transfer coefficient,  $h_{fp}$ , which

indicates the effectiveness of heat transfer between the continuous phase and the discrete phase, is defined as follows:

$$h_{fp} = (c_4 Re^{n_4} Pr^{0.33} + c_5) \frac{k_f}{d_p} \quad (2.27)$$

where  $c_4$ ,  $c_5$  and  $c_6$  and  $n_4$  are input coefficients with values of 0.37, 0.1, 0 W/(m<sup>2</sup>K) and 0.6, respectively, which are substantially fit the experimental data presented by Turton et. al. [60]

### 2.3.5. Particle-Particle Interaction:

Interactions between particles are modeled using a stress function and neglecting the size and velocity of particles in the numerical method. The particle stress function,  $\tau_p$ , is defined by Snider [61] as a function of the particle volume fraction. The stress function is implemented in the numerical model using following equation:

$$\tau_p = \frac{10p_s\theta_p^\beta}{\max[\theta_{cp} - \theta_p, \varepsilon(1 - \theta_p)]} \quad (2.28)$$

where  $p_s$  is a pressure constant,  $\theta_{cp}$  is close pack volume fraction,  $\beta$  is a model constant that varies between 2 and 5 as recommended range,  $\varepsilon$  is a very small coefficient on the order of  $10^{-8}$ . Model constants of particle stress approach with recommended values have a negligible effect on the particle behavior for dilute phase. When the particle volume fraction of a region reaches to close pack value, the stress model begins to affect particles in or moving towards the region.

The close pack volume fraction indicates the maximum possible volume fraction of particles when they are packed randomly in a constant - static volume. The close pack volume fraction is practically obtained from the ratio of the bulk density to solid particle density of the particles: Typical value of the close pack volume fraction is between 0.56 and 0.64 in application.

Application of the stress model prevents particles from entering a region with a close-pack volume fraction. In this case, a particle that approaches to a close pack region is redirected. The redirection physics of a specific solid particle is determined by the

maximum momentum redirection value as a collision parameter. The particle stress tensor determines redirected momentum vector of a particle randomly by considering the given particle incidence angle and a maximum redirection value.

### **2.3.6. Particle-to-Wall Interaction**

Three mechanisms are used to model the physics of particle deflection from a wall boundary:

- Normal-to-wall momentum retention
- Tangent-to-wall momentum retention
- Diffuse bounce

Normal-to-wall momentum retention parameter is the fraction of the remaining normal component of the particle momentum after interaction with the wall. When this parameter is equal to 1, it yields a perfectly elastic collision where the particle is deflected with the same velocity it had before the interaction. Zero retention value indicates that all momentum is lost during collision. A typical value of 0.5 can be used for the normal-to-wall momentum retention of soft particles, while 0.85 or higher is appropriate for hard particles.

Tangent-to-wall momentum retention is the ratio of the remaining tangential component to particle momentum after interaction with the wall. A practical range of 0.85-0.98 can be chosen for the tangent-to-wall momentum retention. The numerical implementation of the normal-to-wall and tangent-to-wall momentum retention parameters is sketched in Figure 2.6.

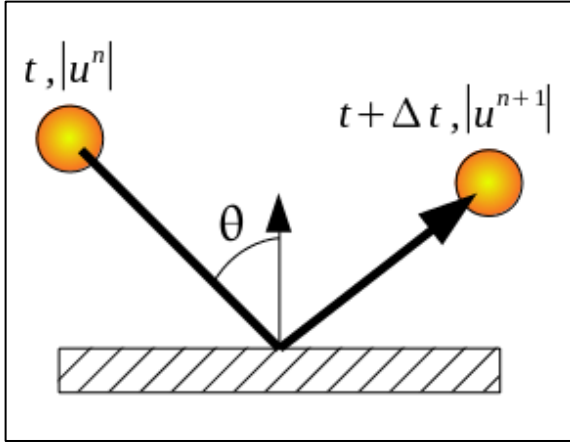


**Figure 2.6** Particle momentum retention model [62]

The particle velocity is calculated by taking normal and tangential momentum losses and the angle of impact into account as described follows:

$$|u^{n+1}| = [(r_T - r_N)(1 - \cos \theta) + r_N]|u^n| \quad (2.29)$$

where,  $u^{n+1}$  and  $u^n$  is particle speed values after and before the wall impact, respectively.  $r_N$  and  $r_T$  are retention factors in normal and tangential directions, respectively. When a solid particle hits a wall boundary with a narrow-angle, there is a small change in the momentum of the particle. In general, momentum loss in the normal component is greater than the loss in the tangential direction.

The diffuse bounce parameter applies a scatter function to particle motion after interaction with the wall. A value of 0 means that the new velocity vector direction for the particles after collision with the wall is computed with no scatter. When a particle cloud collides with a wall with the same velocity vector, then all particles will have an identical velocity vector to each other after deflection (Figure 2.7). However, in the reality, particles are not perfectly spherical and walls are not perfectly smooth. Hence, an average vector is calculated after the collision and some deviation from the average vector for each individual particle is computed according to the model coefficients. Higher values of diffuse bounce parameter yield more scattering and more computational time cost.

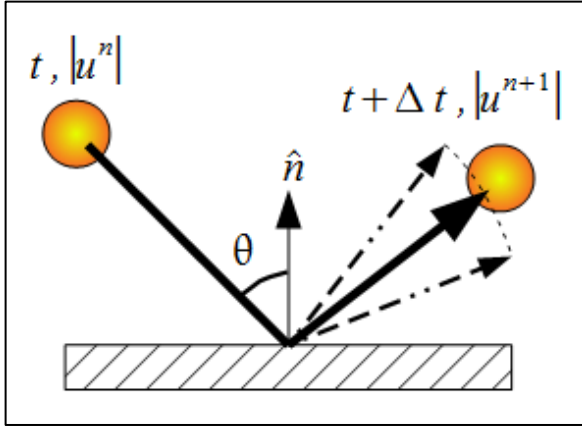


**Figure 2.7** Diffuse bounce model [62]

A stochastic diffuse bounce model is implemented and the scattering function is specified by a pseudo wall-roughness coefficient with a range of 0-5. While zero wall-roughness coefficient refers to a smooth wall, 5 yields a highly rough surface. The maximum scattering angle can be obtained at 15.5 degrees for a normal-to-wall impact condition by using this model. Particles that collide with a wall at a narrow-angle have a near specular bounce regardless of the roughness coefficient. The diffuse bounce model has not any contribution to the magnitude of the particle velocity after wall impact, but the velocity direction of the particle is affected by the diffuse bounce.

### 2.3.7. Turbulence Model

Turbulent viscosity was solved during computations by using Large-Eddy Simulation (LES) model [64]. In the LES model, the large eddies are resolved from the governing flow equations and the sub-grid turbulence is determined by a numeric model. As with other turbulence models, there is no accepted choice for a sub-grid scale (SGS) turbulence model for dense particle flow. SGS model calculates an eddy viscosity based on the notion that the effect of the Reynolds stress is increased transport and dissipation. The SGS eddy viscosity is obtained by the following equations:

$$\nu_t = C_s \rho_f \theta_f \Delta^2 |\bar{S}| \quad (2.30)$$

where,

$$|\bar{S}| = \sqrt{2S_{ij}^2} \quad (2.31)$$

$$\Delta = (\delta x \delta y \delta z)^{1/3} \quad (2.32)$$

$C_s$  is the Smagorinsky constant which can be considered as a function of Reynolds number, geometry, and particle volume fraction in a multiphase flow field. For a channel flow, it has a value of 0.005 and can be larger or smaller with a typical value on the order of 0.01. The time scale is defined as the function of the strain rate tensor,  $1/|\bar{S}|$  and  $\Delta$  is the filter-width length scale.

### 2.3.8. Advection Numerical Scheme

In a typical numerical analysis of a flow with finite volume method, velocity values are obtained at cell faces, while other quantities such as mass, pressure and temperature are calculated at cell centers. When the advection term in mass, momentum, or energy equations is calculated within a computational cell, velocity values of each face can be estimated by using the distance of the face from the cell center with an advection scheme. Partial Donor Cell (PDC) method proposed by Amsden et. al. [65] is used as an advection scheme for the numerical analyses applied in the scope of this study. PDC scheme can be considered as a weighted average of central difference and upwind schemes.

In all advection schemes, the quantity of interest at the cell faces  $Q$ , which can stand for any of variables such as velocity, pressure, turbulence intensity, etc., is calculated from the values of this quantity in two adjacent cells: cell-1 and cell-2. A variable,  $\varphi$ , is used as an indicator of the fluid flow direction. When  $\varphi$  is positive, the flow direction is from cell 1 to cell 2. In the case of a negative indicator value, the flow direction is from cell 2 to cell 1. The flow direction indicator is defined as follows:

$$\varphi = \frac{u_1 A_1 \theta_1 \rho_1 + u_2 A_2 \theta_2 \rho_2}{2} \quad (2.33)$$

In the first order upwind scheme, the quantity of interest at a cell face between two adjacent cells,  $Q_{1/2}$ , is assumed to have the value of the cell from which mass is flowing:

$$Q_{1/2} = \begin{cases} Q_1 & \varphi > 0 \\ Q_2 & \varphi < 0 \end{cases} \quad (2.34)$$

As an effective implementation of higher-order convection schemes for implicit finite volume analyses, upwinding with a deferred correction method involves a correction term,  $\Delta Q$ , added to the upwind advection scheme. This term is obtained for each iteration at the previous time step. Upwind with deferred central difference correction scheme procedure can be described as follows:

$$Q_{1/2} = \Delta Q + \begin{cases} Q_1 & \varphi > 0 \\ Q_2 & \varphi < 0 \end{cases} \quad (2.35)$$

where,

$$\Delta Q = \begin{cases} (Q_2 - Q_1)/2 & \varphi > 0 \\ (Q_1 - Q_2)/2 & \varphi < 0 \end{cases} \quad (2.36)$$

According to the application procedure of PDC differencing method, the first step is the determination of donor (upstream) and acceptor (downstream) cells in which quantities are  $Q_d$  and  $Q_a$ , respectively.

$$Q_d = \begin{cases} Q_1 & \varphi > 0 \\ Q_2 & \varphi < 0 \end{cases} \quad (2.37)$$

$$Q_a = \begin{cases} Q_2 & \varphi > 0 \\ Q_1 & \varphi < 0 \end{cases} \quad (2.38)$$

Then  $Q_{1/2}$  can be calculated by using a weighting factor,  $\Phi$ , as follows:

$$Q_{1/2} = [Q_d(1 + \Phi) + Q_a(1 - \Phi)]/2 \quad (2.39)$$

where,

$$\Phi = \alpha_0 + \beta_0 C \quad (2.40)$$

$$C = \Delta t \frac{2|u_1 A_1 + u_2 A_2|}{\theta_1 V_1 + \theta_2 V_2} \quad (2.41)$$

$C$  is a Courant number based on fluid velocity. The weighting factor,  $\Phi$ , varies between 0 and 1 with adjusted  $\alpha_0$  and  $\beta_0$  coefficients. For  $\alpha_0 = 0$  and  $\beta_0 = 0$ , the convection scheme becomes center differencing approach which is unconditionally unstable without enough diffusion. The convection scheme becomes upwind for the case of  $\alpha_0 = 1$  and  $\beta_0 = 0$  which tends to be stable but over-diffusive. In order to ensure enough diffusion and stability,  $\alpha_0$  and  $\beta_0$  values are taken 0.3 and 1, respectively.

## 2.4. Gasification Chemistry

Gasification process involves many different chemical and physical reactions as mentioned briefly in Chapter 1. Various chemical mechanisms have been developed to define, simplify and model the gasification chemistry over the years. The detailed structure of the chemical and physical mechanisms in a gasifier is not completely understood, even though the latest studies in the last few decades tend to close the gap between operational data obtained from industrial applications and the results of the lab-scale systems. Chemical mechanisms and reaction rates used in the numerical calculations are based on the results of these investigations in literature [66]-[74]. Alternative mechanisms and methodologies for some gasification reactions are also given in this section.

Chemical reactions in a gasifier can be classified into two main groups: Heterogeneous reactions and Homogenous reactions. A typical homogeneous reaction occurs uniformly through a single phase including both reactants and products. In contrast, a heterogeneous reaction takes place within the boundary of two or more phases. Thus, these reactions are sometimes called “surface reactions” in the literature. In the scope of this work, heterogeneous reactions are assumed to occur on the surface of the solid char particles that have %100 Carbon content.

Since the total computation time of a reactive multiphase flow highly depends on the total number of the species and related chemical reaction steps, dominant reaction pathways which have a substantial contribution to the main species concentrations of the syngas, are considered in the numerical analysis. The formation mechanism and other reactions of some species such as larger hydrocarbon molecules are neglected due to their relatively small effects on whole mechanism outputs. This approach yields a considerable decrease in the computational cost of a numerical analysis of a complex transient reactive multiphase flow [75]. Principle heterogeneous and homogenous reactions of the gasification process are discussed under the following sub-section.

Devolatilization and pyrolysis are complex and strongly interlinked processes that depend on both the molecular structure of parent coal and atmospheric conditions in a gasifier

reactor. Hence, a number of detailed models mostly based on comprehensive experimental data have been developed. In this thesis study, two different devolatilization models are used in the simulation and the results are compared to each other. Due to its complexity, details of the devolatilization modeling process are given in a separate section. Approach for Sulfur and Nitrogen reactions are also explained individually.

#### 2.4.1. Principle Gasification Reactions and Kinetic Models Used in CPFD Model

##### Heterogeneous Reactions – Char Conversion

A char particle mostly consists of fix carbon and ash components, with small amounts of other elements. It is a common assumption that only carbon remains as a reactive component in the char structure after the devolatilization process. Char conversion process starts after primary devolatilization phase, but it can also take place simultaneously with secondary pyrolysis phase [90]. The remaining carbon content is consumed by several heterogeneous reactions in the conversion process. Char conversion reactions for a coal gasification process is listed in Table 2.5:

**Table 2.5** Heterogenous reactions for char conversion

| Reaction Description    | Reaction                               | Reaction No |
|-------------------------|----------------------------------------|-------------|
| Char combustion         | $C(s) + O_2 \rightarrow CO_2$          | R1          |
| Partial char oxidation  | $C(s) + 0.5O_2 \rightarrow CO$         | R2          |
| Char-Steam Gasification | $C(s) + H_2O \leftrightarrow CO + H_2$ | R3          |
| Boudouard reaction      | $C(s) + CO_2 \leftrightarrow 2CO$      | R4          |
| Methanation             | $C(s) + 2H_2 \leftrightarrow CH_4$     | R5          |

Walker et al. proposed a three-zone approach for heterogeneous char reactions [76]. They remarked that char combustion is controlled by chemical kinetics at low temperatures, oxygen pore diffusion at moderate temperatures, and oxygen bulk diffusion at high temperatures as illustrated in Figure 2.8.

- In Zone I, reactions take place at low temperatures and the surface reaction rates are relatively slow. In this region, the rate depends on the reactivity of the solid

particle only. Apparent (measured) activation energy is equal to the true activation energy. The reactant gases diffuse into the center of the particle and the particle reacts internally at a constant diameter with a decreasing particle density, leaving an unreacted core.

- In Zone II, the concentrations of reactive species are very small in closer regions to the particle surfaces. The diffusion rate of reactive gases into the interior regions of the char particle through the porous structure is faster than their consumption rate. The apparent activation energy of Zone II is half of the true activation energy. The char particle reacts both internally and externally with a decrease in both particle diameter and particle density.
- In Zone III, reactions occur only on the surface of the particle at high temperatures due to the fast intrinsic reaction. The diffusion rate of the reactant gas into the core is very small and the reaction rate is controlled by diffusion through the surface. In this zone, the particle diameter decreases continuously while the particle density is constant. This char conversion step is also called shrinking-core behavior [90]. The apparent activation energy of this zone is close to zero.

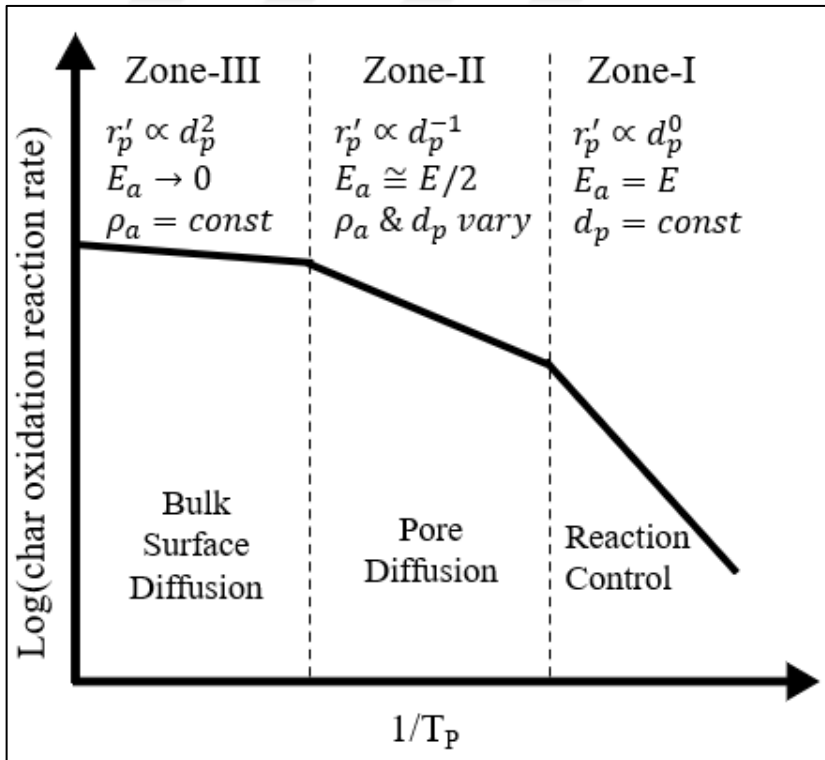


**Figure 2.8** Rate-controlling regimes for char oxidation – Three zone approach ( $r'_p$ : the total normalized particle reaction rate), adapted from [96]

Char conversion process may vary and be more complex than this three-zone approach depending on the type of the oxidizers with different reactivities toward the carbon in the char. For example, Boudouard reaction ( $C - CO_2$  gasification) may occur in a transition regime while solid carbon oxidation is controlled by film diffusion. Additionally, the concentrations of  $O_2$  and  $CO_2$  on the solid particle surface is also affected by the rate of  $CO$  oxidation in the continuous phase. Hence, the conversion rate of a char particle is determined by both homogenous reactions around the particle, diffusion rate of the surrounding gas into the particle and interior/core reactions [96].

Two main approaches are available for numerical modeling of the chemistry in a reactor: Volume average chemistry (VAC) & Discrete particle chemistry (DPC). VAC approach is based on the analysis at the cell level by using the physical properties of continuous phase. Hence, VAC is the most suitable method for the calculation of homogenous reactions. In DPC approach, the chemical reactions and all other related physical properties are calculated individually for each computational particle in a Lagrangian frame. Despite its higher computational cost due to these individual calculations, usage of DPC approach yields higher resolution and better accuracy for the implementation of heterogeneous reactions in the model. Moreover, it is possible to use VAC approach for heterogeneous reactions as well, by using the conversion of solid-dependent terms into volume-average form. In contrast, DPC approach cannot be used for homogeneous reactions.

VAC and DPC methods involve different calculation methods for both physical and chemical properties. The general form of Arrhenius type chemical rate equations can be expressed for VAC and DPC methods by following equations:

$$r_{VAC} = A_0 T^m P^n \rho_f^p \theta_f^t \exp\left(-\frac{E}{T} - E_0\right) \Phi_s \quad (2.42)$$

$$r_{DPC} = A_0 T^m P^n \rho_f^p \theta_f^t (N_p/V)^v \exp\left(-\frac{E}{T} - E_0\right) \Phi_s \quad (2.43)$$

where  $A_0$  is the exponential factor,  $T$  [K] is the temperature,  $E$  [K] is the activation energy divided by the universal gas constant.  $m$ ,  $n$ ,  $p$  and  $t$  are power constants for temperature, pressure, density and fluid volume fraction, respectively.  $\Phi_s$  represents the effect of solid particle property. It can be seen from the general form of rate equations,  $N_p/V$  term with

$v$  constant is additionally used to convert the volume average rate equation into discrete form. The expressions of physical variables for VAC and DPC methods are compared in Table 2.6. It should be taken into consideration that units of mass and area properties differ for each model. While the area unit is  $[m^2]$  in DPC method, it is  $[m^2/m^3]$  for VAC method. Similarly, mass unit is  $[kg]$  in DPC method, it is  $[kg/m^3]$  for VAC method.

**Table 2.6** Calculation of physical properties with VAC and DPC approaches for a solid particle and it's any  $i$ -component

| Physical property                            | Symbol         | VAC                                                                                                            | DPC                                                       |
|----------------------------------------------|----------------|----------------------------------------------------------------------------------------------------------------|-----------------------------------------------------------|
| Volume fraction of $i$ -component            | $\theta_i$     | $\frac{1}{V_{cell}} \sum_p \frac{m_{p,i}}{\rho_i}$                                                             | $\frac{1}{V_{cell}} \sum_p \frac{m_{p,i}}{\rho_i}$        |
| Volume fraction of all particles in the cell | $\theta_{all}$ | $\frac{1}{V_{cell}} \sum_p \sum_i \frac{m_{p,i}}{\rho_i}$                                                      | $\frac{1}{V_{cell}} \sum_p \sum_i \frac{m_{p,i}}{\rho_i}$ |
| Surface area of $i$ -component               | $a_i$          | $\frac{1}{V_{cell}} \sum_p \frac{m_{p,i} a_{p,i}}{\rho_i v_p}$                                                 | $\frac{m_{p,i}}{m_p} a_p$                                 |
| Surface area of all particles in the cell    | $a_{all}$      | $\frac{1}{V_{cell}} \sum_p a_p$                                                                                | $a_p$                                                     |
| Diameter of $i$ -component                   | $d_i$          | $6 \left( \sum_p \frac{m_{p,i}}{\rho_i} \right) \left( \sum_p \frac{m_{p,i} a_{p,i}}{\rho_i v_p} \right)^{-1}$ | $\frac{m_{p,i}}{m_p} d_p$                                 |
| Mean Diameter                                | $d_{all}$      | $d_{32} = 6 \frac{\sum_p v_p}{\sum_p a_p}$                                                                     | $d_p$                                                     |
| Mass of $i$ -component                       | $m_i$          | $\frac{1}{V_{cell}} \sum_p m_{p,i}$                                                                            | $m_{p,i}$                                                 |
| Mass of all particles in the cell            | $m_{all}$      | $\frac{1}{V_{cell}} \sum_p \sum_i m_{p,i}$                                                                     | $\sum_i m_{p,i}$                                          |

More than ten kinetic models are available in the literature in order to predict and describe the actual chemistry of char conversion [99]. The closest representation of reality with a simple mathematical approach yields an optimized chemistry model with reasonable computational cost. Three kinetic models for char conversion are considered: the progressive-conversion model, the shrinking unreacted-core model and random pore model.

In progressive-conversion model, the solid char particle is converted continuously and its carbon content is consumed progressively with a constant particle diameter assumption as shown in Figure 2.9-a.

When a partially reacted solid coal particle is examined closely, it can be easily seen that unreacted solid core is surrounded by an ash layer. By considering this, the shrinking-core model separates the particle into two regions: an outer ash layer and an interior char core. According to the shrinking-core model, overall particle diameter remains constant during char conversion while the thickness of the ash layer increasing unreacted core diameter is decreasing (Figure 2.9-b).

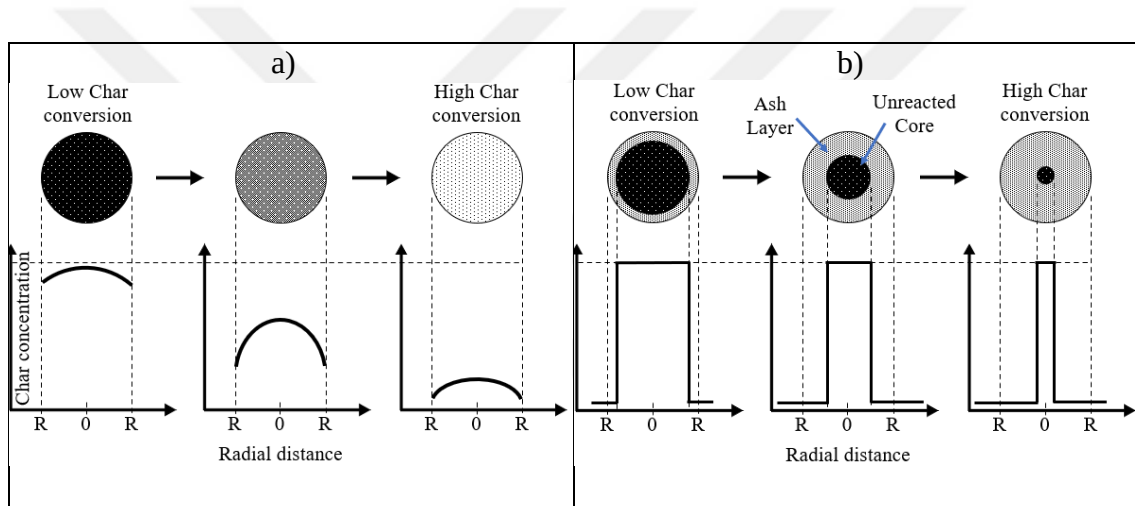


**Figure 2.9** Char conversion kinetic models a) Progressive-conversion model, b) Shrinking-core model, adapted from [99]

Although the model uses a homogenous layer thickness and spherical core assumptions, the shrinking-core model represents actual particle reactivity more closely than the progressive conversion model in most cases. The shrinking-core model is useful for the investigation of the combustion/gasification behavior of coal and biomass particles. A representation of the shrinking-core model is given in Figure 2.10.

Shrinking-core model was firstly proposed by Yagi and Kunii [100] and they defined the model with five steps:

- Diffusion of gaseous reactant through a film layer to the particle surface.
- Diffusion of gaseous reactant through an ash layer to the unreacted core

- The reaction of gaseous reactant with char in the core
- Reverse diffusion of the gaseous products to the exterior solid surface
- Reverse diffusion of the gaseous products into the main stream.

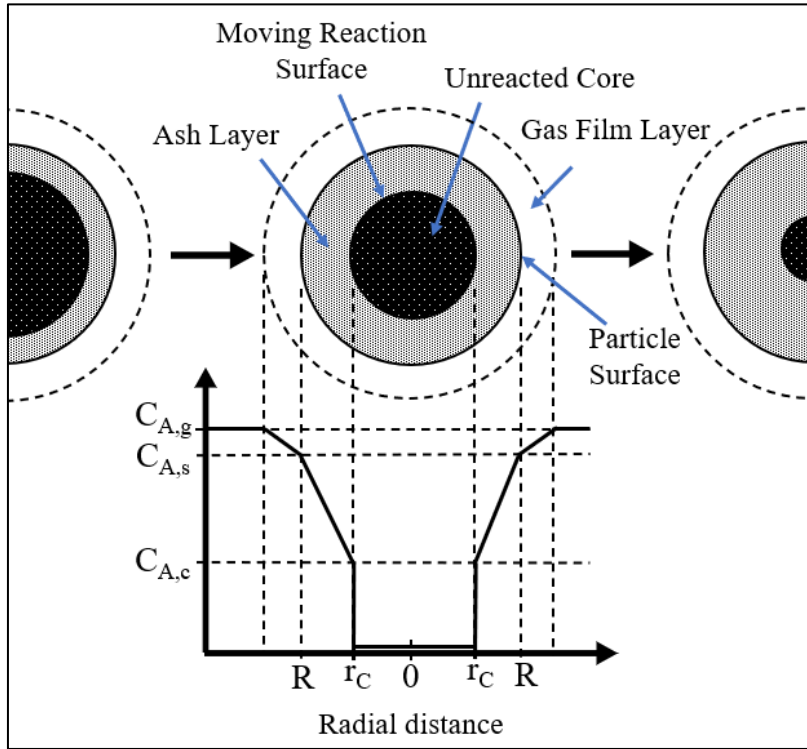


**Figure 2.10** Representation of shrinking-core model with the concentration values of typical heterogeneous conversion reaction  $A(g) + B(s) \rightarrow \text{Solid} + \text{gaseous products}$ , adapted from [99]

Shrinking-core method is implemented into CPFD model by defining the mass concentration of gaseous reactant on unreacted solid particle surface,  $X_c$ , as follows:

$$X_c = \frac{X_\infty}{k_R \left( \frac{1}{k_R} + \frac{1}{k_D} + \frac{1}{k_B} \right)} \quad (2.44)$$

where,  $X_\infty$  is the mass concentration of gaseous reactant surrounding the particle.  $k_R$  is the first order reaction rate for the core,  $k_D$  is the diffusion rate through the ash layer and  $k_B$  is the boundary layer mass transfer rate.

The diffusion rate of the gaseous reactant through the ash layer is defined as follows:

$$k_D = \frac{4\pi D_{m,s}}{\frac{1}{r_p} - \frac{1}{r_c}} \quad (2.45)$$

where,  $D_{m,s}$  is diffusion coefficient for solid phase,  $r_p$  is particle radius and  $r_c$  is unreacted core radius. Unreacted core radius can be calculated by using the following equation:

$$r_c = \left( \frac{m_c}{4\pi\rho_c/3} \right)^{1/3} \quad (2.46)$$

where,  $m_c$  and  $\rho_c$  are the mass and density of the solid reactant in the core, respectively.

Boundary layer gas mass transfer rate can be expressed by the following set of equations:

$$k_B = h_m A_p \quad (2.47)$$

$$h_m = \frac{Sh D_{m,g}}{l} \quad (2.48)$$

$$l = \min(2d_p; 0.1V_{cell}^{1/3}) \quad (2.49)$$

$$Sh = 2 + 0.6Re^{1/2} \quad (2.50)$$

$$Re = \frac{2r_p\rho_f|u_p - u_f|}{\mu_f} \quad (2.51)$$

$$D_{m,g} = \frac{\mu_f}{Sc\rho_f} \quad (2.52)$$

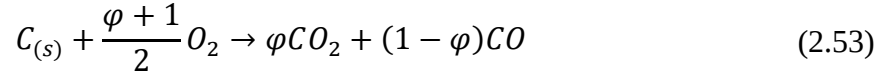
Here,  $h_m$  and  $D_{m,g}$  are the mass transfer coefficient and diffusion coefficient of the gas phase, respectively. Sh, Re and Sc are Sherwood, Reynolds and Schmidt numbers, respectively. Schmidt number can be practically taken as 0.9.

Since the physical properties of individual particles are used as input parameters of this model, Shrinking-core method can only be applied with discrete particle chemistry approach.

According to the Shrinking-core method, the ash layer limits the heat and mass transfer during gasification and combustion processes. But it is shown in the literature that this effect can be neglected for the chemical conversion of coal char particles [101]. Thus, VAC approach is chosen and Progressive-conversion model is used for the numerical analyses in this thesis study.

### Char Oxidation/Combustion:

Char-oxygen reactions are commonly combined and expressed as a unique reaction with a stoichiometric coefficient:



The stoichiometric coefficient  $\varphi$ , used in this reaction depends on the type of coal, temperature and particle diameter. It is stated in the literature that  $\varphi$  gets values very close to one for large coal particles, especially with size values higher than 1 mm [67]. Since the coal particle size is between 0.5-1.0 mm in the scope of this study, partial combustion is also taken into account during the numerical analysis setup.

Oxidation/combustion reactions have relatively faster chemical kinetic rates than gasification reactions. While combustion reactions are defined as exothermic processes, gasification reactions have endothermic behavior except heterogeneous methanation reaction. Methanation is the slowest reaction in a gasification process. Thus, heterogeneous methanation reaction is usually neglected for numerical simulation of coal gasification. But methanation is taken into consideration in the scope of this study.

### Kinetic Rates of Chemical Reactions:

Gasification process consists of many thermal and chemical steps as mentioned in Chapter 1. Although all the chemical reactions and their rates could be described, it is not possible to examine a large number of coupled reactions numerically for such a complex process with a realistic computation time. The general approach is to use a reduced number of reactions by considering the dominating reaction rates for the process. Chemical reactions are modeled in Arrhenius form where the chemical reaction rates can be defined as:

$$k = A_0 T^m \exp\left(-\frac{E}{T} - E_0\right) C_1^a C_2^b C_3^c \quad (2.54)$$

where  $A_0$  is the exponential factor,  $T$  [K] is the temperature,  $E$  [K] is the activation energy divided by the universal gas constant,  $C_1$  and  $C_2$  are molar concentrations, and  $a$  and  $b$

are the reaction orders in mol/m<sup>3</sup>.  $C_3$  is another term that can be mass ( $m_c$ ) or volume fraction ( $v_c$ ) of carbon for surface reactions or the mole fraction of some reactant for gas phase reactions. Table 2.7 and Table 2.8 show gas phase and solid surface reactions that are included in the numerical setup and the Arrhenius kinetic parameters of these reactions.

**Table 2.7** Homogenous reactions and kinetic parameters:

| Reaction                                      | $A_0$                  | $E$   | $E_0$ | $m$ | $a$  | $b$  | $c$ | $C_3$  | Ref  |
|-----------------------------------------------|------------------------|-------|-------|-----|------|------|-----|--------|------|
| $CO + H_2O \rightarrow CO_2 + H_2$            | $7.68 \times 10^{10}$  | 36640 | 0     | 0   | 0.5  | 1    | 0   |        | [69] |
| $CO_2 + H_2 \rightarrow CO + H_2O$            | $6.4 \times 10^9$      | 39260 | 0     | 0   | 1    | 0.5  | 0   |        | [70] |
| $CO + 0.5O_2 \rightarrow CO_2$                | $1.259 \times 10^{10}$ | 20130 | 0     | 0   | 1    | 0.25 | 0.5 | $H_2O$ | [66] |
| $CH_4 + 0.5O_2 \rightarrow CO + 2H_2$         | $2.474 \times 10^9$    | 15107 | 0     | 0   | 0.5  | 1.25 | 0   |        | [68] |
| $H_2 + 0.5O_2 \rightarrow H_2O$               | $3.824 \times 10^{13}$ | 20130 | 0     | -1  | 0.25 | 1.5  | 0   |        | [68] |
| $CH_4 + H_2O \rightarrow CO_2 + 3H_2$         | $3.0 \times 10^5$      | 15107 | 0     | 0   | 1    | 1    | 0   |        | [68] |
| $NH_3 + 0.75O_2 \rightarrow 0.5N_2 + 1.5H_2O$ | $5.07 \times 10^{14}$  | 35200 | 0     | 0   | 1    | 0.75 | 0   |        | [71] |

**Table 2.8** Heterogeneous reactions and kinetic parameters:

| Reaction                            | $A_0$                  | $E$   | $E_0$ | $m$ | $a$ | $b$ | $c$ | $C_3$ | Ref  |
|-------------------------------------|------------------------|-------|-------|-----|-----|-----|-----|-------|------|
| $C(s) + H_2O \rightarrow CO + H_2$  | $7.519 \times 10^{-3}$ | 15097 | 0     | 1   | 0   | 1   | 1   | $m_c$ | [67] |
| $CO + H_2 \rightarrow C(s) + H_2O$  | $6.174 \times 10^{-7}$ | 1233  | 17.29 | 2   | 1   | 1   | 1   | $m_c$ | [67] |
| $C(s) + CO_2 \rightarrow 2CO$       | $7.519 \times 10^{-3}$ | 15097 | 0     | 1   | 0   | 1   | 1   | $m_c$ | [67] |
| $2CO \rightarrow C(s) + CO_2$       | $6.174 \times 10^{-7}$ | 1233  | 20.92 | 2   | 2   | 0   | 1   | $m_c$ | [67] |
| $0.5C(s) + H_2 \rightarrow 0.5CH_4$ | $6.836 \times 10^{-3}$ | 8078  | 7.087 | 1   | 0   | 1   | 1   | $m_c$ | [67] |
| $0.5CH_4 \rightarrow 0.5C(s) + H_2$ | $7.545 \times 10^{-1}$ | 13128 | 0.372 | 0.5 | 0.5 | 0   | 1   | $m_c$ | [67] |
| $C(s) + O_2 \rightarrow CO_2$       | 12224                  | 13587 | 0     | 1   | 0   | 1   | 1   | $m_c$ | [72] |
| $2C(s) + O_2 \rightarrow 2CO$       | 12224                  | 13587 | 0     | 1   | 0   | 1   | 1   | $m_c$ | [72] |

While the temperature used in the chemical reaction calculations is assumed to be equal to the fluid temperature for volume average approach for homogeneous phase reactions, temperature can be calculated considering the effect of particle temperature by using a weighting factor,  $\omega$ , as follows:

$$T = \omega_f T_f + \omega_p T_p \quad (2.55)$$

where,  $\omega_f$  and  $\omega_p$  are weighting factor for fluid and particle, respectively. For this weighting method,  $\omega_f + \omega_p = 1$ . Thus, the effect of particle temperature increases when higher  $\omega_p$  value is chosen. Since  $\omega_p = 0$  for homogeneous reactions,  $\omega_f = 1$  and temperature value is equal to fluid temperature ( $T = T_f$ ).

#### 2.4.2. NO<sub>x</sub> Formation

Three main mechanisms for NO<sub>x</sub> formation are defined in the literature: Thermal NO<sub>x</sub>, Prompt NO<sub>x</sub> and Fuel NO<sub>x</sub>. Fuel NO<sub>x</sub> is formed by the reaction of nitrogen bound in the fuel with oxygen molecules in the air. While Fuel NO<sub>x</sub> formation is negligible for the combustion of gaseous fuels, liquid fuels involve a considerable amount of fuel-bound Nitrogen and ratio of fuel NO<sub>x</sub> may reach up to 50% of overall NO<sub>x</sub> emissions. Thermal NO<sub>x</sub> formation mechanism was first described in Zeldovich's study who stated that the oxygen and nitrogen molecules would react at high temperatures to form NO<sub>x</sub> in a combustion or gasification process. Zeldovich also remarked that the mechanisms forming NO<sub>x</sub> strongly depend on the temperature [77]. Zeldovich mechanism is defined by following three reactions:



By using these mechanisms, the net rate of change in NO concentration can be expressed as follows [103]:

$$\begin{aligned} \frac{d[NO]}{dt} = & k_1[O][N_2] + k_2[N][O_2] + k_3[N][OH] - k_{-1}[NO][N] \\ & - k_{-2}[NO][O] - k_{-3}[NO][H] \end{aligned} \quad (2.59)$$

Prompt NO<sub>x</sub> formation is proposed by Fenimore [78] and it was remarked that prompt NO<sub>x</sub> formation may reach a significant level quantity under fuel-rich and low-temperature conditions with short residence time. However, prompt NO<sub>x</sub> is relatively very small when it is compared to the overall amount of NO<sub>x</sub> generated from gasification or combustion

processes. The contribution from prompt NO is not large especially in coal-fired systems. Despite numerous possible reactions involving hydrocarbon fragments, two of them are believed to be prominent for prompt NO<sub>x</sub> formation [103]:



The cyanide *CN*, and hydrogen cyanide *HCN* is further oxidized and contributed to NO<sub>x</sub> formation.

The temperature dependency of these three mechanisms is shown in Figure 2.11 [79]. Moreover, the amount of nitrogen contained in the fuel is very small (<1%) compared to air and the fluidized bed temperature is between 800-900°C. Hence, NO<sub>x</sub> formation reactions are not taken into account in this study.

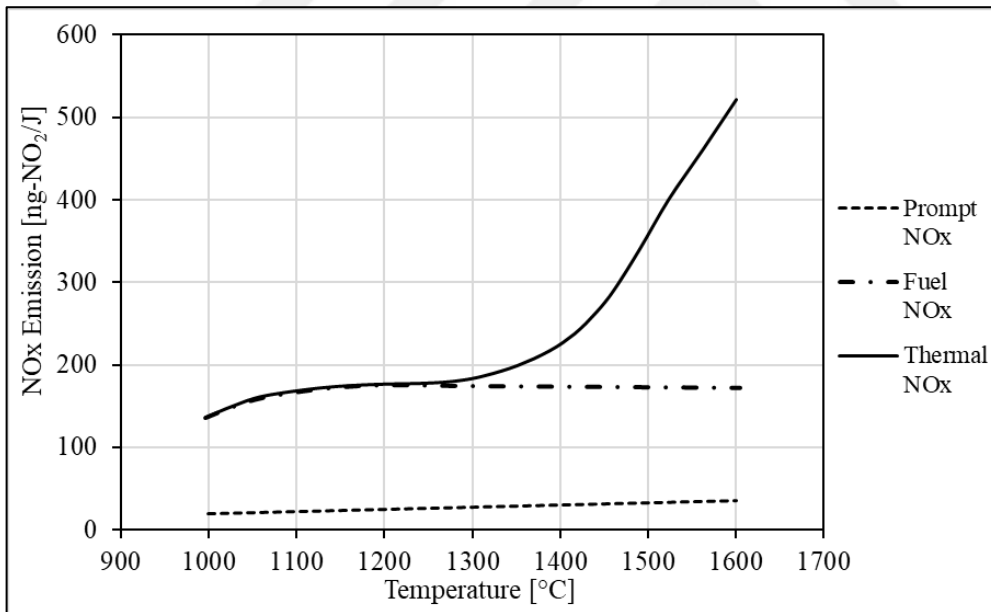


**Figure 2.11** Schematic representation of the distribution of NO<sub>x</sub> formation as a function of temperature, adapted from [79]

### 2.4.3. Sulfur Conversion

The conversion of sulfur during the combustion or gasification of coal is still a hot topic for researchers. Various mechanisms have been presented in recent studies involving

various experimental and numerical techniques [80]-[85]. Sulfur conversion is a very complex chemical process including numerous different steps. Leeds University suggested a sulfur mechanism involves 102-main elementary reactions [82]. Li et. al. reduced this mechanism into a ten-step form by using dominant reactions and simulated the sulfur conversion during a steady-state pulverized coal combustion process [84]. Ten-step reduced mechanism is given in the following reactions:



However, this mechanism still requires a very high computational cost for modeling sulfur conversion in the transient gasification process of coal particles.

Hasse et al. remarked that sulfur is mostly released in  $SO_2$  form with minor amounts of COS during the combustion process. In their review study about advances in CFD simulation of coal combustion and gasification, it is stated that  $H_2S$  is the main released product that involves sulfur content [90].

In the scope of this thesis study, the conversion of sulfur content for chosen coal samples is assumed to be in the volatile release process governed by the devolatilization models described in the next sub-section. While MGAS model converts all sulfur content directly into  $H_2S$  form, CPD model first converts it into tar form during the devolatilization process. Then, by oxidation of tar, it is converted into  $SO_3$  form at the end of the process in CPD model.

#### 2.4.4. Devolatilization Models

Many approaches have been suggested in the literature in order to model devolatilization mechanisms. Pyrolysis models are mainly classified into two groups: Empirical models & Network models. A detailed classification of these models is given in Figure 2.12.

Empirical pyrolysis models use a small number of simple reaction rates to simulate the pyrolysis process mathematically. While some empirical models are based on single or multi-step global kinetic approach and some of them use distributed activation energy (DAE) approach [86]. As one of the primary global kinetic approaches, Gregory and Littlejohn proposed an empirical correlation as a function of volatile matter content and temperature in order to estimate the devolatilization rate [87]. Loison and Chauvin presented a set of second-order polynomial equations based on only volatile matter content to determine the composition of the devolatilization products in their study [88]. Gungor and Eskin developed a dynamic 2D model for CFB combustors by using these correlations and validated the model with experimental data obtained from a pilot-scale (50kWth) and industrial scale (160MWth) CFB reactors [89].

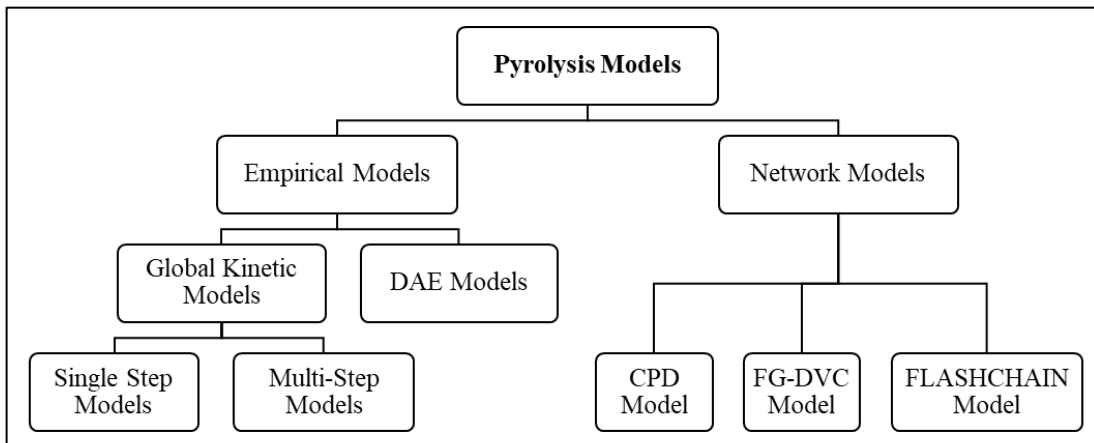


**Figure 2.12** Classification of pyrolysis models, adapted from [90]

MGAS, CPD, PCCL, FLASHCHAIN and FG-DVC are the most known advanced tools for modeling the devolatilization process of coal. Network devolatilization models such as PC- Coal Lab (PCCL) and FG-DVC (Functional Group, Depolymerization, Vaporization, Cross-linking) involve inherent complexity and it is challenging to implement the results into a CFD code [91]. However, outputs of MGAS and CPD models

can be easily applied to a CFD code. Also, they are applicable to a very wide range of coal types. Hence, MGAS and CPD devolatilization models are applied to numerical analyses in this study in order to predict the volatile matter content of coal particles by using C3M code.

The mass of all elements determined by ultimate analysis is carefully conserved within C3M. However, the amount of fixed carbon and volatile matter defined in the proximate analysis of the coal can shift depending on the specific pyrolysis conditions due to the applied heating profile during proximate analysis according to ASTM method. Thus, the proximate analysis is adjusted by considering this change in C3M.

The fixed carbon content,  $FC_m$ , is reported by a sub-model used for devolatilization modeling with both MGAS and CPD models. Once this value is calculated, an adjusted proximate analysis is determined and the composition of the volatile matter and tar can be obtained. Fixed carbon content is assumed to be pure carbon and the sum of the adjusted value of fixed carbon, adjusted value of volatile matter, moisture and ash is equal to 1. Hence, first mass balance equation can be written for proximate analysis as follows:

$$FC_m + VM_m = 1 \quad (2.72)$$

where,  $FC_m$  is fixed carbon and  $VM_m$  is volatile matter content of the fuel on dry ash free (DAF) basis. Other elements in the char such as oxygen, nitrogen, hydrogen, and sulfur may be calculated by a sub-model. However, these elements are assumed to be included in the volatile matter and/or tar during pyrolysis because char is pure carbon in C3M reactions.

While  $FC_m$  is obtained internally in CPD model, MGAS model calculates it by the following equation:

$$FC_m = \frac{FC_{PA}}{(1 - M_{PA} - A_{PA})} \quad (2.73)$$

Once the  $FC_m$  is obtained, the proximate analysis is adjusted by following equations:

$$FC_{PA\_adj} = FC_m(1 - M_{PA} - A_{PA}) \quad (2.74)$$

$$VM_{adj} = 1 - FC_{PA_{adj}} - M_{PA} - A_{PA} \quad (2.75)$$

The ultimate analysis results are defined on a DAF basis. If they are not defined in that way, they can be converted by using the following equation for any element,  $X$ :

$$VM_x = \frac{X_{UA} - FC_{PA}}{(1 - FC_{PA})} \quad (2.76)$$

where,  $VM_x$  is the mass fraction of any element in the volatile matter and  $X_{UA}$  is the fraction in the ultimate analysis.

Both MGAS and CPD models take the moisture release into account. Moisture release sub-model is defined by an Arrhenius form equation as follows:

$$r_{mr} = \frac{k_d \exp\left(-\frac{E_{a,d}}{RT_s}\right) \varepsilon_s \rho_s X_{H_2O}}{MW_{H_2O}} \quad (2.77)$$

where,  $k_d$  and  $E_{a,d}$  are the kinetic parameters of the moisture release sub-model,  $R$  is the universal gas constant.  $T_s$ ,  $\varepsilon_s$  and  $\rho_s$  are local temperature, local volume fraction and local density of solid particles, respectively.  $X_{H_2O}$  and  $MW_{H_2O}$  are local mass fraction and molecular weight of water in the coal, respectively.

Theoretical approaches, assumptions and some details of MGAS and CPD models are explained below:

### **MGAS Model**

The first devolatilization model considered in this study is a numerical model called METC-Gasifier Advanced Simulation (MGAS), presented by Syamlal and Bisset for the analysis and design of various types of gasification reactors [92]. The MGAS model can simulate a gasifier with given specifications and estimate its gasification performance. MGAS model provides a methodology to determine the products of volatile cracking based on proximate and ultimate analyses of the feedstock. The rate of moisture release from the coal is also calculated by using a sub-model with the mechanisms for tar cracking and tar combustion. This model is capable to obtain the syngas composition and calculate

mass flow rates of gasification agents required to achieve a desired carbon conversion level. Thus, MGAS model is practical for design studies.

MGAS uses a set of balance equations for mass and energy in differential form in order to determine the carbon conversion rate, maximum temperature and the transient behavior of the bed. MGAS model can predict outputs of both four sub-mechanisms (drying, pyrolysis, combustion and reduction) of the gasification process. But only one-dimensional pyrolysis sub-model of MGAS tool is used within the frame of this study.

MGAS model has an empirical kinetic approach that enables the model to obtain the tar yield and to obtain the composition of syngas. MGAS tracks the flow and solid particle temperatures separately with two energy equations to accurately calculate the temperature changes during large particle gasification and link the devolatilization kinetics with coal heat-up.

The product gas composition and temperature predicted by the model were compared with data from a METC gasifier depicted in Figure 2.13. Experimental and predicted tar yields were compared to adjust the rate of the tar cracking reaction. Also, experimental and calculated temperature data were compared to adjust the partitioning of the heat of the carbon combustion. The model validation was satisfied with a good agreement between numerical and experimental data.

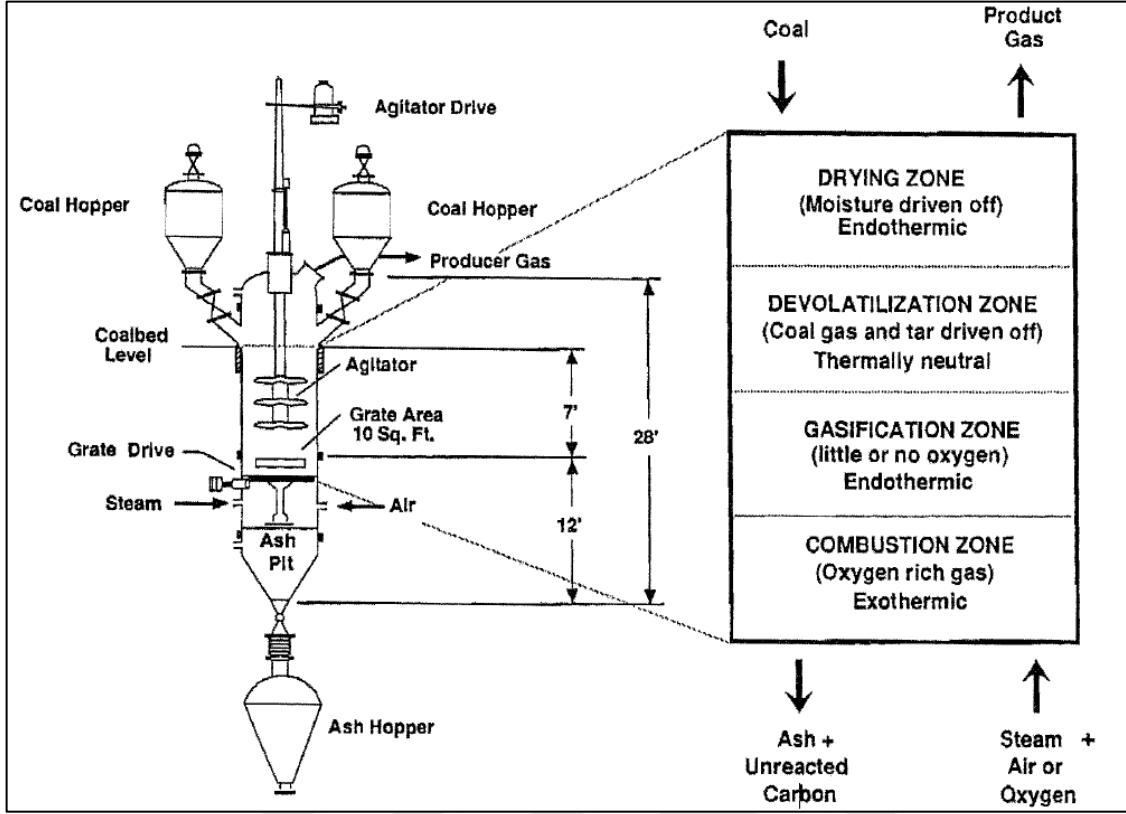
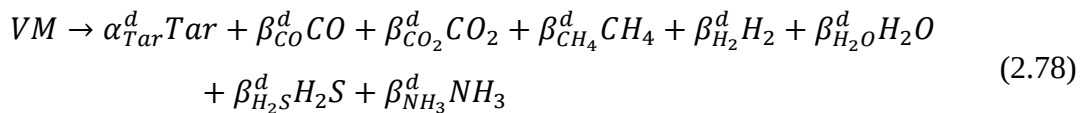


**Figure 2.13** MTEC Gasifier [92]

MGAS model can track fourteen gas species: CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>, H<sub>2</sub>O, H<sub>2</sub>S, N<sub>2</sub>, O<sub>2</sub>, NH<sub>3</sub>, Tar, C<sub>2</sub>H<sub>4</sub>, C<sub>2</sub>H<sub>6</sub>, C<sub>3</sub>H<sub>8</sub> and C<sub>6</sub>H<sub>6</sub>. Only CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub>, H<sub>2</sub>O, H<sub>2</sub>S, N<sub>2</sub>, O<sub>2</sub>, NH<sub>3</sub> and tar species are taken into account in this study. MGAS model involves three stages for the pyrolysis process: Devolatilization, tar cracking and tar oxidation.

The elemental composition of the tar is obtained using the devolatilization reaction with  $\alpha$  and  $\beta$  coefficients in the model. The devolatilization approach of MGAS model can be expressed as following reaction in mass terms:



It is assumed that unit mass of volatile matter breaks up into the mass fractions from the  $\alpha^d$  and  $\beta^d$ 's. Thus, the sum of  $\alpha^d$  and all  $\beta^d$  coefficients is equal to one. These coefficients for each component except Hydrogen, is defined by using DAF basis mass fractions of solid ( $f_s$ ) and related volatile component ( $f_g$ ) as follows:

$$\beta_g^d = \frac{f_g}{(1 - f_s)} \quad (2.79)$$

$$\alpha_{Tar}^d = \frac{f_{tar} + f_{other}}{(1 - f_s)} \quad (2.80)$$

The elemental mass fractions of tar components in MGAS devolatilization model can be expressed as follows:

$$Tar_C = \frac{VM_C - (\beta_{CO}^d \delta_{CO}^C + \beta_{CO_2}^d \delta_{CO_2}^C + \beta_{CH_4}^d \delta_{CH_4}^C)}{\alpha d_{tar}} \quad (2.81)$$

$$Tar_H = \frac{VM_H - (\beta_{CH_4}^d \delta_{CH_4}^H + \beta_{H_2}^d \delta_{H_2}^H + \beta_{H_2O}^d \delta_{H_2O}^H + \beta_{H_2S}^d \delta_{H_2S}^H + \beta_{NH_3}^d \delta_{NH_3}^H)}{\alpha d_{tar}} \quad (2.82)$$

$$Tar_O = \frac{VM_O - (\beta_{CO}^d \delta_{CO}^O + \beta_{CO_2}^d \delta_{CO_2}^O + \beta_{H_2O}^d \delta_{H_2O}^O)}{\alpha d_{tar}} \quad (2.83)$$

$$Tar_N = \frac{VM_N - \beta_{NH_3}^d \delta_{NH_3}^H}{\alpha d_{tar}} \quad (2.84)$$

$$Tar_S = \frac{VM_S - \beta_{H_2S}^d \delta_{H_2S}^H}{\alpha d_{tar}} \quad (2.85)$$

where,  $\delta_Y^g$  is the mass ratio of element  $g$  in the molecule. However, as an assumption of MGAS model, it is always predicted for tar content to have an ultimate analysis of 0.9 C, 0.08 H, and 0.02 O. Moreover, it is assumed that any sulfur in the coal used for the formation of  $H_2S$  and all Nitrogen ends up as  $NH_3$ . The remaining species distribution is set to maintain elemental mass balance.

Molar fraction conversion from mass units can be obtained by using the following equation for any species,  $g$ :

$$n_X = \frac{\beta_g^d MW_{VM}}{MW_g} \quad (2.86)$$

The chemistry and reaction kinetics called Two Parameter - Single Stage Model (2P1SM) is used in MGAS based on the results presented by Wen et al.[67]. 2P1SM is a modified Arrhenius form with a switch based on reaction temperature that accounts for the fraction

of volatiles that will remain in the coal if the solids are not heated high enough to complete the devolatilization. 2P1SM devolatilization rate,  $r_d$ , can be expressed as follows:

$$r_d = \begin{cases} \frac{A \exp\left(-\frac{E}{RT_s}\right) \varepsilon_s \rho_s (X_{VM} - X^*)}{MW_{VM}} & X_{VM} \geq X^* \\ 0 & X_{VM} < X^* \end{cases} \quad (2.87)$$

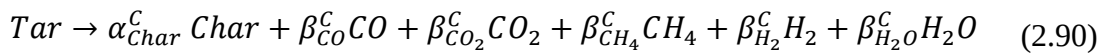
where,

$$X^* = (X_{VM}^0 + X_{FC}^0) X_{min}^* \quad (2.88)$$

$$X_{min}^* = \begin{cases} \frac{\left(\frac{867.2}{T_s - 273}\right)^{3.914}}{100} & T_s < 1223 \\ 0 & T_s \geq 1223 \end{cases} \quad (2.89)$$

Here,  $A$  is the pre-exponential factor,  $E$  is the activation energy in cal/mol,  $R$  is universal gas constant in cal/mol/K,  $T_s$  is the solid particle temperature in K,  $\varepsilon_s$  is the local volume fraction of the solids and  $\rho_s$  is the density of solid in g/cm<sup>3</sup>.  $X$  and  $MW$  are the mass fraction and molecular weight values, respectively. Superscript  $*$  denotes the temperature switch condition.

The tar cracking reaction in MGAS is defined to produce a mixture of char, CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub> and H<sub>2</sub>O. The tar cracking reaction and its rate,  $r_{tc}$ , are defined in the following equations:

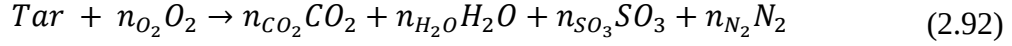


$$r_{tc} = \frac{A \exp\left(-\frac{E_a}{RT_s}\right) \varepsilon_g \rho_g X_{Tar}}{MW_{Tar}} \quad (2.91)$$

where,  $\varepsilon_g$  is local volume fraction,  $\rho_g$  is local density.  $X_{Tar}$  and  $MW_{Tar}$  are local mass fraction and molecular weight of the tar, respectively.

MGAS model also calculates the molecular weight of tar component. However, the molecular weight of volatile matter is assumed to be 56 g/mol.

The tar produced as a result of the pyrolysis process is oxidized with available oxygen in the reactor. Molar balance for tar oxidation reaction is given in the following equation:



where,  $n_g$  is the stoichiometric coefficient of any species,  $g$ . All nitrogen content of the tar is assumed to be converted into  $N_2$  due to low temperature characteristics of the pyrolysis process in a gasifier reactor. The stoichiometric coefficients can be obtained for each species by using the following equations:

$$n_{CO_2} = \frac{MW_{Tar} Tar_C}{AW_C} \quad (2.93)$$

$$n_{H_2O} = \frac{MW_{Tar} Tar_H}{2AW_H} \quad (2.94)$$

$$n_{SO_3} = \frac{MW_{Tar} Tar_S}{AW_S} \quad (2.95)$$

$$n_{N_2} = \frac{MW_{Tar} Tar_N}{2AW_N} \quad (2.96)$$

$$n_{O_2} = \frac{2n_{CO_2} + n_{H_2O} + 3n_{SO_3} - \frac{MW_{Tar} Tar_O}{AW_O}}{2} \quad (2.97)$$

The reaction rate of the tar oxidation in MGAS model,  $r_{TO}$ , is assumed to be the rate for  $C_{10}H_{22}$  proposed by Westbrook and Dryer [66] expressed as follows:

$$r_{TO} = 3.8 \times 10^{11} \exp\left(-\frac{30000}{RT_g}\right) \varepsilon_g \left(\frac{\rho_g X_{O_2}}{MW_{O_2}}\right)^{1.5} \left(\frac{\rho_g X_{Tar}}{MW_{Tar}}\right)^{0.25} \quad (2.98)$$

where,  $\varepsilon_g$  is local volume fraction,  $\rho_g$  is local density and  $X$  is local mass fraction value of the species.

MGAS model allows to use 5 different coal types as kinetic rate constants as listed in the table below for modelling devolatilization (Table 2.9). Since the proximate analysis of the coal samples used in this study is close to North Dakota lignite, its kinetic data is used

for devolatilization modeling. Additionally, kinetic rates of char oxidation and Boudouard reactions are calculated by referencing this assumption.

**Table 2.9** Kinetic rate constants used by MGAS Model, adapted from [67]

| Reaction              | Const.   | Unit    | Arkwright<br>Pittsburgh<br>(Bitum.) | Pittsburgh<br>No.8<br>(Bitum.) | Illinois<br>No.6<br>(Bitum.) | Rosebud<br>(Sub-bitum.) | North<br>Dakota<br>(Lignite) |
|-----------------------|----------|---------|-------------------------------------|--------------------------------|------------------------------|-------------------------|------------------------------|
| Char<br>Oxidation     | <i>A</i> | 1/atm.s | 600                                 | 930                            | 3000                         | 70                      | 1.1                          |
|                       | <i>E</i> | K       | 22646                               | 22646                          | 21136                        | 15097                   | 15097                        |
| Boudouard             | <i>A</i> | 1/atm.s | 600                                 | 930                            | 3000                         | 70                      | 1.1                          |
|                       | <i>E</i> | K       | 22646                               | 22646                          | 21136                        | 15097                   | 15097                        |
| Devolati-<br>lization | <i>A</i> | 1/atm.s | $1.1 \times 10^5$                   | $1.1 \times 10^5$              | $1.1 \times 10^5$            | $5.1 \times 10^4$       | $5.1 \times 10^4$            |
|                       | <i>E</i> | K       | 10669                               | 10669                          | 10669                        | 9411                    | 8153                         |
| Tar<br>Cracking       | <i>A</i> | 1/atm.s | $2.5 \times 10^7$                   | $9.7 \times 10^9$              | $9.7 \times 10^9$            | $8.0 \times 10^{10}$    | $2.1 \times 10^8$            |
|                       | <i>E</i> | K       | 14594                               | 14594                          | 14594                        | 13965                   | 13336                        |

### CPD Model

In the second pyrolysis model used in the scope of this study, an empirical approach to modeling pyrolysis is taken into account for the numerical analysis due to the complexity of the coal structure and its chemistry. Chemical Percolation model for Devolatilization (CPD) proposed by Grant et al. [73] and Fletcher et al. [74], uses measured chemical structure coefficients from coal and Carbon-13 Nuclear Magnetic Resonance ( $^{13}\text{C}$  NMR) analysis to predict pyrolysis behavior. The CPD model includes the principal thermochemical processes involved in coal pyrolysis. Figure 2.14 illustrates the main concept used to quantify the coal structure in CPD model.

The CPD model includes following features [93]:

- The chemical structure description of the parent coal
- Kinetic scheme for breaking labile bridges ( $\xi^*$ ) and side chains ( $\delta$ )
- Percolation approach based on a Bethe lattice to detach molecules from the coal particle
- A vapor–liquid equilibrium treatment between metaplast and tar content
- A cross-linking mechanism for reattaching metaplast to the solid char.

According to the CPD model, coal structure is defined as a macromolecular array consisting of a single aromatic ring or aromatic ring clusters with various types and sizes. These clusters are connected to each other by chemical bridges of different bond strengths. Some of these bonds are labile bridges that can break readily during coal pyrolysis, whereas others are stable at a certain operating temperature.

A labile bridge decomposes to form a reactive bridge intermediate, which is unstable and quickly reactive in one of two competitive reactions. The reactive intermediate bridge is cleaved and two halves form side chains that remain attached to the aromatic cluster for the first mechanism. Tar fragments which consist of such finite clusters, are generated as a result of the integration of a sufficient number of bridges completely detached from the lattice network. The side chains expose a cracking reaction to form light gas during the pyrolysis process. Side chains are branching element of a molecule that is attached to a larger hydrocarbon backbone. If a side chain is connected to the same aromatic cluster (monomer) at multiple points, it is known as a loop. Other bridges in given thermochemical process conditions are defined as char bridges (*c*). For the competing reaction mechanism, the reactive intermediate is stabilized and a relatively stable char bridge is formed with the consequent release of light gas [9].

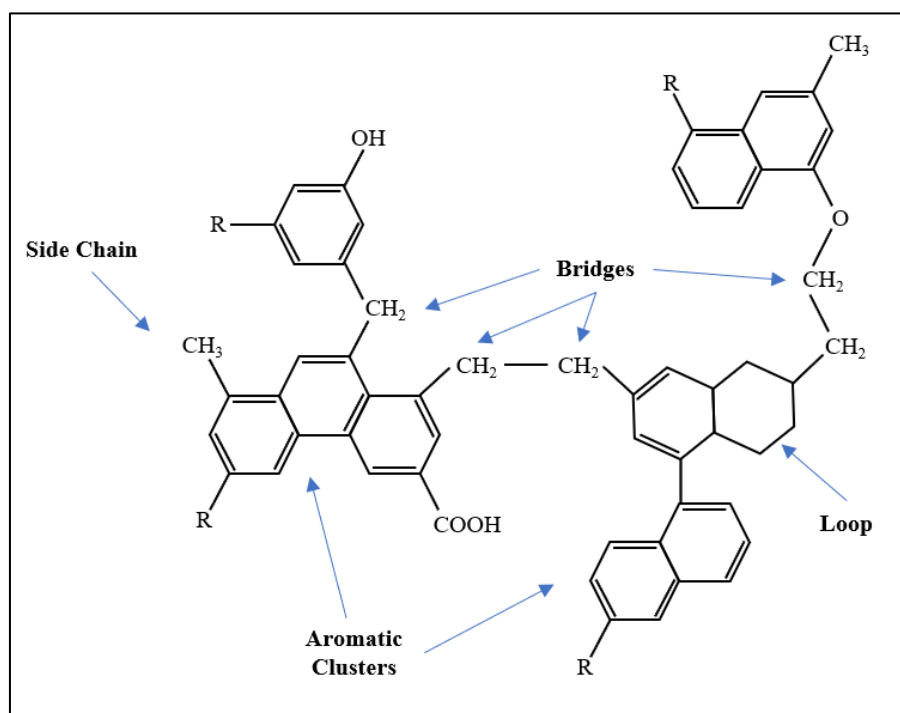


**Figure 2.14** Schematic representation of chemical structures identified in  $^{13}\text{C}$  NMR analyses and used in the CPD model, adapted from [74]

During the pyrolysis process, solid coal particles typically is converted into a semi-liquid phase; which decomposes to solid char and volatiles. The liquid fraction of this phase is called a metaplast. The metaplast is exposed to two competing reactions. While the first reaction causes splitting of large fragments into two smaller fragments, char and two smaller fragments are formed in the second reaction [9]. However, the amount of generated metaplast during pyrolysis of lignite is very small, hence its effect is limited [94].

The ultimate tar output, the molecular weight of tar and char's behavior are determined by cross-linking reactions which are essential in a typical pyrolysis process. Cross-linking does not generate any new attachment sites and occurs only at existing sites that are initially occupied by a side chain. There are various types of side chains in coal, and the cross-linking reactions of different side chains are different. However, the models to describe cross-linking reactions are usually simplified. The amount of metaplast recombined with coal matrix was calculated by a first-order reaction and a second-order reaction, respectively. The metaplast cross-linking mechanism is decoupled from the percolation statistics, there is not any difference between the cross-linking behavior of bituminous coals and lignite [94].

It is not possible to define the statistics of real 2D and 3D arrays analytically due to the looped structure of sites and bridges within the lattice. Nevertheless, Bethe lattices that involve a class of pseudo-lattices based on percolation theory, can be solved analytically. These Bethe lattices are similar to real lattices with respect to the possible characterization by a coordination number and a bridge population parameter. However, contrary to real lattices, any sites in a Bethe lattice are only connected to each other by a single path, so the structure doesn't involve any loop. Bethe lattice approach is illustrated in Figure 2.15.

Chemical reaction rate constants are calculated using the structural coefficients of the CPD model, which can be measured using  $^{13}\text{C}$  NMR method, to determine the output of the pyrolysis process without considering the coal type in the CPD model has been successfully applied to a wide range of coal resources with chemical variability and structural complexity. Since  $^{13}\text{C}$  NMR data for all coal types are not available, correlations have been developed in order to determine the CPD model parameters [95].

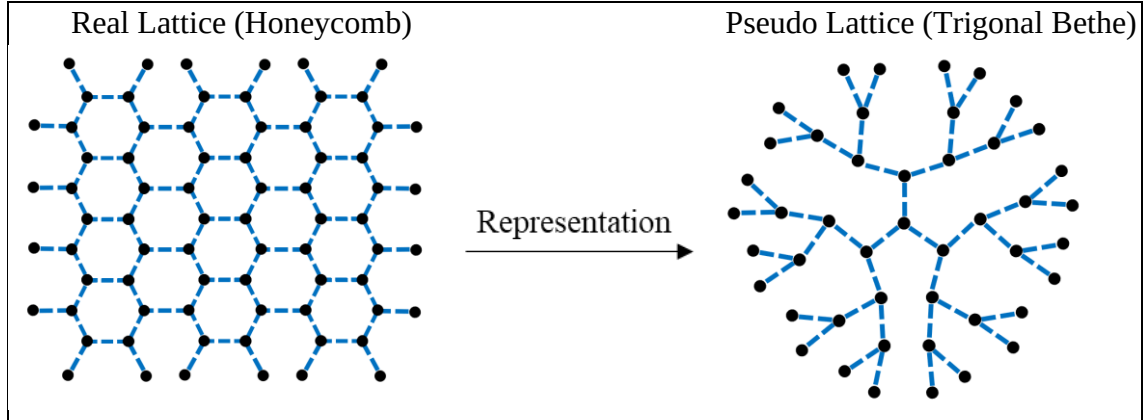


**Figure 2.15** Schematic illustration of Bethe lattice approach to represent real lattice structure in CPD model, adapted from [90]

The reaction mechanism of CPD model is illustrated in Figure 2.16. The formation and kinetic rates for the individual species in CPD reaction mechanism are given in equations (2.99) - (2.105) [74][96].

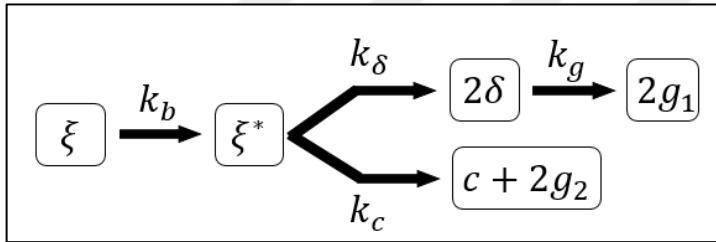


**Figure 2.16** CPD reaction mechanism, adapted from [74].

The conversion rate of a labile bridge  $\xi$  into a relatively smaller reactive unstable bridge intermediate  $\xi^*$ , is defined by the following reaction:

$$\frac{d\xi}{dt} = -k_b \xi \quad (2.99)$$

The change in  $\xi^*$  can be defined as the sum of the formation and consumption reactions rate of this species as follows:

$$\frac{d\xi^*}{dt} = k_b \xi - (k_\delta + k_c) \xi^* \quad (2.100)$$

Other reaction rates in CPD model are defined by assuming that the transient consumption and formation rate of reactive bridges are equal (i.e.  $\partial \xi^* / \partial t = 0$ ). By this assumption,

the reaction rates for the production of char bridges and side chains can be simplified with eliminating  $k_\delta$  variable as follows:

$$\frac{dc}{dt} = -k_b \frac{\xi}{(k_c + 1)} \quad (2.101)$$

$$\frac{d\delta}{dt} = \left[ 2k_c k_b \frac{\xi}{(k_c + 1)} \right] - k_g \delta \quad (2.102)$$

Production rates of light gases obtained from side chains ( $g_1$ ) and char bridges ( $g_2$ ) are defined in the following equations:

$$\frac{dg_1}{dt} = k_g \delta \quad (2.103)$$

$$\frac{dg_2}{dt} = 2 \frac{dc}{dt} \quad (2.104)$$

A simple cross-linking mechanism is used to determine the reattachment rate of metaplast to the infinite matrix. This mechanism is described in the model by a one-step Arrhenius expression as follows:

$$\frac{dm_{cross}}{dt} = k_{cross} m_{meta} \quad (2.105)$$

where,  $m_{meta}$  is the mass of metaplast (tar precursor fragments),  $m_{cross}$  is the amount of reattached metaplast.

$\delta$ ,  $\xi$ , and  $c$ , are dynamic bridge population variables from which the number of intact bridges,  $p$ , is calculated as  $p = \xi + c$ . The fraction of broken bridges  $f$ , is also defined as  $f = 1 - p$ . Conservation of mass dictates that  $g_1 = 2f - \delta$  and  $g_2 = 2(c_0 - c)$  so that only three differential equations [(2.99),(2.101)&(2.102)] are solved for bridge parameters. According to the model, the dynamic variables are matched with the variables of percolation theory, the number of intact bridges, and the lattice coordination number,  $\sigma + 1$  [73].  $c_0$ ,  $\xi_0$ ,  $\delta(0) = 2f_0$  and  $g_1(0) = g_2(0) = 0$  are the initial conditions for the solution of these three equations.

$k_b$ ,  $k_g$ ,  $k_c$ ,  $k_\delta$ , and  $k_{cross}$  are kinetic rate constants of corresponding reactions expressed above, in Arrhenius form ( $k = \exp(-E/RT)$ ). Kinetic rate relations, frequency factor and activation energy values for each rate are given in Table 2.10.

**Table 2.10** Kinetic rate parameters in reaction mechanism of CPD Model [96]

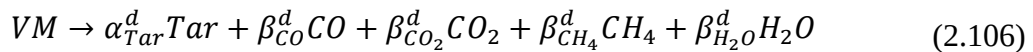
| Reaction                                     | Parameter                   | Symbol         | Value                | Unit            |
|----------------------------------------------|-----------------------------|----------------|----------------------|-----------------|
| Bridge Breaking, $k_b$                       | Activation energy           | $E_b$          | 232                  | $\text{kJ/mol}$ |
|                                              | Frequency factor            | $A_b$          | $2.6 \times 10^{15}$ | $1/\text{s}$    |
|                                              | Standard deviation of $E_b$ | $\sigma_b$     | 7.5                  | $\text{kJ/mol}$ |
| Light Gas Release, $k_g$                     | Activation energy           | $E_g$          | 232                  | $\text{kJ/mol}$ |
|                                              | Frequency factor            | $A_b$          | $2.6 \times 10^{15}$ | $1/\text{s}$    |
|                                              | Standard deviation of $E_b$ | $\sigma_b$     | 7.5                  | $\text{kJ/mol}$ |
| Char Bridge, $k_c$<br>Side Chain, $k_\delta$ | Composite rate constant     | $k_\delta/k_c$ | 0.9                  | —               |
| Cross-link, $k_{cross}$                      | Activation energy           | $E_{cross}$    | 272                  | $\text{kJ/mol}$ |
|                                              | Frequency factor            | $A_{cross}$    | $3.0 \times 10^{15}$ | $1/\text{s}$    |

CPD model uses five coal-specific parameters to solve the set of equations and corresponding kinetic rate constants [96]:

- lattice coordination number,  $\sigma + 1$
- initial number of bridges in the coal lattice,  $p_0 = c_0 + \xi_0$
- mass of the coal in the clusters,  $m_{cluster}$
- mass of coal in the bridges  $m_b$
- initial number of char bridges  $c_0$

The first four quantities are the structural parameters of coal type that can be obtained from NMR experiments. The last one is estimated from the initial char content of the coal. Smith et al. published the values of these parameters for nineteen different coals with various ranks and other parameters independent from coal type in the CPD model [96].

The elemental composition of the tar is obtained using the devolatilization reaction with  $\alpha^d$  and  $\beta^d$  coefficients in the model. CPD model can be expressed as the following reaction in mass terms:



Here, the definition of  $\alpha^d$  and  $\beta^d$  coefficients are same as MGAS model.

CPD model computes the mass fractions of solids, H<sub>2</sub>O, CO<sub>2</sub>, CH<sub>4</sub>, CO, Tar, and “other”. The mass fraction of “other” is the combined fractions of light hydrocarbons not heavy enough to be considered tar. CPD does not explicitly predict Hydrogen, but rolls it up in the “other” fraction with higher hydrocarbons. Hence, it is not possible to directly obtain the mass fraction of Hydrogen.

The elemental mass fractions of tar components are defined as follows (equivalent to an ultimate analysis):

$$Tar_C = \frac{VM_C - (\beta_{CO}^d \delta_{CO}^C + \beta_{CO_2}^d \delta_{CO_2}^C + \beta_{CH_4}^d \delta_{CH_4}^C)}{\alpha_{tar}^d} \quad (2.107)$$

$$Tar_H = \frac{VM_H - (\beta_{CH_4}^d \delta_{CH_4}^H + \beta_{H_2O}^d \delta_{H_2O}^H)}{\alpha_{tar}^d} \quad (2.108)$$

$$Tar_O = \frac{VM_O - (\beta_{CO}^d \delta_{CO}^O + \beta_{CO_2}^d \delta_{CO_2}^O + \beta_{H_2O}^d \delta_{H_2O}^O)}{\alpha_{tar}^d} \quad (2.109)$$

$$Tar_N = \frac{VM_N}{\alpha_{tar}^d} \quad (2.110)$$

$$Tar_S = \frac{VM_S}{\alpha_{tar}^d} \quad (2.111)$$

where,  $\delta_Y^g$  is the mass ratio of element  $g$  in molecule.

The  $n^{\text{th}}$  order Arrhenius model is used in CPD model with coefficients based on experimental data determined by TGA. The devolatilization rate,  $r_d$ , is expressed in g/cm<sup>3</sup>/s as follows:

$$r_d = \frac{A \exp\left(-\frac{E}{RT_s}\right) (1 - \zeta)^n \varepsilon_s \rho_s X_{VM}^0}{MW_{VM}} \quad (2.112)$$

where,  $A$  is the pre-exponential factor,  $E$  is the activation energy in cal/mol,  $R$  is universal gas constant in cal/mol/K,  $T_s$  is the solid particle temperature in K,  $\varepsilon_s$  is the local volume fraction of the solids and  $\rho_s$  is the density of solid in g/cm<sup>3</sup>.  $X_{VM}^0$  and  $MW_{VM}$  are the mass fraction and molecular weight of volatile matter, respectively. The model parameters  $A$ ,  $E_a$  and  $n$  are determined through a least squared approach fits to TGA data.  $\zeta$  variable refers to the conversion of volatile matter. The fitting process is conducted numerically

and aim to minimize the numerical integral of the devolatilization rate equation for each particle.

Tar weight assumption and tar oxidation methodology of CPD model are also same as MGAS model.



### 3. EXPERIMENTAL AND NUMERICAL ANALYSIS OF LAB-SCALE SYSTEM

#### 3.1. Experimental Results

Gasification experiments of four different coal samples are conducted by using an atmospheric bubbling fluidized bed gasifier described in Section 2.2. Experimental results are given in Table 3.1, Table 3.2, Table 3.3 and Table 3.4 for Sample-1, Sample-2, Sample-3 and Sample-4, respectively. Mass flow rate of coal, volumetric flow rate of air and volumetric flow rate of nitrogen feed with coal are given as measured input parameters of these experiments. Air need for stoichiometric combustion is calculated by using equation (2.2) and ER values are obtained by using equation (2.1).

According to the experimental data, it can be said that lower reactor temperature ( $T_{1-2}$ ) and upper reactor temperature ( $T_{1-2}$ ) values remained constant and close to the chosen operating temperature (825°C) for different ER values.

As mentioned in Section 2.2, only CO, CO<sub>2</sub>, CH<sub>4</sub>, H<sub>2</sub> and O<sub>2</sub> species are measured. Nevertheless, syngas involves various unmeasured gas molecules such as, N<sub>2</sub>, NO, H<sub>2</sub>S, COS, etc. Despite that, it can be assumed that the major amount of unmeasured content is composed of N<sub>2</sub>. The volumetric ratio of unmeasured content of syngas is obtained by subtracting the sum of the measured species from 100%. This content is called “Other” species in both experimental and numerical results.

It can be easily seen that the volumetric ratio of other species increases with increasing ER values. Since air is mostly composed of N<sub>2</sub>, the volumetric ratio of useful and combustible species (such as CO, CH<sub>4</sub> and H<sub>2</sub>) in syngas composition decreases by increasing air flow rate. Relatively high CH<sub>4</sub> and H<sub>2</sub> concentrations are obtained for ER3, ER4 and ER5 values in the syngas obtained from Sample-1 & Sample-2. A similar trend is seen in the gasification experiments of Sample-3 & Sample-4 for ER4 and ER5 values. While higher CO<sub>2</sub> concentration values are measured with increasing air flow rates for Sample-3 & Sample-4, it is almost exact opposite for Sample-1 & Sample-2.

**Table 3.1** Lab-scale experimental results - Gasification of coal Sample-1.

| Parameter          | Unit               | ER1    | ER2    | ER3    | ER4    | ER5    |
|--------------------|--------------------|--------|--------|--------|--------|--------|
| Coal MFR           | kg/h               | 1.037  | 1.037  | 1.037  | 1.037  | 1.037  |
| Air VFR            | m <sup>3</sup> /h  | 1.630  | 1.380  | 1.140  | 0.900  | 0.650  |
| ER                 | -                  | 0.797  | 0.676  | 0.557  | 0.438  | 0.318  |
| N <sub>2</sub> VFR | m <sup>3</sup> /h  | 0.320  | 0.320  | 0.320  | 0.320  | 0.320  |
| T <sub>3-4</sub>   | °C                 | 817    | 824    | 823    | 825    | 826    |
| T <sub>1-2</sub>   | °C                 | 819    | 827    | 824    | 825    | 824    |
| Other              | vol%               | 76.35  | 68.65  | 63.30  | 61.77  | 50.76  |
| CO                 | vol%               | 2.92   | 4.87   | 5.99   | 4.78   | 9.08   |
| CO <sub>2</sub>    | vol%               | 15.32  | 16.74  | 17.50  | 18.67  | 17.70  |
| CH <sub>4</sub>    | vol%               | 1.22   | 1.39   | 1.57   | 1.84   | 2.03   |
| H <sub>2</sub>     | vol%               | 3.79   | 7.93   | 11.24  | 12.51  | 20.12  |
| O <sub>2</sub>     | vol%               | 0.40   | 0.42   | 0.40   | 0.43   | 0.31   |
| Total              | vol%               | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 |
| LHV                | MJ/Nm <sup>3</sup> | 1.22   | 1.97   | 2.53   | 2.61   | 4.05   |

**Table 3.2** Lab-scale experimental results - Gasification of coal Sample-2.

| Parameter          | Unit               | ER1    | ER2    | ER3    | ER4    | ER5    |
|--------------------|--------------------|--------|--------|--------|--------|--------|
| Coal MFR           | kg/h               | 1.042  | 1.042  | 1.042  | 1.042  | 1.042  |
| Air VFR            | m <sup>3</sup> /h  | 1.630  | 1.380  | 1.140  | 0.890  | 0.650  |
| ER                 | -                  | 0.638  | 0.541  | 0.445  | 0.350  | 0.254  |
| N <sub>2</sub> VFR | m <sup>3</sup> /h  | 0.320  | 0.320  | 0.320  | 0.320  | 0.320  |
| T <sub>3</sub>     | °C                 | 829    | 830    | 823    | 825    | 825    |
| T <sub>2</sub>     | °C                 | 825    | 825    | 827    | 824    | 823    |
| Other              | vol%               | 73.83  | 66.58  | 62.43  | 58.58  | 50.56  |
| CO                 | vol%               | 5.81   | 7.72   | 9.47   | 9.53   | 11.76  |
| CO <sub>2</sub>    | vol%               | 13.19  | 15.59  | 16.20  | 17.06  | 17.06  |
| CH <sub>4</sub>    | vol%               | 1.23   | 1.39   | 1.44   | 1.68   | 2.11   |
| H <sub>2</sub>     | vol%               | 5.66   | 8.31   | 10.05  | 12.90  | 18.40  |
| O <sub>2</sub>     | vol%               | 0.28   | 0.41   | 0.41   | 0.25   | 0.11   |
| Total              | vol%               | 100.00 | 100.00 | 100.00 | 100.00 | 100.00 |
| LHV                | MJ/Nm <sup>3</sup> | 1.79   | 2.37   | 2.80   | 3.20   | 4.23   |

Lower heating values of syngas at the reactor exit are calculated from the volumetric concentrations of CO, CH<sub>4</sub> and H<sub>2</sub> by using formula given in equation 2.3. According to the experimental results, it can be said that LHV of the syngas per unit volume decreases with increasing ER values for all cases. Moreover, unreacted oxygen concentration is

measured below %1 for all samples and all ER values. Hence, it is evaluated that almost all amount of oxygen is consumed during gasification experiments.

Graphical comparisons of the experimental results for molar concentration of each gaseous species and LHV values can be seen from the figures presented in Appendix-A.

**Table 3.3** Lab-scale experimental results - Gasification of coal Sample-3.

| Parameter          | Unit               | ER2    | ER3    | ER4    | ER5    |
|--------------------|--------------------|--------|--------|--------|--------|
| Coal MFR           | kg/h               | 1.039  | 1.039  | 1.039  | 1.039  |
| Air VFR            | m <sup>3</sup> /h  | 3.260  | 2.700  | 2.060  | 1.560  |
| ER                 | -                  | 0.558  | 0.463  | 0.353  | 0.267  |
| N <sub>2</sub> VFR | m <sup>3</sup> /h  | 0.320  | 0.320  | 0.320  | 0.320  |
| T <sub>3</sub>     | °C                 | 825    | 827    | 825    | 827    |
| T <sub>2</sub>     | °C                 | 835    | 832    | 825    | 826    |
| Other              | vol%               | 78.26  | 73.23  | 66.76  | 63.22  |
| CO                 | vol%               | 2.68   | 6.20   | 10.76  | 11.41  |
| CO <sub>2</sub>    | vol%               | 15.40  | 14.09  | 11.63  | 12.42  |
| CH <sub>4</sub>    | vol%               | 1.18   | 1.61   | 1.83   | 2.13   |
| H <sub>2</sub>     | vol%               | 1.50   | 4.15   | 8.14   | 10.52  |
| O <sub>2</sub>     | vol%               | 0.98   | 0.72   | 0.88   | 0.30   |
| Total              | vol%               | 100.00 | 100.00 | 100.00 | 100.00 |
| LHV                | MJ/Nm <sup>3</sup> | 0.92   | 1.81   | 2.89   | 3.34   |

**Table 3.4** Lab-scale experimental results - Gasification of coal Sample-4.

| Parameter          | Unit               | ER2    | ER3    | ER4    | ER5    |
|--------------------|--------------------|--------|--------|--------|--------|
| Coal MFR           | kg/h               | 1.049  | 1.049  | 1.049  | 1.049  |
| Air VFR            | m <sup>3</sup> /h  | 3.290  | 2.730  | 2.080  | 1.570  |
| ER                 | -                  | 0.487  | 0.404  | 0.308  | 0.233  |
| N <sub>2</sub> VFR | m <sup>3</sup> /h  | 0.320  | 0.320  | 0.320  | 0.320  |
| T <sub>3</sub>     | °C                 | 825    | 825    | 825    | 827    |
| T <sub>2</sub>     | °C                 | 827    | 830    | 825    | 826    |
| Other              | vol%               | 79.06  | 76.46  | 73.71  | 61.53  |
| CO                 | vol%               | 2.20   | 4.70   | 7.81   | 13.45  |
| CO <sub>2</sub>    | vol%               | 15.88  | 14.03  | 10.94  | 12.71  |
| CH <sub>4</sub>    | vol%               | 1.40   | 1.66   | 2.00   | 2.48   |
| H <sub>2</sub>     | vol%               | 1.18   | 2.85   | 5.15   | 9.81   |
| O <sub>2</sub>     | vol%               | 0.28   | 0.30   | 0.39   | 0.02   |
| Total              | vol%               | 100.00 | 100.00 | 100.00 | 100.00 |
| LHV                | MJ/Nm <sup>3</sup> | 0.91   | 1.50   | 2.26   | 3.65   |

### 3.2. Repeatability and Uncertainty of the Measurements for Syngas Output

In order to calculate the error in experimental results and to determine measurement uncertainty during the experiments due to measuring equipment, operating conditions, personnel, etc.; additional experiments are carried out on two different dates. Sample-2 is chosen for repeatability experiments. Three cases with different ER values given in Table 3.2 are investigated twice experimentally.

**Table 3.5** The results of repeatability experiments – Sample-2

| Measured Variable | ER1   |       |       | ER2   |       |       | ER3   |       |       |
|-------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
|                   | Exp-1 | Exp-2 | Diff. | Exp-1 | Exp-2 | Diff. | Exp-1 | Exp-2 | Diff. |
| N <sub>2</sub>    | 73.83 | 70.46 | 3.37  | 66.58 | 65.17 | 1.41  | 62.43 | 61.7  | 0.73  |
| CO                | 5.81  | 5.66  | 0.15  | 7.72  | 7.44  | 0.28  | 9.47  | 9.83  | 0.36  |
| CO <sub>2</sub>   | 13.19 | 16.63 | 3.44  | 15.59 | 17.06 | 1.47  | 16.2  | 16.04 | 0.16  |
| CH <sub>4</sub>   | 1.23  | 1.3   | 0.07  | 1.39  | 1.45  | 0.06  | 1.44  | 1.48  | 0.04  |
| H <sub>2</sub>    | 5.66  | 5.8   | 0.14  | 8.31  | 8.47  | 0.16  | 10.05 | 10.64 | 0.59  |
| O <sub>2</sub>    | 0.28  | 0.15  | 0.13  | 0.41  | 0.41  | 0     | 0.41  | 0.31  | 0.1   |
| LHV               | 1.79  | 1.81  | 0.02  | 2.37  | 2.37  | 0     | 2.8   | 2.92  | 0.12  |

The results of repeatability experiments are tabulated in Table 3.5. In this table, syngas composition values are given as volumetric fractions (vol%) and LHV is given in unit MJ/Nm<sup>3</sup>. The difference between the 1<sup>st</sup> and 2<sup>nd</sup> experiments is quite small in the measurement values of the product gas components as seen in the table. In the measurement values of CO, CH<sub>4</sub> and H<sub>2</sub> components, which play a role in the calculation of LHV, the difference between the 1<sup>st</sup> and 2<sup>nd</sup> experiments was in the range of 0.04% - 0.59%. The difference in the calculated LHV values was 0.00-0.12 MJ/Nm<sup>3</sup>. As a result, the difference for LHV parameter is assumed to be  $\pm 0.12$  MJ/Nm<sup>3</sup>.

In addition, the measurement uncertainty values of gas analyzer and its modules used in the experiments for each gas species are given in Table 3.6. According to this table, it can be said that uncertainty values of CO, CH<sub>4</sub> and H<sub>2</sub> species obtained from the experiments are in the range of the analyzer datasheet values.

**Table 3.6** Measurement uncertainty of gas analyzer and its modules

| Gas analyzer  | Species         | Module     | Principle            | Uncertainty |
|---------------|-----------------|------------|----------------------|-------------|
| ABB<br>AO2040 | CO <sub>2</sub> | Uras26     | IR                   | 1%          |
|               | CO              | Uras26     | IR                   | 1%          |
|               | CH <sub>4</sub> | Uras26     | IR                   | 1%          |
|               | H <sub>2</sub>  | Caldos 17  | Thermal conductivity | 1%          |
|               | O <sub>2</sub>  | Magnos 106 | Magneto-mechanical   | 1%          |

### 3.3. Results of Devolatilization Modelling

Mass fraction coefficients of devolatilization mechanism calculated by using MGAS model for each different coal sample are given in Table 3.7. Since Sample-2 and Sample-4 are dried forms of Sample-1 and Sample-4, respectively, the volatile composition was obtained very close to each other for dry and wet samples.

**Table 3.7** Mass fraction coefficients obtained from MGAS model (equation 2.78)

| Mass fraction    | Sample-1  | Sample-2  | Sample-3  | Sample-4  |
|------------------|-----------|-----------|-----------|-----------|
| $\alpha_{Tar}^d$ | 0.3002610 | 0.3007400 | 0.4459390 | 0.4459920 |
| $\beta_{CO}^d$   | 0.0893131 | 0.0892671 | 0.0476297 | 0.0476264 |
| $\beta_{CO_2}^d$ | 0.0651524 | 0.0651189 | 0.0347451 | 0.0347427 |
| $\beta_{CH_4}^d$ | 0.0634598 | 0.0633245 | 0.1318050 | 0.1317840 |
| $\beta_{H_2}^d$  | 0.0135858 | 0.0135568 | 0.0282176 | 0.0282130 |
| $\beta_{H_2O}^d$ | 0.2995240 | 0.2993700 | 0.1597330 | 0.1597220 |
| $\beta_{H_2S}^d$ | 0.1440890 | 0.1440200 | 0.1176790 | 0.1176720 |
| $\beta_{NH_3}^d$ | 0.0246148 | 0.0246029 | 0.0342502 | 0.0342481 |

Same mass coefficients of tar cracking reactions are used for different coal samples. Additionally, mass coefficients of tar oxidation reaction are constant for different samples as shown in Table 3.8.

**Table 3.8** Mass fraction coefficients of tar cracking (equation 2.90) and tar oxidation reactions (equation 2.92) obtained with MGAS model

| Tar cracking reaction |           | Tar oxidation reaction |          |
|-----------------------|-----------|------------------------|----------|
| Mass fractions        | Value     | Mass fractions         | Value    |
| $\alpha_{char}^c$     | 0.646661  | $\beta_{O_2}^o$        | 3.01265  |
| $\beta_{CO}^c$        | 0.0350145 | $\beta_{CO_2}^o$       | 3.29771  |
| $\beta_{CO_2}^c$      | 0         | $\beta_{H_2O}^o$       | 0.714935 |
| $\beta_{CH_4}^c$      | 0.318324  | $\beta_{SO_3}^o$       | 0        |
| $\beta_{H_2}^c$       | 0         | $\beta_{N_2}^o$        | 0        |
| $\beta_{H_2O}^c$      | 0         |                        |          |

Reaction rates are obtained for devolatilization, tar cracking and tar oxidation mechanisms by MGAS model for all coal samples as shown in Table 3.9.

**Table 3.9** Reaction rate parameters used in MGAS model

| Mechanism        | $A$ [variable]                 | $E$ [K] |
|------------------|--------------------------------|---------|
| Devolatilization | $5.1 \times 10^4$ [1/s]        | 8153    |
| Tar Cracking     | $2.1 \times 10^8$ [1/s]        | 13336   |
| Tar Oxidation    | $3.8 \times 10^{11}$ [1/atm.s] | 15097   |

Since CPD model takes operating conditions into account, physical properties such as reactor temperature, static pressure, heating rate, etc. should be defined as input parameters. CPD model input parameters are given in Table 3.10.

**Table 3.10** Input parameters used in CPD model

| Input Parameters    | Unit | Value   |
|---------------------|------|---------|
| Initial Temperature | K    | 293.15  |
| Final Temperature   | K    | 1098.15 |
| Absolute Pressure   | Pa   | 110000  |
| Heating Rate        | K/s  | 2       |
| Period              | s    | 600     |
| Specific gravity    | -    | 1.17    |

At the end of the calculations, it is seen that CPD model is not capable to solve coal types that involve very high-ash and high-moisture content. The solver crashed and did not give any results for Sample-1 and Sample-2. It's thought that proximate and ultimate analysis data of these samples are out of the range of the NMR database of the CPD model. Hence, the solver could not able to solve the problem. However, the numerical results are obtained for Sample-1 & Sample-2.

Devolatilization reaction constants obtained from the calculations by using CPD model are listed in Table 3.11. Mass fractions of the species for tar oxidation reaction are given in Table 3.12. Kinetic rates of devolatilization and tar oxidation reactions used in CPD model are also given in Table 3.13.

**Table 3.11** Mass fraction coefficients of devolatilization reaction obtained from CPD model for Sample-3 & Sample-4 (equation 2.106)

| Mass fraction    | Sample-3 & Sample-4 |
|------------------|---------------------|
| $\alpha_{Tar}^d$ | 0.522614            |
| $\beta_{CO}^d$   | 0.0565361           |
| $\beta_{CO_2}^d$ | 0.0598456           |
| $\beta_{CH_4}^d$ | 0.140099            |
| $\beta_{H_2O}^d$ | 0.220905            |

**Table 3.12** Mass fraction coefficients of tar oxidation reaction (equation 2.92) obtained with CPD model for Sample-3 & Sample-4

| Tar oxidation reaction<br>Mass fraction | Value     |
|-----------------------------------------|-----------|
| $\beta_{O_2}^o$                         | 3.22635   |
| $\beta_{CO_2}^o$                        | 1.34661   |
| $\beta_{H_2O}^o$                        | 2.05829   |
| $\beta_{SO_3}^o$                        | 0.745482  |
| $\beta_{N_2}^o$                         | 0.0759611 |

**Table 3.13** Reaction rate parameters used in CPD model

| Mechanism        | $A$ [variable]                 | $E$ [K] | $n$     |
|------------------|--------------------------------|---------|---------|
| Devolatilization | 7.52446 [1/s]                  | 12392   | 1.16047 |
| Tar Oxidation    | $3.8 \times 10^{11}$ [1/atm.s] | 15097   | -       |

### 3.4. Numerical Analysis Setup

An atmospheric, autothermal bubbling fluidized bed gasifier consisting of a reactor, a cyclone and a screw feeder is taken into consideration for lab-scale studies. A 3D geometrical model of the flow domain of lab-scale gasification reactor created with CAD software (Solidworks) is given in Figure 3.1. For the computational setup, the real geometry was simplified in order to reduce computational time cost by removing the cyclone and using air injection points instead of distributor plate geometry. Cartesian grid generation method is applied to this simplified geometry shown in Figure 3.2 and a computational grid with hexahedral elements was generated. Appropriate mesh size is determined by a mesh independency study.

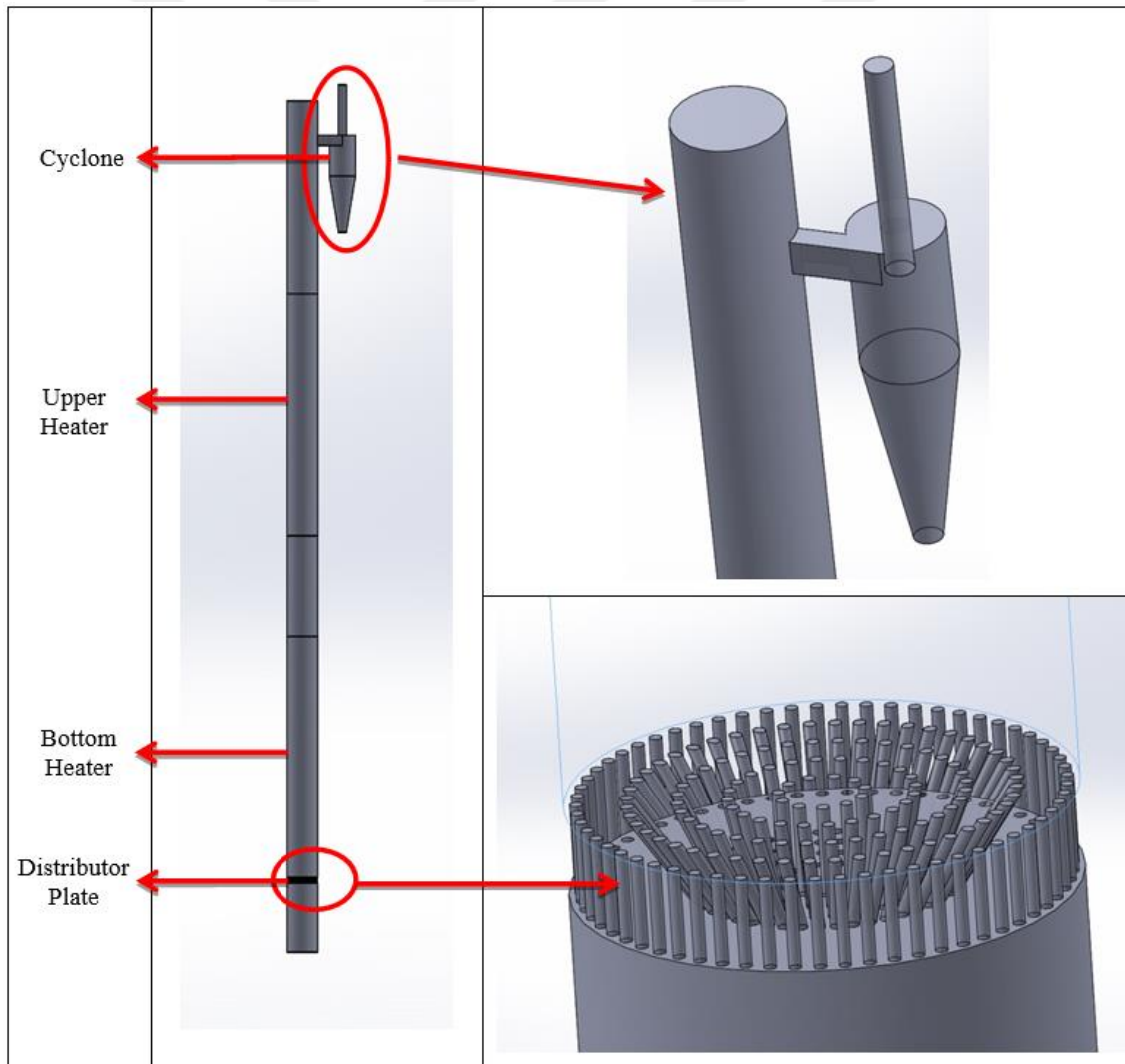


**Figure 3.1** 3D geometrical model of flow domain gasifier reactor

Sample-3 is chosen for numerical validation studies. Approximate and ultimate analyses of the coal Sample-3 (washed sample) given in Table 2.3 are used in the numerical calculations. Coal particles fed to the reactor are carried by a  $N_2$  stream from an injection point located 200 mm above the reactor bottom. Particle sizes of the coal vary between 0.5-1 mm. Since a narrow particle size range is chosen, linear size distribution is applied during numerical setup.

### 3.4.1. Initial, Boundary and Operating Conditions

The temperature of the gasification process was controlled by applying an adiabatic boundary condition to the gasifier walls. Air mass flow rate is calculated to reach desired equivalence ratio for gasification reactions.

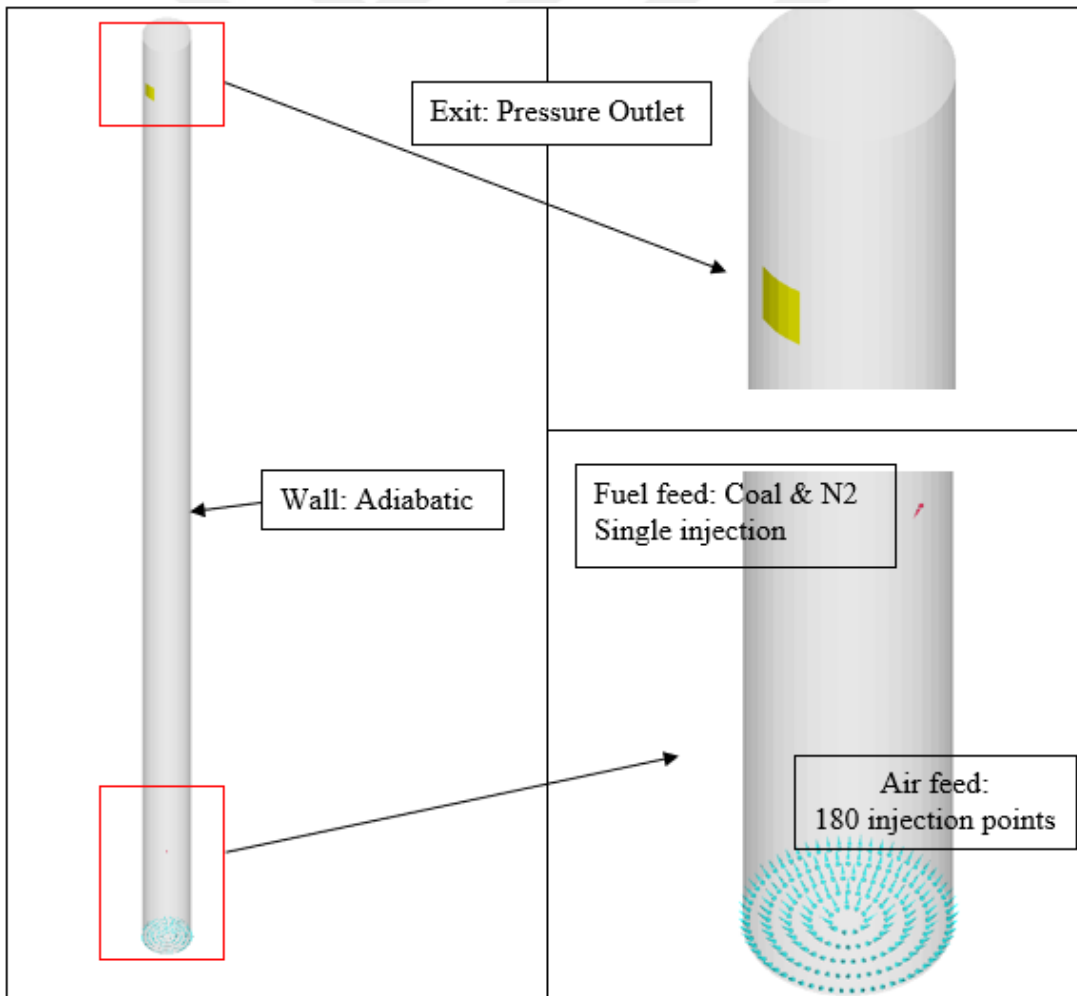


**Figure 3.2** Boundary conditions on simplified geometrical model

Air is fed to the reactor by equally spaced 180 injection points at the reactor bottom with various injection angles. Coal and nitrogen are supplied to the reactor at a single injection point with 200 mm height from reactor bottom. A pressure outlet boundary condition is defined for syngas exit at the coordinates of cyclone inlet. All boundary conditions of the computational model are shown in Figure 3.2. The specifications of the boundary conditions are listed in Table 3.14.

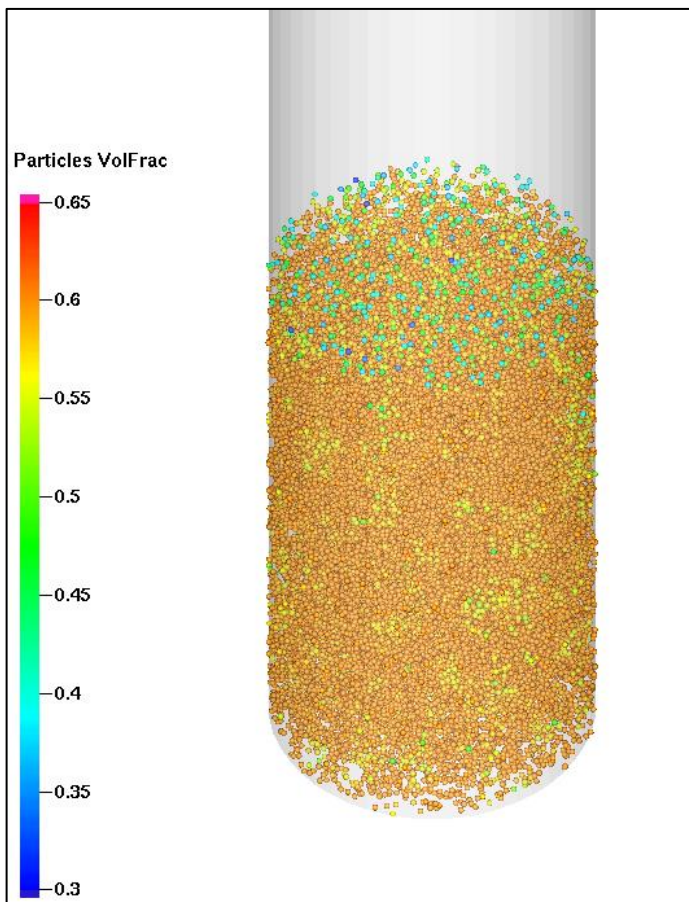
**Table 3.14** Boundary condition specifications

| Boundary Condition                         | Continuous Phase                                    | Discrete Phase                                                                                 |
|--------------------------------------------|-----------------------------------------------------|------------------------------------------------------------------------------------------------|
| Distributor Plate:<br>180 injection points | Air mass flow rate:<br>Varies with ER val.<br>@20°C | -                                                                                              |
| Fuel feeding:<br>Single injection          | N2 mass flow rate:<br>0.32 m <sup>3</sup> /h @20°C  | Coal mass flow rate:<br>~1.04 kg/h @20°C                                                       |
| Gasifier wall                              | Adiabatic wall                                      | Particle momentum retention<br>coefficients [97]:<br>Normal dir.: 0.85<br>Tangential dir.: 0.9 |
| Particle-particle<br>interaction           | -                                                   | Close-pack volume fraction: 0.61 [98]<br>Momentum redirection coefficient:<br>0.40 [97]        |
| Exit                                       | Pressure outlet:<br>101325 Pa                       | Particle exit                                                                                  |

In order to initialize the discrete phase computations, the numerical calculations are started with 150 mm static bed height, which contains 100% Silica sand at operating temperature. Initial velocity magnitude value for both air domain and solid particles is taken as zero. The static pressure of air is assumed to be atmospheric pressure for initialization (Table 3.15). Particle distributions with volume fraction for initial conditions are shown in Figure 3.3.

**Table 3.15** Initial conditions

| Initial Condition  | Continuous Phase | Discrete Phase                                          |
|--------------------|------------------|---------------------------------------------------------|
| Bed material       | 100% Air         | 100% Silica, 150mm initial bed height at reactor bottom |
| Temperature        | 825°C            | 825°C                                                   |
| Absolute Pressure  | 101325 Pa        | N/A                                                     |
| Velocity Magnitude | 0 m/s            | 0 m/s                                                   |

**Figure 3.3** Particle initialization – particle volume fraction

### 3.4.2. Mesh Independency Study

It is aimed to obtain proper computational mesh size to represent real fluid-particle interaction in the fluidized bed. Non-reactive cold-flow simulations (excluding energy equation) are carried out with 4 different mesh sizes. These grids involve 18k, 39k, 77k and 154k cartesian elements, respectively. Computational meshes prepared for mesh

independency study are presented in Figure 3.4 by using vertical midplane and horizontal section of the gasifier.

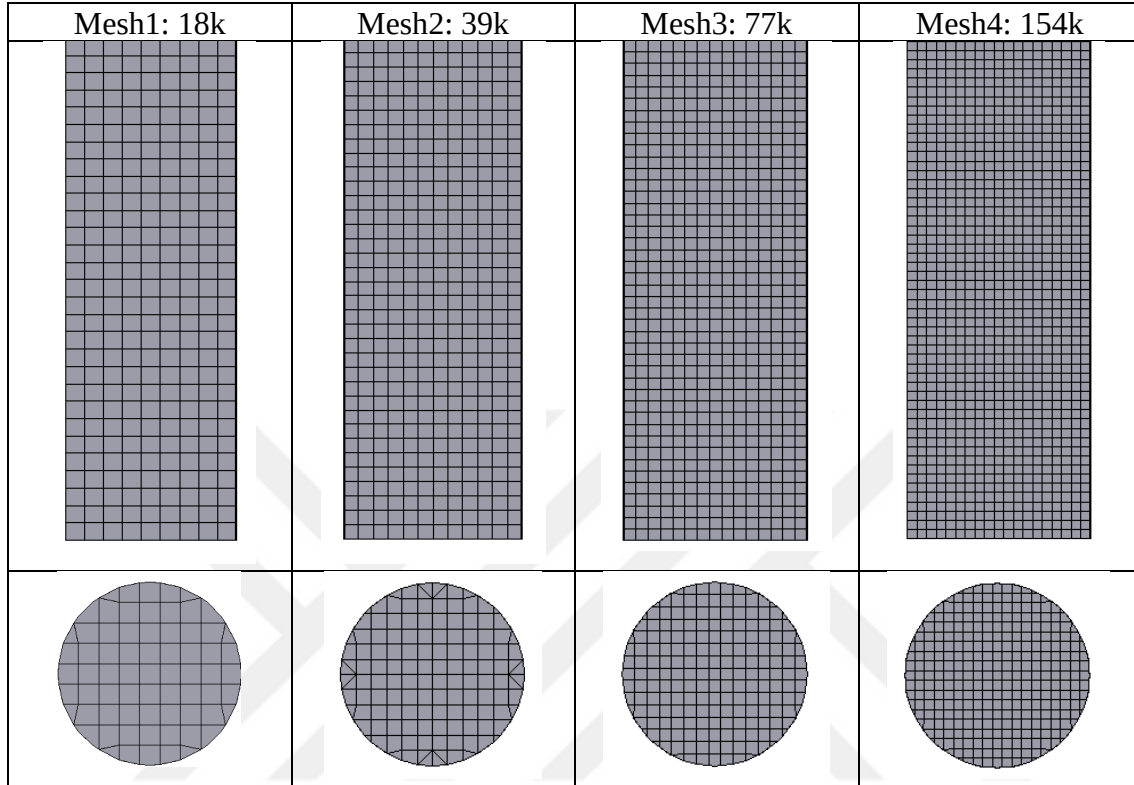


**Figure 3.4** Different mesh sizes used for mesh independency calculations

The computations are carried out with 0.001s time step for 100s physical time period by a GPU-based solver. Maximum computational cost is obtained 130 GPU-hours for Mesh-4 case. Time-averaged static pressure values on the vertical axis of the reactor for each case are given in Figure 3.5. As seen in the figure, while the static pressure values in the riser region are very close to each other, there is a variation between cases in the bed region. It can be seen from the figure that, relatively lower pressure values are obtained for Mesh-1.

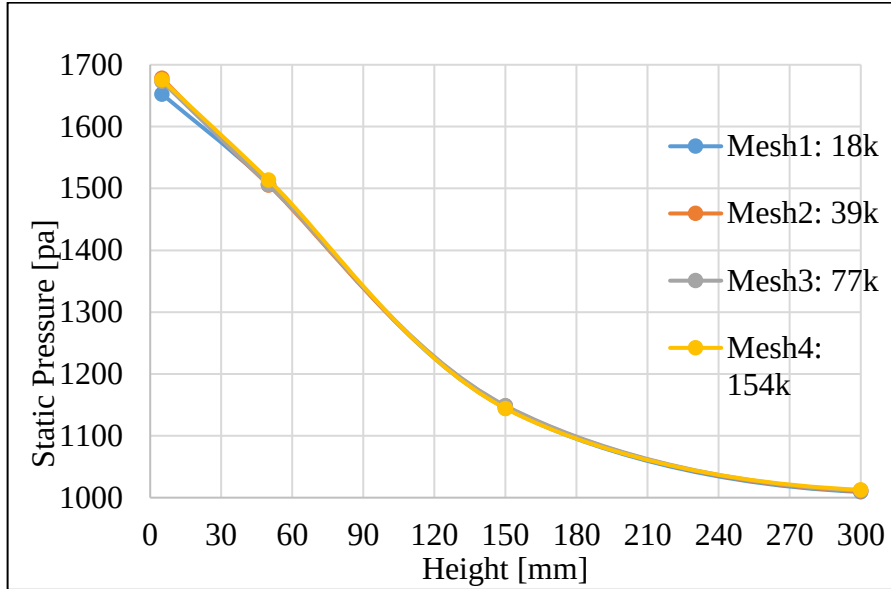


**Figure 3.5** Mean static pressure values on vertical axis

Additionally, instantaneous particle volume fraction distributions at fluidized bed regions for different meshes are plotted and compared (Figure 3.6). According to this comparison, the case involves 77k elements yields satisfactory bubbling fluidized bed characteristics and structure. It can be seen that there is a convergence for particle volume fraction distribution and bubble formation at Mesh-3.

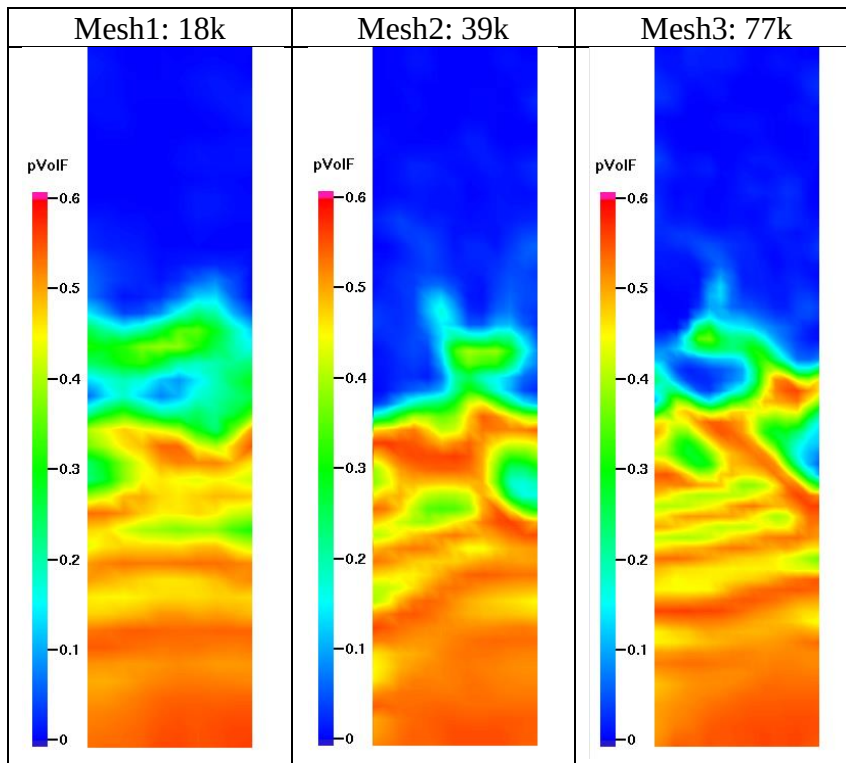


**Figure 3.6** Particle volume fraction distribution in fluidized bed region for different mesh sizes

At the end of the mesh independency study, minimum mesh size is determined approximately 80k for the lab-scale gasifier.

### 3.4.3. Other Solver Properties

A standard gravity condition is defined for numerical setup. It is assumed that coal particles enter the reactor with an injection expansion angle. LES constant, residual values for governing equations and ODE solver are chosen according to the suggestions in the literature. CPFD Barracuda VR software uses an adaptive time-step method that adjusts the time-step size during computations by checking convergence and the limits of the variables such as pressure and temperature. At the end of many calculations,  $5 \times 10^{-5}$  second is obtained as an appropriate time step for the cases of this thesis study. Investigated physical time for the simulations is determined as 50 s. All other solver settings are given in Table 3.16.

**Table 3.16** Other solver settings

| Parameter                             | Value                         | Comment                          |
|---------------------------------------|-------------------------------|----------------------------------|
| Gravity                               | $9.81 \text{ m/s}^2$          | -z direction                     |
| Coal feed expansion angle             | $15^\circ$                    | For both directions              |
| LES constant                          | 0.01                          | Smagorinsky constant             |
| Residual val. for continuity equation | $1 \times 10^{-7}$            |                                  |
| Residual val. for pressure term       | $1 \times 10^{-6}$            |                                  |
| Residual val. for momentum equations  | $1 \times 10^{-7}$            |                                  |
| Residual val. for energy equation     | $1 \times 10^{-6}$            |                                  |
| ODE solver tolerance val.             | $1 \times 10^{-9}$            |                                  |
| Initial particle cloud resolution     | 5 clouds/cell                 |                                  |
| Coal particle feed – Number Density   | 250                           | Particle feed per average volume |
| Time-step                             | $5 \times 10^{-5} \text{ s.}$ | Maximum time step                |
| Physical solution time period         | 50 s.                         |                                  |

### 3.5. Results of Numerical Analyses

The numerical calculations of reactive thermal cases for Sample-3 with different ER values are carried out by using MGAS devolatilization model. The computations are conducted on a GPU-based workstation in parallel. The computational cost for each case is approximately 105 GPU-hours.

Due to the usage of adaptive time step methodology, time-step had been varied between  $1 \times 10^{-5}$  s. and  $5 \times 10^{-5}$  s. during calculations. Time-step is reduced by the solver, especially in the case of a large temperature gradient value occurring in a cell close to the coal &  $N_2$  injection point.

While total number of coal particles is approximately  $4.3 \times 10^6$  in reality, these particles are represented by  $4.0 \times 10^4$  particle clouds in numerical calculations for chosen total time period. Hence, it can be commented that each numerical particle cloud represents 100 real coal particles on average.

In order to obtain syngas composition, a flux plane is defined at the pressure outlet and time-dependent mass fluxes of gaseous species are recorded. Then, mass fraction values are converted into mole fractions by using the molecular weight of each species. Transient syngas compositions obtained from the CFD calculations for Sample-3 with various ER values are plotted in Figure 3.7, Figure 3.8, Figure 3.9 and Figure 3.10. In these figures, mole fraction of  $N_2$  is shown on secondary vertical axis in order to investigate other species in detail with a narrow scale. According to these numerical results, it can be seen that the syngas composition reaches a quasi-steady-state condition after 30 seconds of physical time as shown in Figure 3.7. This situation is valid for the numerical analyses of all ER cases.

According to the time-dependent syngas compositions, there is a sudden change for  $H_2O$ ,  $N_2$ ,  $O_2$  and  $CO_2$  species at the transient period of 0-4s. High  $H_2O$  concentration values at the reactor exit show that the moisture release process is dominant in this period. Since high  $O_2$  concentrations are available at initial conditions, char content of coal particles rapidly and easily oxidized and high  $CO_2$  concentrations are obtained. Increase in  $H_2O$

and CO<sub>2</sub> concentrations induces a sudden decrease in O<sub>2</sub> and N<sub>2</sub> volume fractions at the end of this period.

After a physical time period of the first 4 seconds, firstly hydrogen then CO, H<sub>2</sub>S, NH<sub>3</sub> and CH<sub>4</sub> molecules start to appear in the syngas composition. Zero oxygen concentration values are calculated at the reactor exit after this period for all ER cases. Additionally, zero tar concentration in the syngas is obtained at any physical time during calculations. Thus, it can be said that all tar content released by the devolatilization process is cracked or oxidized and all oxygen molecules fed by air injection points are consumed by combustion and oxidation reactions in the reactor.

While mole fraction values for CO and CH<sub>4</sub> are decreasing with increasing air flow rates, it is vice versa for the concentration values of CO<sub>2</sub> and N<sub>2</sub> in syngas output. When the concentrations of H<sub>2</sub> for different ER cases are evaluated comparatively, it can be seen that ER5 case yields higher concentration than other cases. The molar concentration value of H<sub>2</sub> remains almost constant for ER4, ER3 and ER2 cases. Moreover, lower H<sub>2</sub>O content syngas output is obtained by increasing ER values.

As a general trend, higher fluctuations are observed for mole fraction values of all gaseous species with increasing ER values. When the time needed for the convergency of the concentration values for different gaseous species are compared for a specific ER value, it can be said that each of them has a different rate. In addition, it is seen that higher ER values yield more time to reach a quasi-steady-state regime. For example, while CO molar concentration reaches steady-state at a physical time of approximately 22s. in ER5 case, 30 s. physical time is needed for ER2 case.

Time-averaged data is obtained for molar concentrations of each gas species, fluid temperature and particle volume fraction parameters on the vertical mid-plane of the gasifier in order to analyze the numerical results of the gasification process. Time-averaged data comprise only the variable values obtained during the steady-state regime (i.e., physical time period of 30s.-50s.). Time-averaged distributions of these variables are shown in Figure 3.11, Figure 3.12, Figure 3.13 and Figure 3.14 for different ER cases.

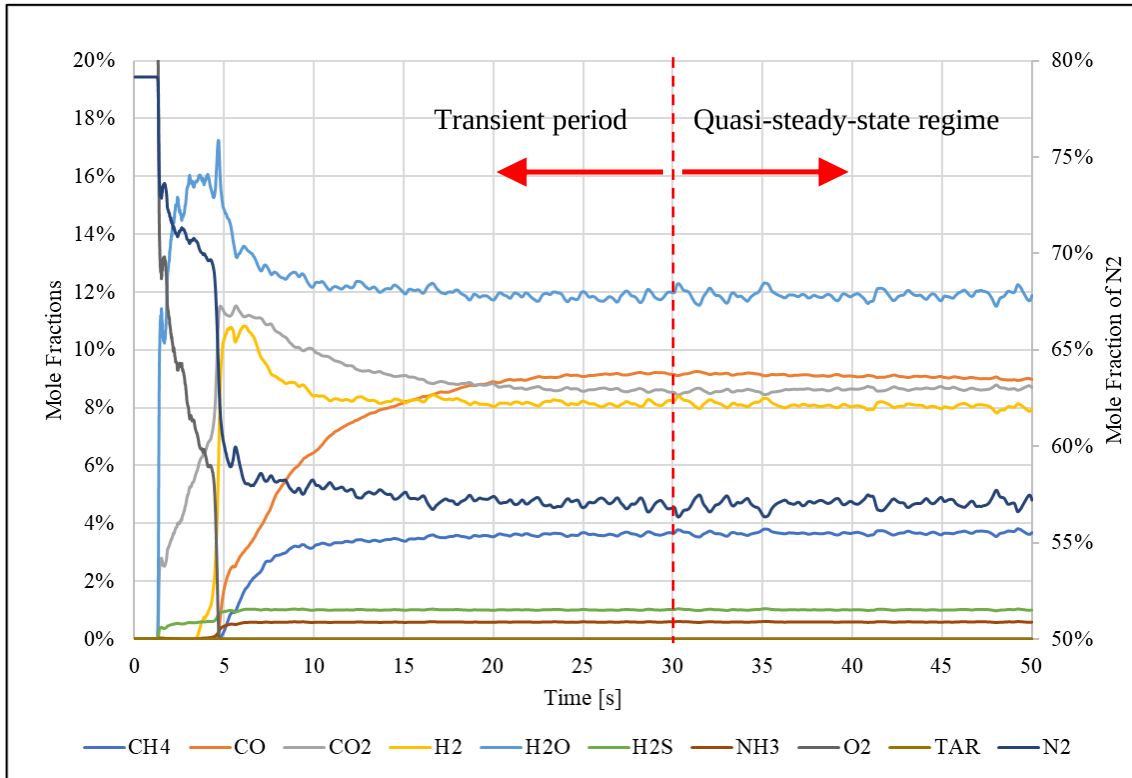


**Figure 3.7** Sample-3 ER5 - Transient syngas composition at reactor exit

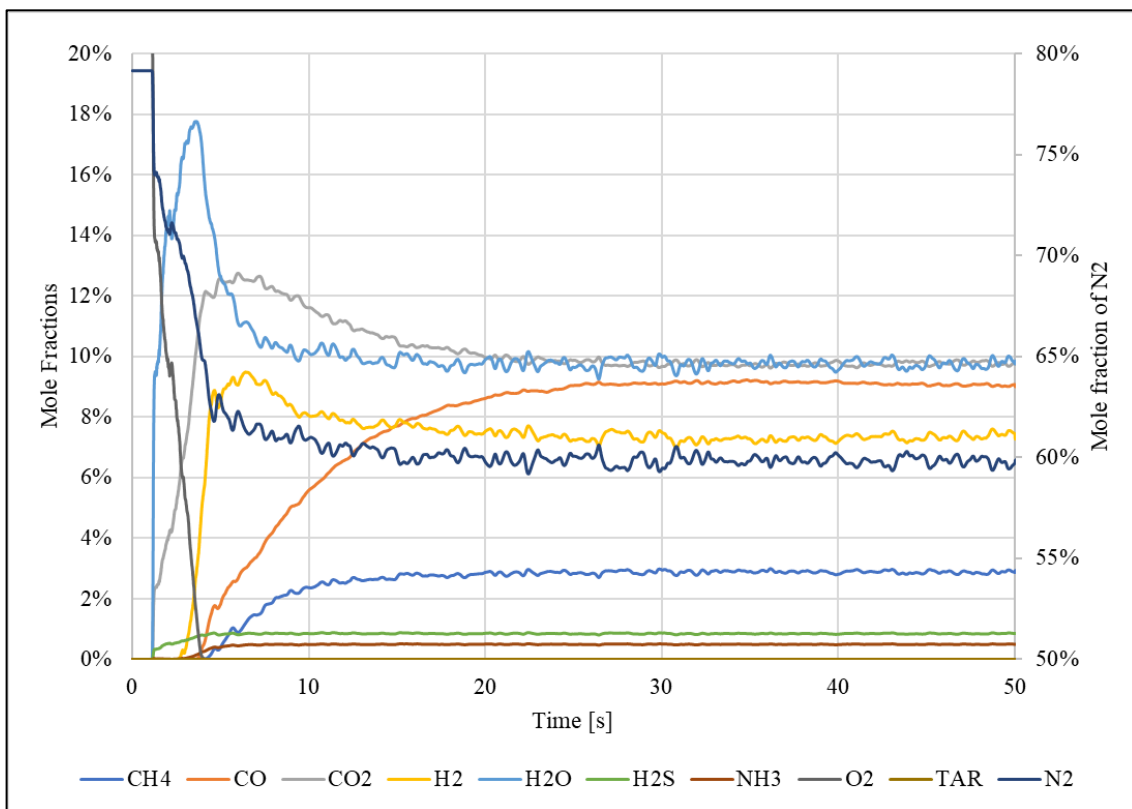


**Figure 3.8** Sample-3 ER4 - Transient syngas composition at reactor exit

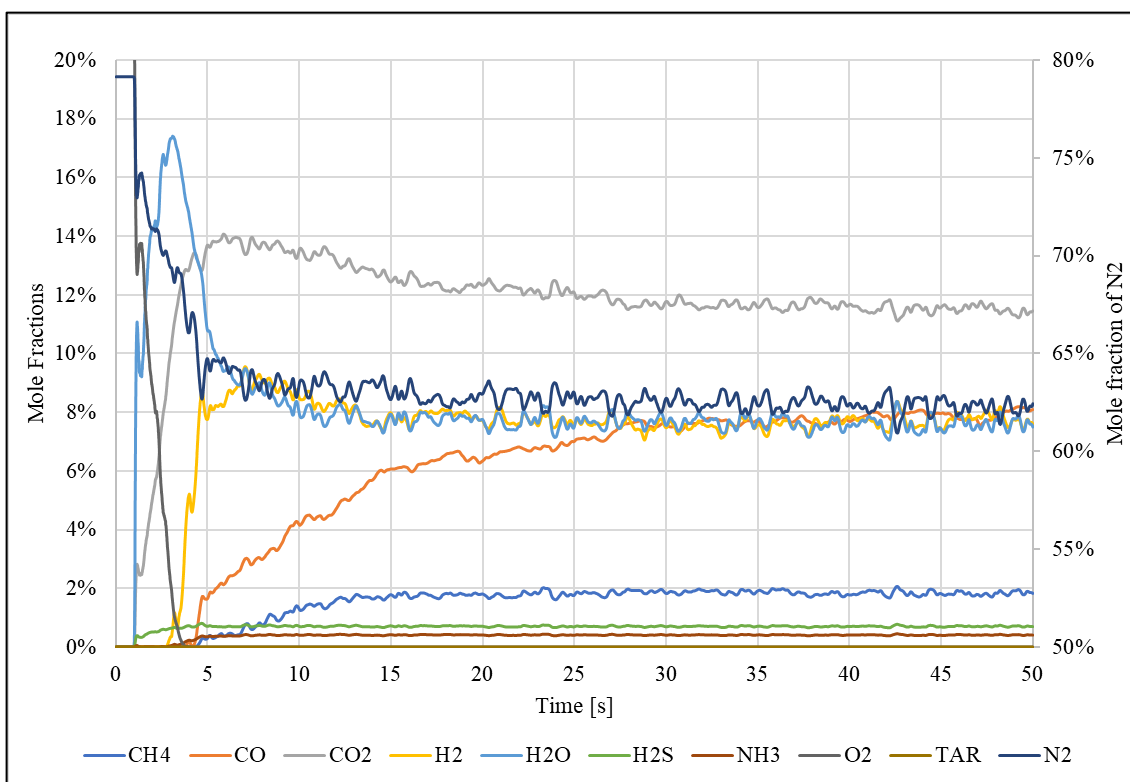


**Figure 3.9** Sample-3 ER3 - Transient syngas composition at reactor exit

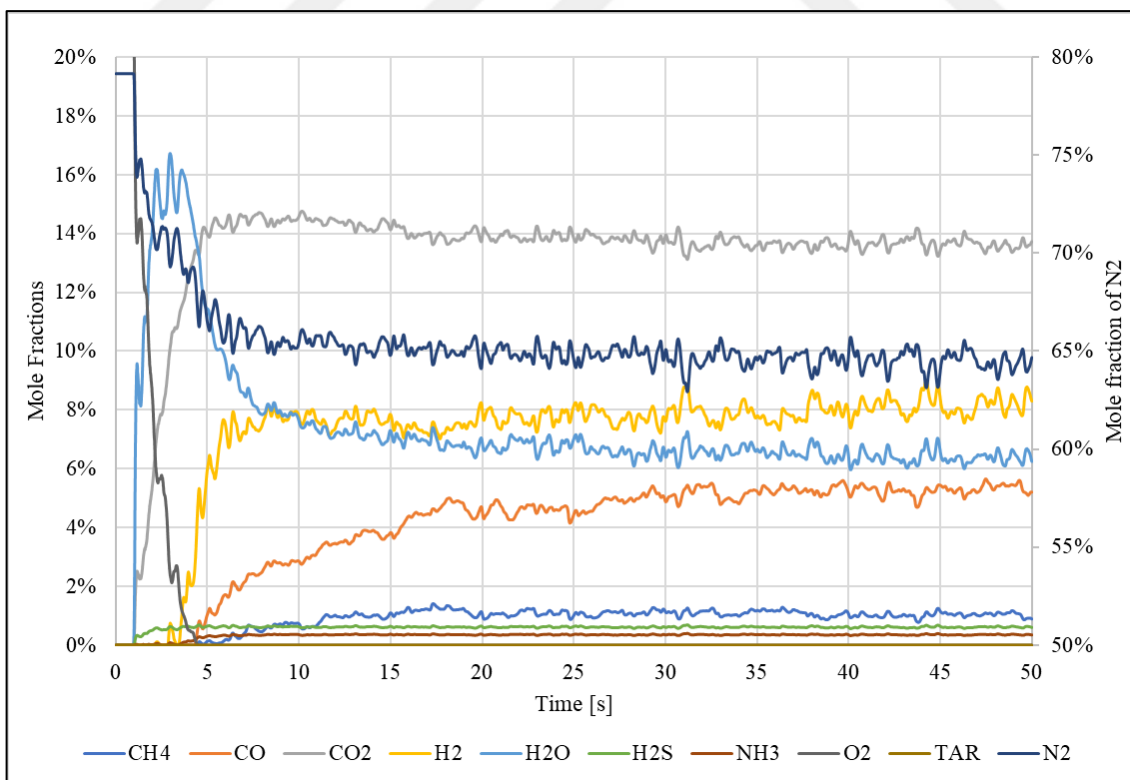


**Figure 3.10** Sample-3 ER2 - Transient syngas composition at reactor exit

All scales are kept same for the contour plot of specific variable in these figures in order to ease the comparison between numerical results of different ER cases.

When the particle volume fraction contours for different ER values are compared to each other, it can be seen that higher ER values yield higher bed heights and lower particle volume fraction values at the bed region.

Temperature distributions on the vertical axis show that while bed temperatures are very close to each other, there is a considerable difference in the free-stream region for different ER values. It can be commented that combustion reactions become dominant with increasing air flow rates and this causes higher temperature values.

High molar concentrations are obtained for  $\text{CH}_4$  and  $\text{H}_2\text{O}$  species in the coal feeding region for all cases. It is observed that these values decrease in the vertical direction with increasing height depending on gasification reactions. Additionally, it is seen that higher  $\text{H}_2$  concentrations are observed in the regions where the decrease in molar concentrations of  $\text{CH}_4$  and  $\text{H}_2\text{O}$  species occurs. It can be said that the methane reforming reaction becomes more effective in the upper regions of the gasifier and this reaction dominates especially for high ER values.

It is also determined that the molar concentrations of  $\text{CH}_4$ ,  $\text{CO}$ ,  $\text{H}_2\text{O}$ ,  $\text{H}_2\text{S}$  and  $\text{NH}_3$  in syngas decrease with increasing ER values while the molar concentrations of  $\text{N}_2$  and  $\text{CO}_2$  increase as a general trend.

Tar species appear only in the feed region for all ER cases. Numerical results show that the size of this region gets smaller with increasing ER values. It can be evaluated that kinetic rates of tar cracking and tar oxidation reactions increase with increasing bed temperatures on oxygen concentrations.

$\text{NH}_3$  and  $\text{H}_2\text{S}$  are the species released into the reactor by the devolatilization reaction. While an oxidation reaction is introduced for ammonia, released hydrogen sulfur does not react with any other species according to the chemistry defined in the numerical setup.

Contour plots show that the molar concentration of ammonia decreases in the vertical direction while no change is observed in the molar concentration of  $\text{H}_2\text{S}$ .

It can be said that  $\text{CO}$  and  $\text{CO}_2$  molecules start to be produced at relatively lower regions of the fluidized bed in all ER cases. According to the results, oxygen molecules in air supplied from the distributor plate into the reactor first react with the char content of the solid particles which substantially completed their own devolatilization process and reach relatively high temperatures. Moreover, it can be easily seen from the figures that most of the oxygen fed as a gasification agent is consumed in this region. However, with increasing air flow rates, oxygen molecules are able to reach the upper regions of the bed and are able to react with more molecules released as output of the devolatilization process.

Since  $\text{NO}_x$  formation reactions are not considered in the computations due to relatively low operating temperatures,  $\text{N}_2$  molecules do not react with any other species. Thus, there is not any change in the molar concentration of nitrogen in the free-stream region of the reactor. However, it is affected by ER value due to the flow rate of air.

In order to investigate the relationship between particle volume fraction and static pressure, time-averaged data is obtained on the reactor axis. Results are scaled to focus on the fluidized bed region and compared for different ER conditions in Figure 3.15. It is determined that pressure change with height is almost linear for all ER cases. A non-linear relationship between particle volume fraction and the bed height is obtained from the calculations. However, a general trend is obtained that lower particle concentrations yield higher pressure values at lower regions of the bed for all cases. In addition, relatively higher dynamic fluidized bed heights are observed for ER2 and ER3 cases.

Instantaneous contour plots are given in Figure A.6, Figure A.7, Figure A.8 and Figure A.9 for each ER case in Appendix-B.

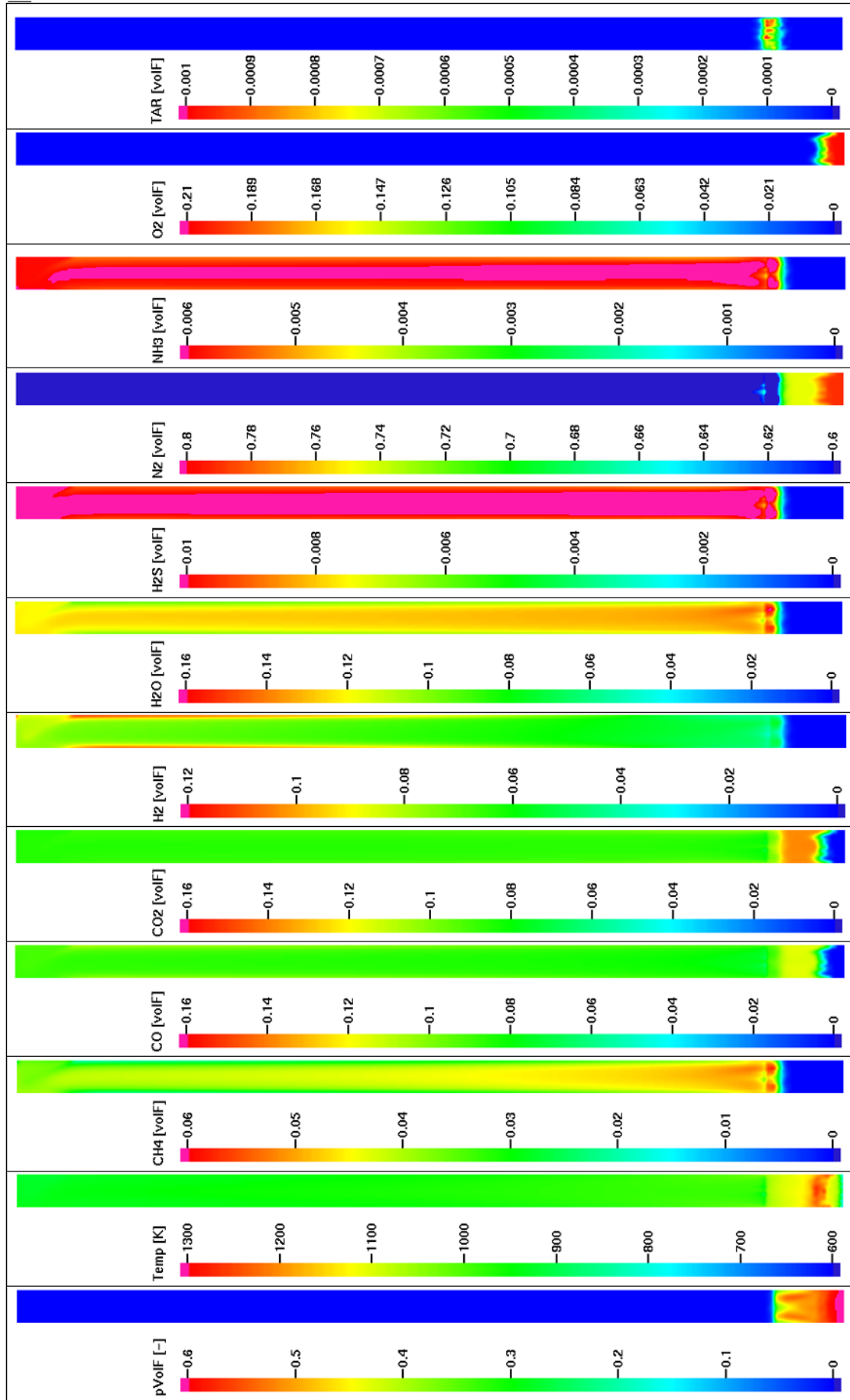


**Figure 3.11** Sample-3 ER5—Time averaged contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas species

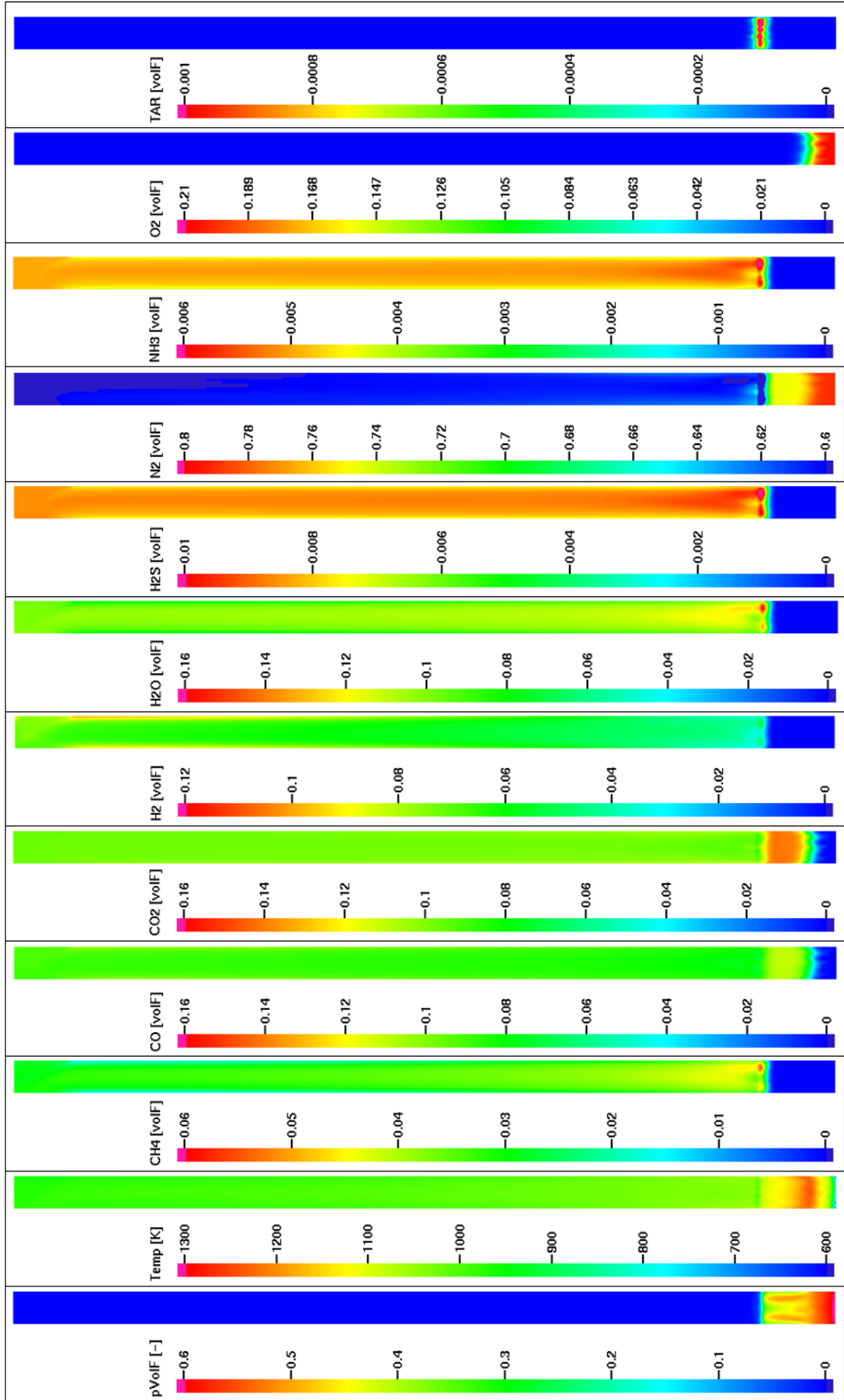


**Figure 3.12** Sample-3 ER4–Time averaged contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas. species

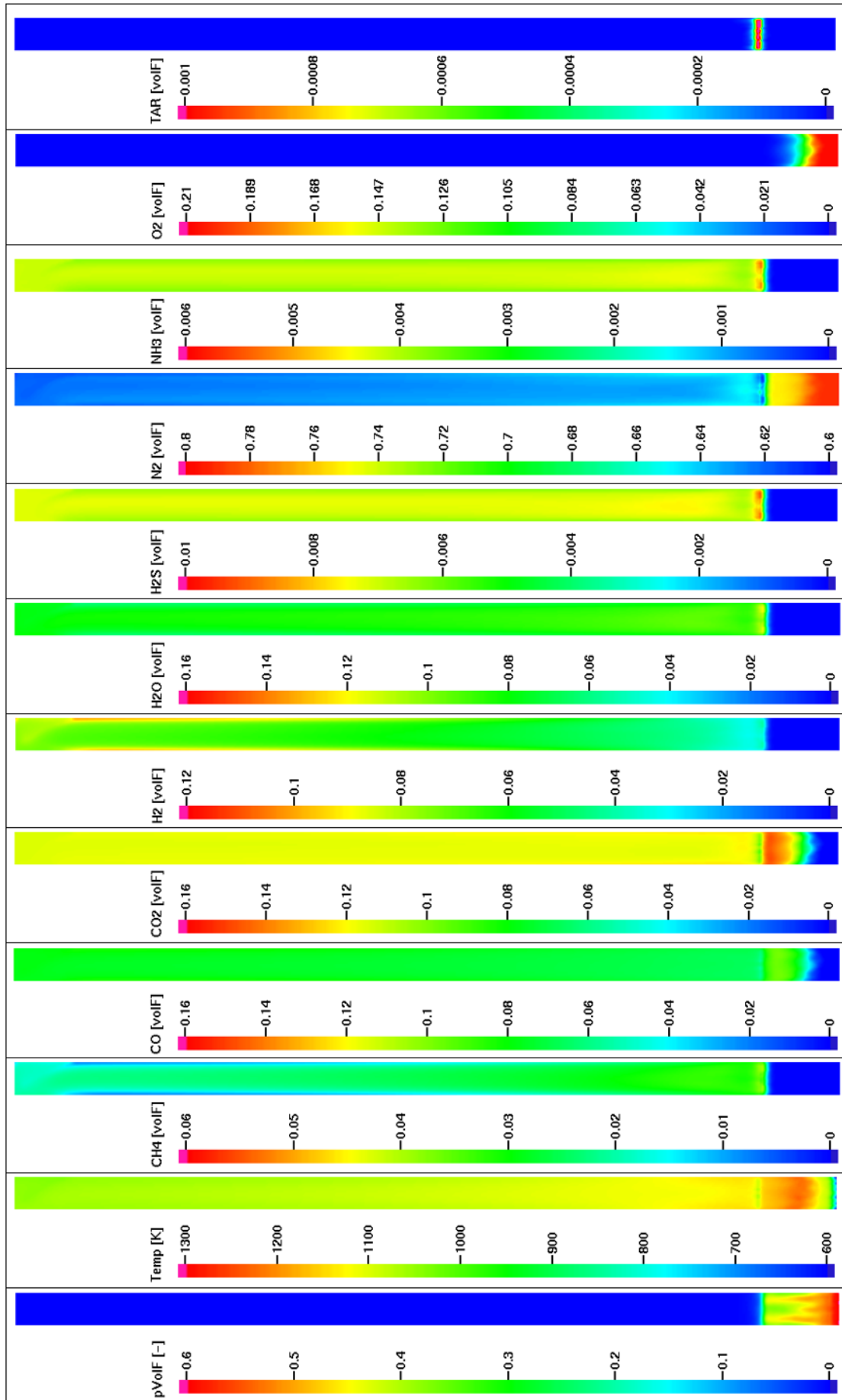


**Figure 3.13** Sample-3 ER3—Time averaged contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas species

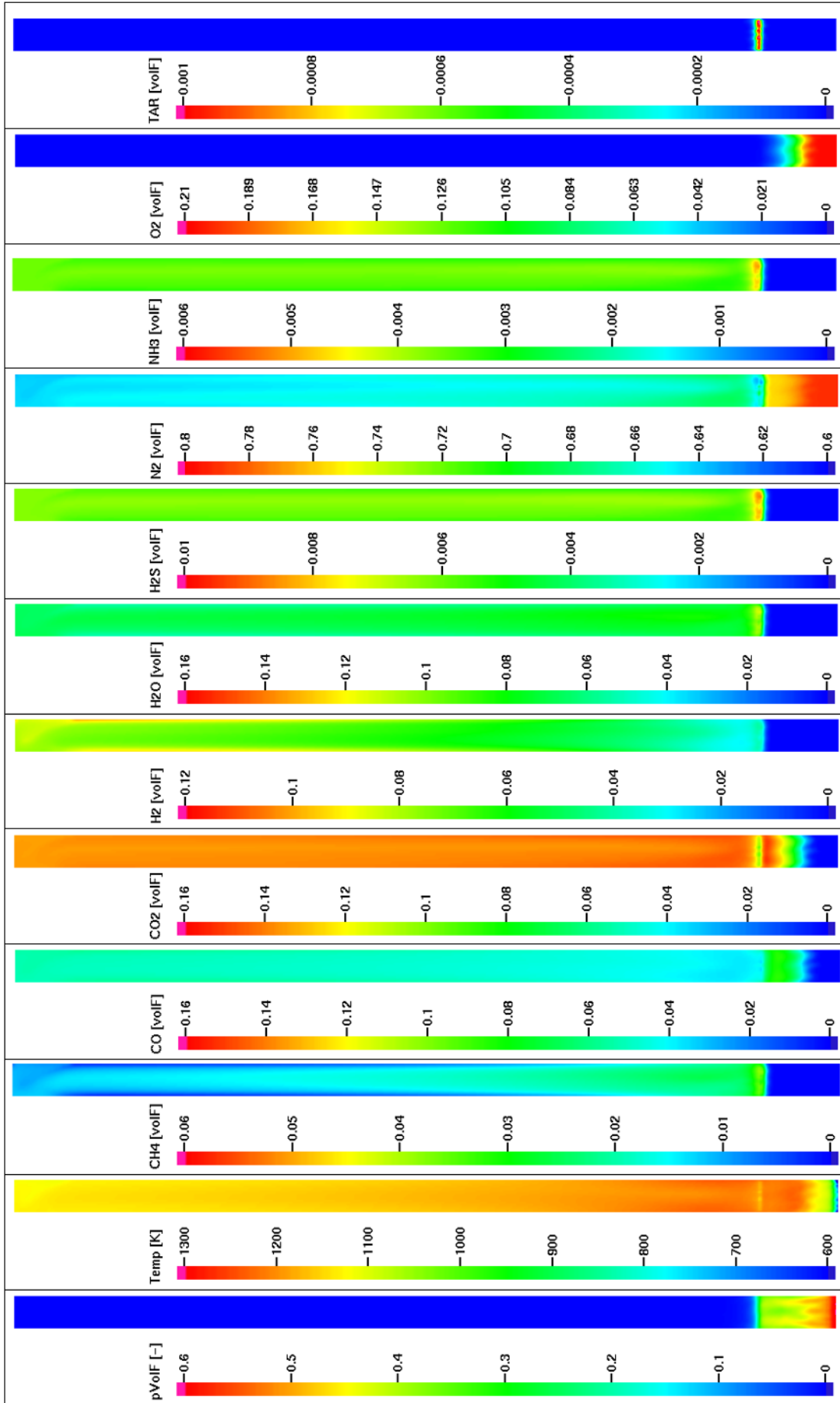


**Figure 3.14** Sample-3 ER2—Time averaged contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas species

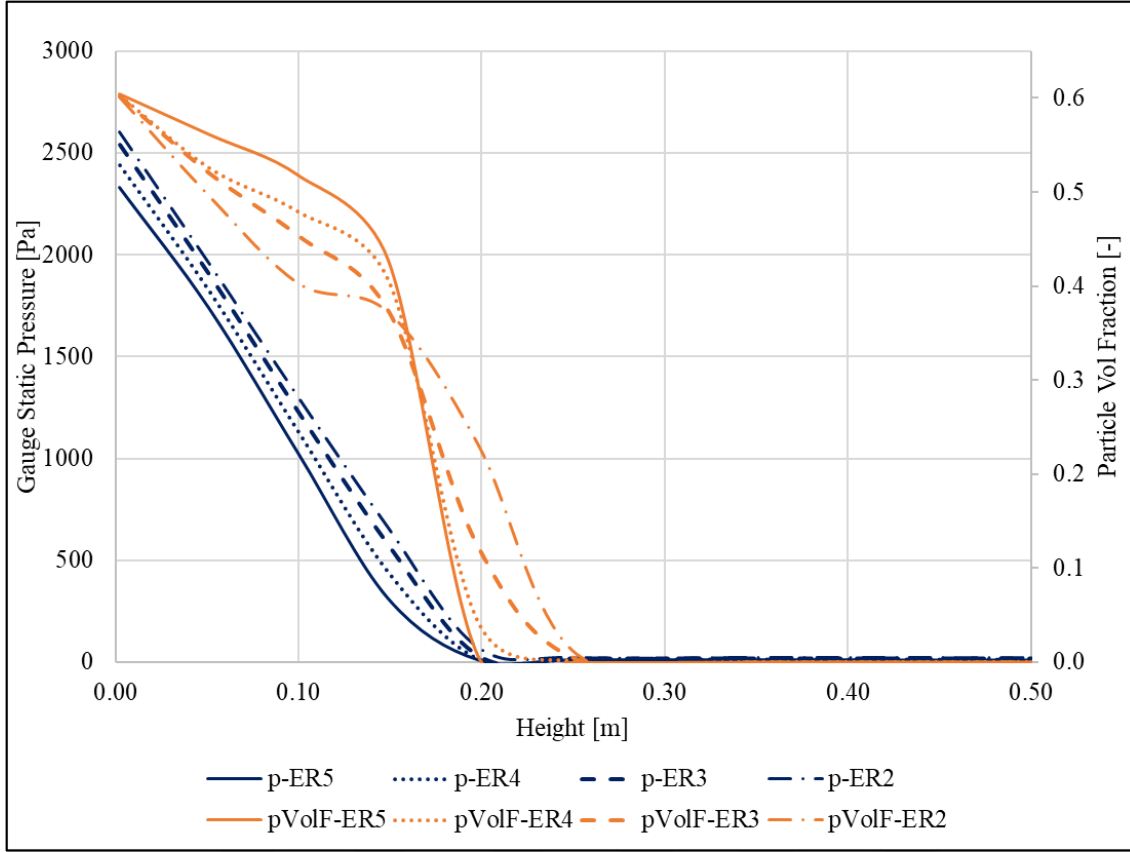


**Figure 3.15** Time-averaged particle volume fraction and static pressure values on reactor axis at fluidized bed region for different ER conditions

### 3.6. Comparison of Numerical and Experimental Results

The comparison of numerical and experimental results obtained for the molar concentrations of all gaseous species in syngas for different ER cases is given in Figure 3.16. Since five species are measured by an online gas analyzer as explained in Chapter 2, unmeasured content is represented as “other” species. As described in Chapter 2, the syngas vacuumed from the cyclone outlet is cooled before the inlet of the gas analyzer during experiments. This causes condensation of water and tar molecules in the cooling unit before analysis of gas composition. According to this assumption, other species involve  $\text{H}_2\text{S}$ ,  $\text{N}_2$  and  $\text{NH}_3$  for the numerical results obtained with MGAS model, while it is composed of  $\text{SO}_3$  and  $\text{N}_2$  for CPD model. Hence, the molar fraction values of methane, carbon monoxide, carbon dioxide, hydrogen and oxygen species obtained from the numerical analyses are converted into a “Dry and Tar Free (DTF)” form in order to compare numerical and experimental results accordingly. For this conversion, it is assumed that all water and tar content is condensed in the syngas cooler.

Other species are evaluated on a secondary axis in the graphic. Single mark is used for each experimental data and lines with marks are used for numerical results. Also, same color is used for both numerical and experimental values of a specific syngas content in order to ease the comparison.

It can be said that molar concentration values of CO, CO<sub>2</sub>, H<sub>2</sub>O and Other species obtained from numerical calculations are very close to the experimental data for ER5, and ER4 cases. According to the figure, there is a considerable difference is obtained for methane volume fraction in syngas in these cases. While CH<sub>4</sub> content of syngas is predicted approximately 4% at the end of the computations, it is measured by the gas analyzer as approximately 2% in experiments.

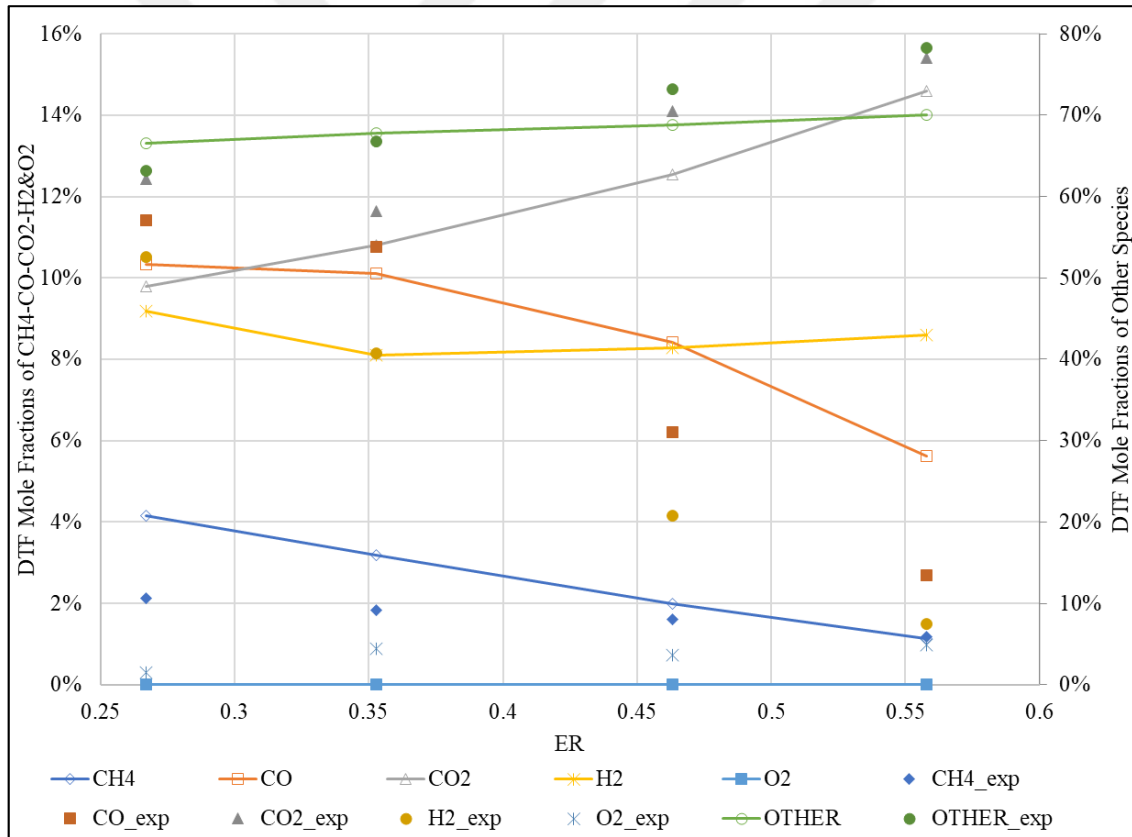


**Figure 3.16** Comparison of numerical and experimental concentration of gaseous species in the syngas output of gasification process of Sample-3 with different ER values

The gap between numerical and experimental data gets bigger for ER3 and ER2 cases. It is observed that calculated molar concentration of CO and H<sub>2</sub> species are higher than the experimental values. However, methane concentrations obtained from computations are very close to the experimental data for these ER cases.

Almost zero oxygen concentration in the syngas is numerically found for each case. However, the molar concentrations of  $O_2$  in the syngas are measured less than 1% for all ER cases in experiments, but are not equal to zero. Since experimental values are very small, the difference can be neglected.

Transient DTF mole fraction values for all ER cases can be seen in Figure A.10, Figure A.11, Figure A.12 and Figure A.13 in Appendix-C.

Lower heating value of the syngas is calculated by using volume fractions of methane, carbon monoxide and hydrogen species obtained from both numerical and experimental data (see equation 2.3). These results are compared in Figure 3.17. According to the calculated LHV values, a good agreement is obtained between experimental and numerical analysis, especially for lower ER values. Additionally, a similar trend is obtained between experimental and numerical results by considering the change in ER values. It can be said that higher ER values yield lower LHV values for syngas output.

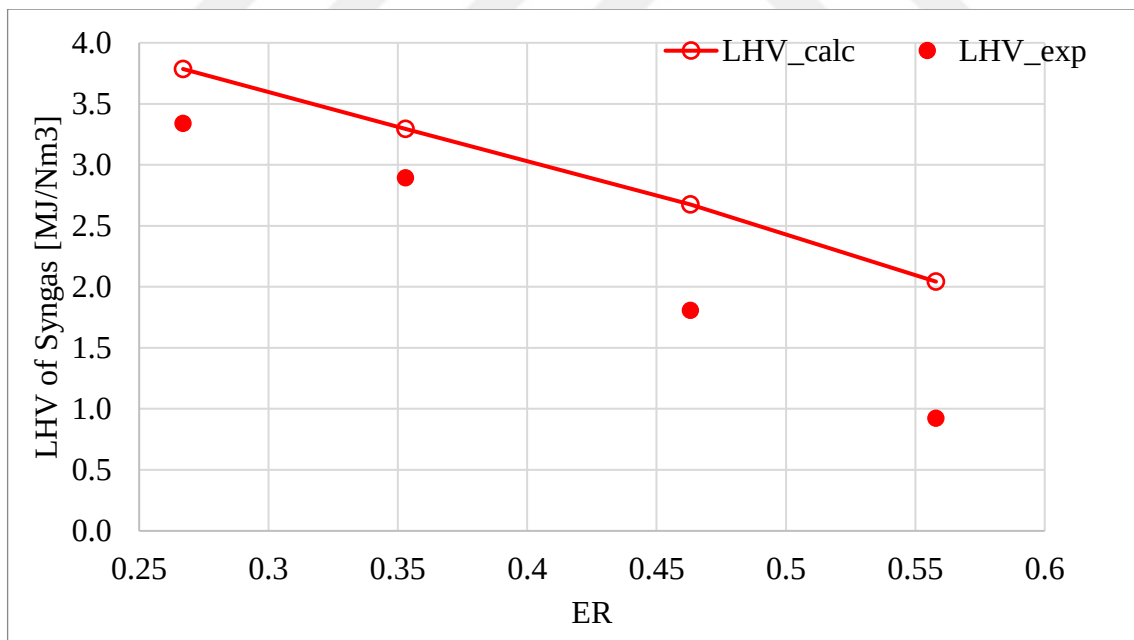


**Figure 3.17** Comparison of experimental and numerical results for LHV of syngas output

### 3.6.1. Effect of Thermal Boundary Condition of Reactor Walls

The experimental setup involves a PID temperature controller which tries to keep the wall temperature of the reactor constant. However, there is a variable temperature distribution through the reactor axis in actual operation. Since the combustion reactions of volatiles and char particles occur in the bed region, higher temperature values are expected at the bottom of the gasifier. Additionally, coal is fed with nitrogen into the reactor through a pipe at a 200 mm height from the bottom of the reactor at atmospheric temperature levels. Hence, a temperature decrease occurs in the feeding region. Temperature distributions on the reactor axis obtained from numerical analyses are given in Figure 3.18 for each ER case.

According to the computational results, while a temperature increase occurs in the bed region due to the combustion reactions, a sharp decrease in temperature value is observed at the fuel feeding level. Moreover, reactor temperatures of ER5 and ER4 cases are lower than the operating temperature, especially in the free-stream region. Hence, it can be said that furnaces are activated by the PID controller in order to reach the operating temperature for these cases.

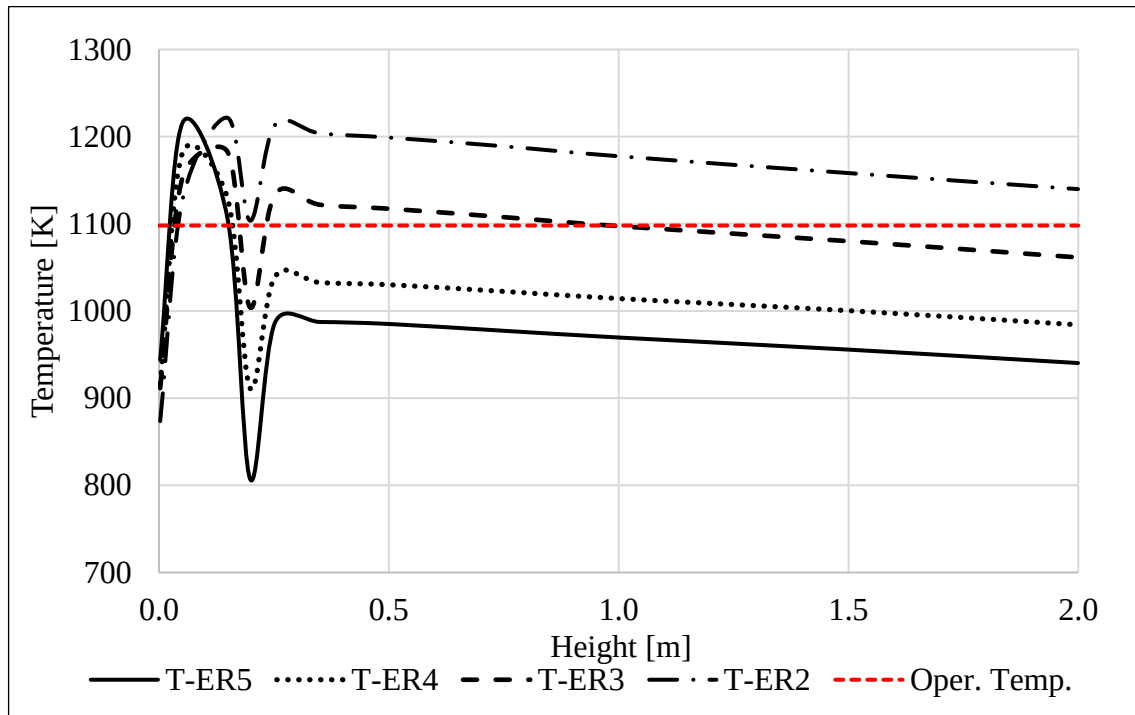


**Figure 3.18** Time-averaged temperature distribution on the vertical axis of the reactor for different ER values

Constant temperature boundary condition is used for water-cooled reactor walls in some of the numerical studies on the gasification process of coal and biomass in the literature [102]. Nevertheless, this approach can not be applied to the case studied in the scope of this thesis. In the case of usage of constant thermal boundary condition, there will be a very high heat loss from the bed to the reactor walls.

### **3.6.2. Effect of Methane Reforming Reaction on Syngas Composition**

Since a considerable difference in  $H_2$  and CO molar concentrations between numerical results and experimental data for higher ER values is observed, it is needed to develop an alternative approach. By evaluating the numerical results in Figure 3.13 and Figure 3.14, it can be easily seen that most of the hydrogen amount is formed in the free-stream region where the molar concentration of methane decreases for these cases. This indicates the dominant effect of the kinetics of methane reforming reaction. It's decided that the effect of methane reforming reaction on the gasification process should be investigated separately. In accordance with this purpose, all ER cases are calculated again by excluding the methane reforming reaction from the numerical setup.

The effect of excluding methane reforming reaction on syngas composition is shown in Figure 3.19 in comparison with experimental data. A very good agreement between numerical and experimental results is obtained for all molar concentration values of gaseous species in syngas for ER2 and ER3 cases. However, significant differences are observed for lower ER values, especially in molar concentrations of  $H_2$ ,  $CH_4$  and CO. Thus, it can be commented that the kinetics of methane reforming reaction has a determinant effect on syngas composition. Advanced and detailed chemical kinetics studies for this reaction including other physical parameters such as temperature are needed in order to obtain consistent results for different ER cases in a gasification process. Moreover, it may be concluded that different reaction kinetics may be needed for different fuel-oxidizer mixing compositions.

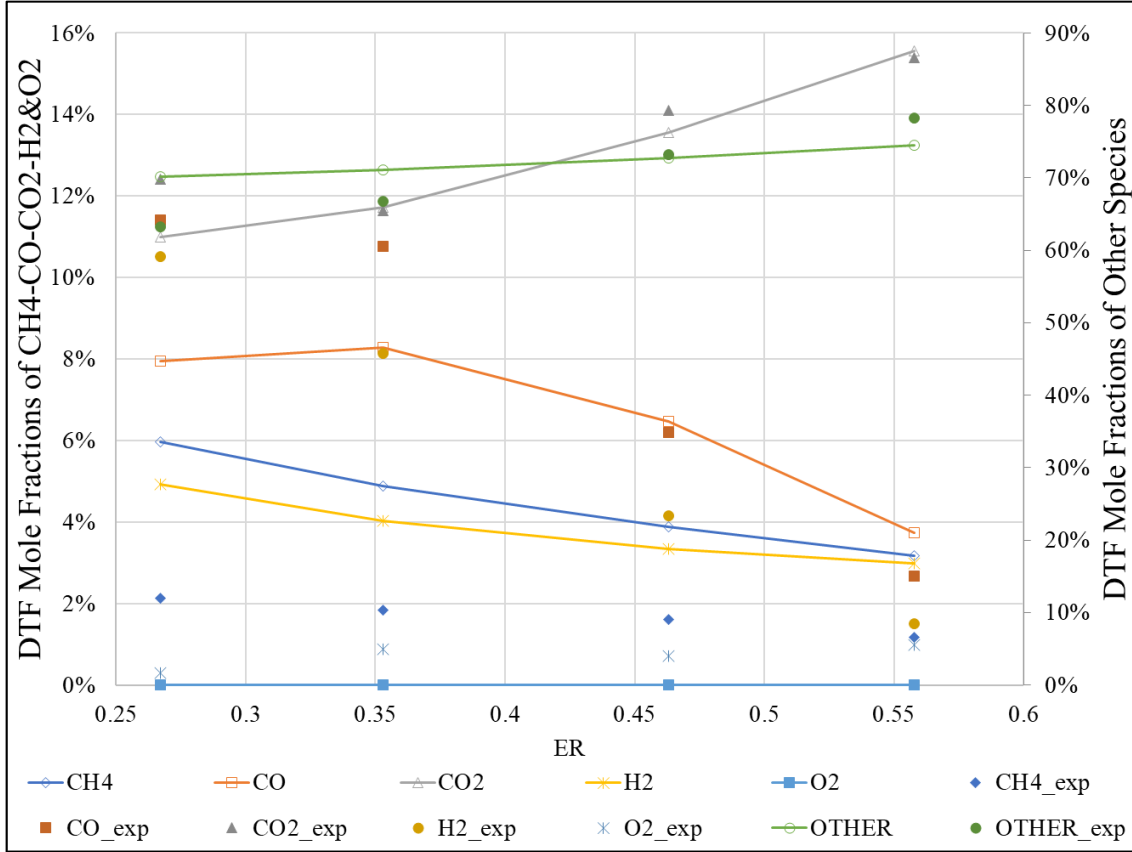


**Figure 3.19** Comparison of numerical and experimental concentration of gaseous species in the syngas output of gasification process of Sample-3 for the case of excluding methane reforming reaction during computations

### 3.6.3. Effect of Devolatilization Model on Syngas Composition

Numerical analysis with CPD model is conducted as an alternative approach to simulate the devolatilization process during the gasification process of Sample-3. ER4 condition for this sample is chosen as a reference case in order to compare the numerical and experimental results and evaluate the effect of the devolatilization model on syngas composition.

Time-dependent syngas composition at the reactor exit is shown in Figure 3.20 in terms of dry and tar-free volumetric fraction of each gaseous species. It is found that volumetric concentration values of all species reach convergence to a steady-state regime at the end of a physical time of 25 s. In order to calculate time-averaged results for syngas composition transient data between 25s and 30s is taken into account.

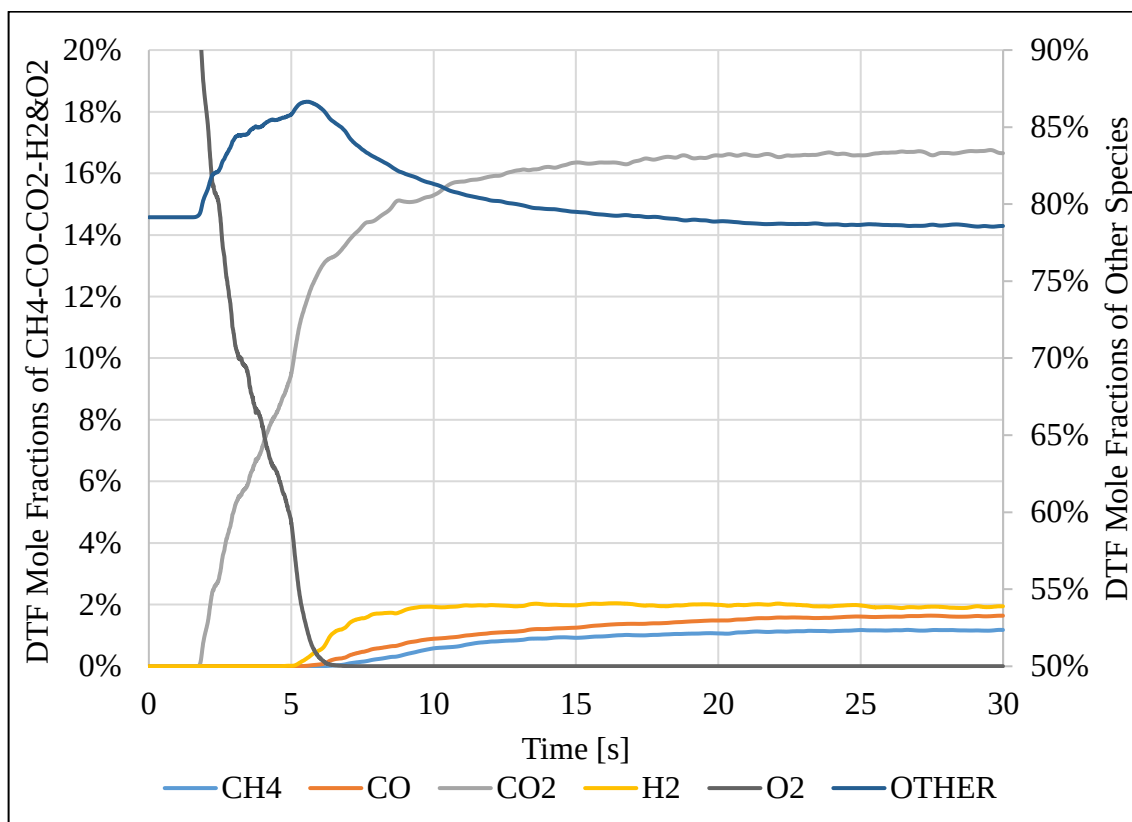


**Figure 3.20** Transient DTF mole fractions of the species at syngas output calculated from numerical results by using CPD model

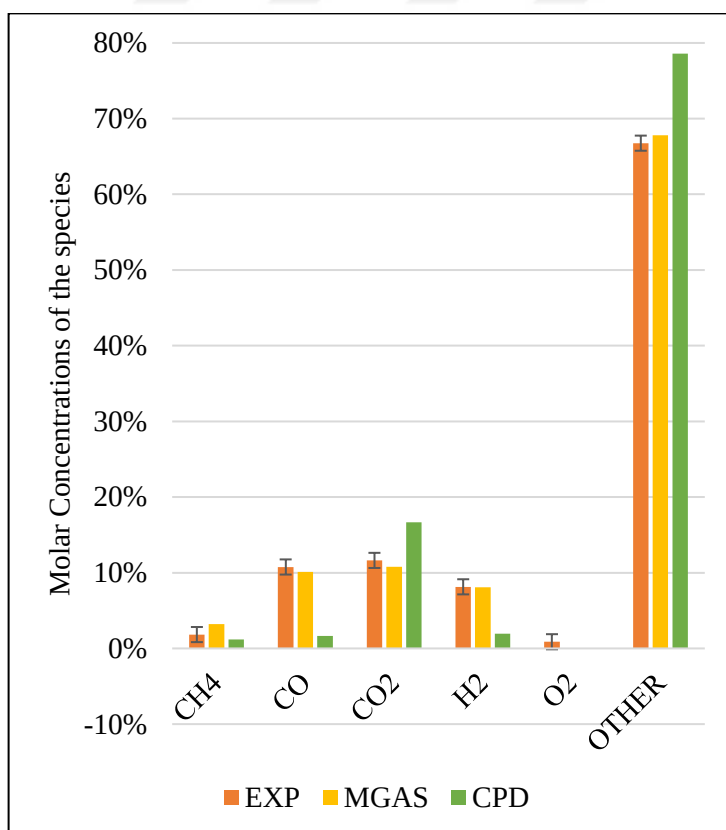


**Figure 3.21** Comparison of different devolatilization models – Syngas Composition

The effect of the devolatilization model on syngas composition predicted by time-averaged numerical data in the steady-state regime and comparison with experimental results are shown in Figure 3.21. According to the comparison with experimental results, there is an overestimation observed in molar concentration of  $\text{CH}_4$ ,  $\text{CO}$  and  $\text{H}_2$  by using MGAS model, while very low values are obtained for these species from the computational outputs of CPD model. Hence, these results yield higher numerical LHV values for syngas output calculated by MGAS model. Furthermore, MGAS devolatilization model predicts closer results than CPD model for all species concentrations except for methane.

#### **3.6.4. Effect of Initial Bed Height on Syngas Composition**

As described in Chapter 2, the bottom region of the lab-scale gasifier is filled with silica sand having  $450\mu\text{m}$  mean particle diameter in order to supply an initial static bed structure in the experiments. The initial static bed height is 150mm for all cases. This bed is heated up only by furnaces until reactor temperature reaches  $500^\circ\text{C}$ . Then, the coal is started to feed into the reactor to initiate the reactive mode. It is needed a couple of hours to reach operating temperature. This yields an increase in bed height due to the accumulation of ash particles in the bed. However, the present experimental setup is not capable of measuring the dynamic height of the bed. Hence, the effect of initial bed height is examined numerically in this study.

Three cases involving different initial bed height values (150mm, 300mm and 450mm) are evaluated in order to understand the effect of the initial amount of sand particles. While initial bed height varies, all other parameters such as fuel feeding coordinates, ER value, etc. used in the numerical setup are kept constant for these cases. Sample-3/ER2 case is chosen for numerical analyses conducted for the investigation of the effect of initial bed height.

Transient molar concentrations of  $\text{CO}_2$  and  $\text{H}_2$  obtained from 3 cases with different initial bed heights can be seen in Figure 3.22 and Figure 3.23, respectively. According to the numerical results, it can be said that the molar concentration of carbon monoxide is not

affected by the change in initial bed height while it has a considerable effect on the molar concentration of hydrogen. While 150 mm initial bed height value yields approximately 8%  $H_2$  concentration in the steady-state region, it is approximately 6% for 300mm and 450mm cases. The molar concentration of  $CO_2$  almost remained constant for all cases around 15%. In the case of 150mm initial bed, the coal feeding level is higher than the fluidized bed height. However, increasing initial bed height from 150 mm to 300 or 450 mm causes a shift in coal feeding level from the upper region to the inner regions of the fluidized bed, relatively. This also causes the motion of char particles at higher bed levels. In higher initial bed cases, oxygen can also reach higher levels and can react with more volatile matter such as methane, hydrogen, etc. Hence, the decrease in molar concentration of  $H_2$  can be explained by increasing oxygen concentrations with height for 300mm and 450mm initial bed height cases.

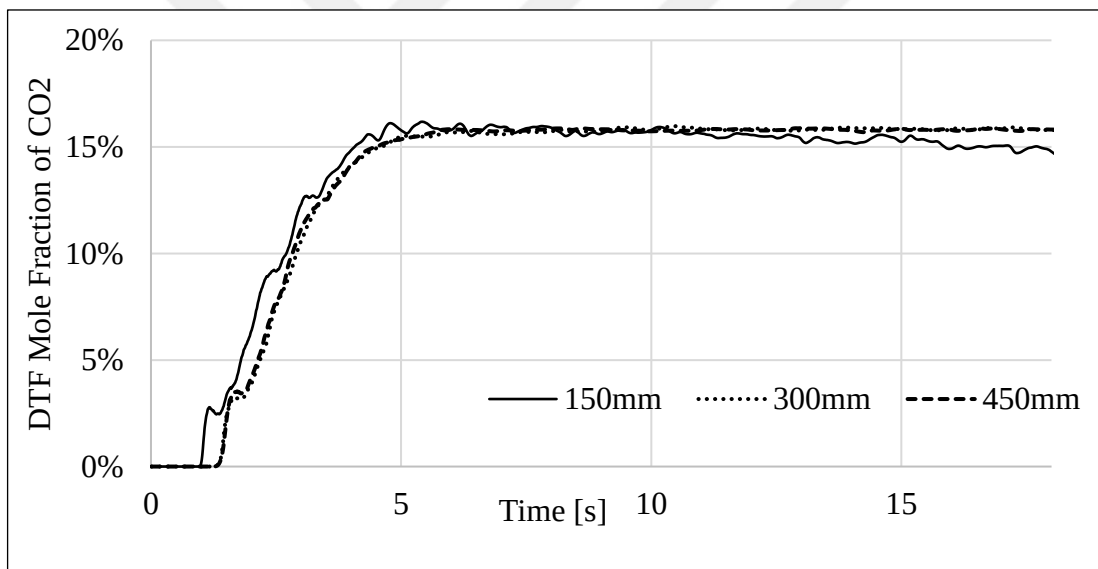


**Figure 3.22** Transient DTF mole fractions of  $CO_2$  for different initial bed height cases

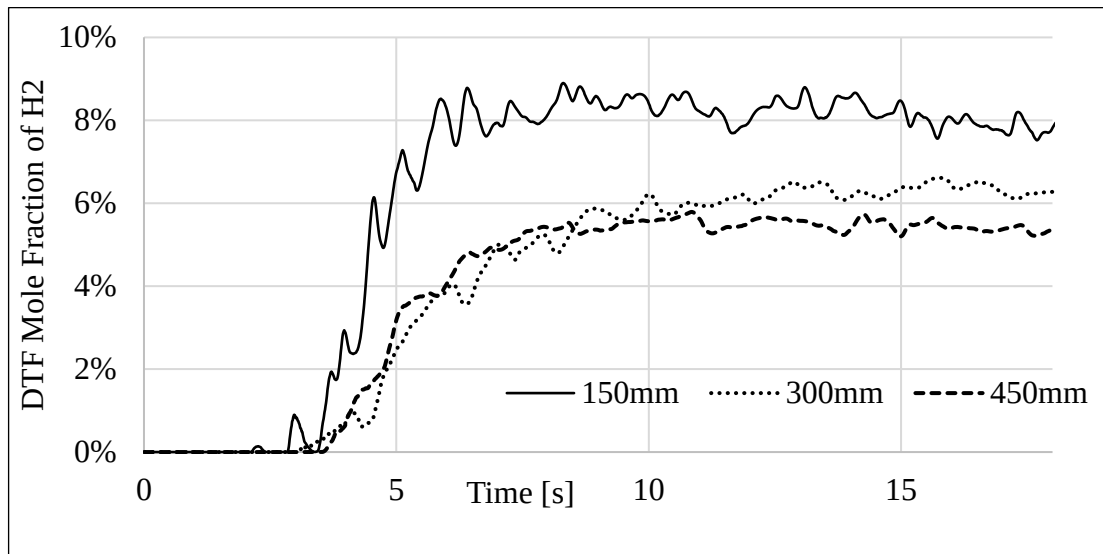


**Figure 3.23** Transient DTF mole fractions of H<sub>2</sub> for different initial bed height cases

### 3.6.5. Thermal Efficiency of the Gasification Process

The thermal efficiency of the gasification process is calculated by using both LHV values obtained from the results of numerical and experimental studies. Process efficiency is obtained in terms of cold gas efficiency and hot gas efficiency by using equation 2.4 and equation 2.5, respectively.

There is not any ash removal process in the experimental procedure. Additionally, the system has a bubbling bed structure with lower superficial velocities. Hence, there is no unreacted char particle content expected in fly ash. Consequently, it can be said that char conversion efficiency of 100% in the experiments and the mass flow rate of syngas obtained from numerical results can be used for the calculations of thermal efficiency of the experiments. However, temperature values are taken constant and equal to the operating temperature (1100K) to calculate the efficiency values as measured during experiments.

Calculated cold gas efficiency and hot gas efficiency values of the gasification process of Sample-3 for different ER cases are shown in Figure 3.24 and Figure 3.25, respectively. An agreement is observed between experimental and numerical results, especially for lower ER values. The highest hot and cold gas efficiencies are observed ER value of around 0.45 however it is predicted about ER=0.35 in experiments. The difference observed between numerical and experimental results for ER3 and ER2 is consistent with the results obtained for LHV values.

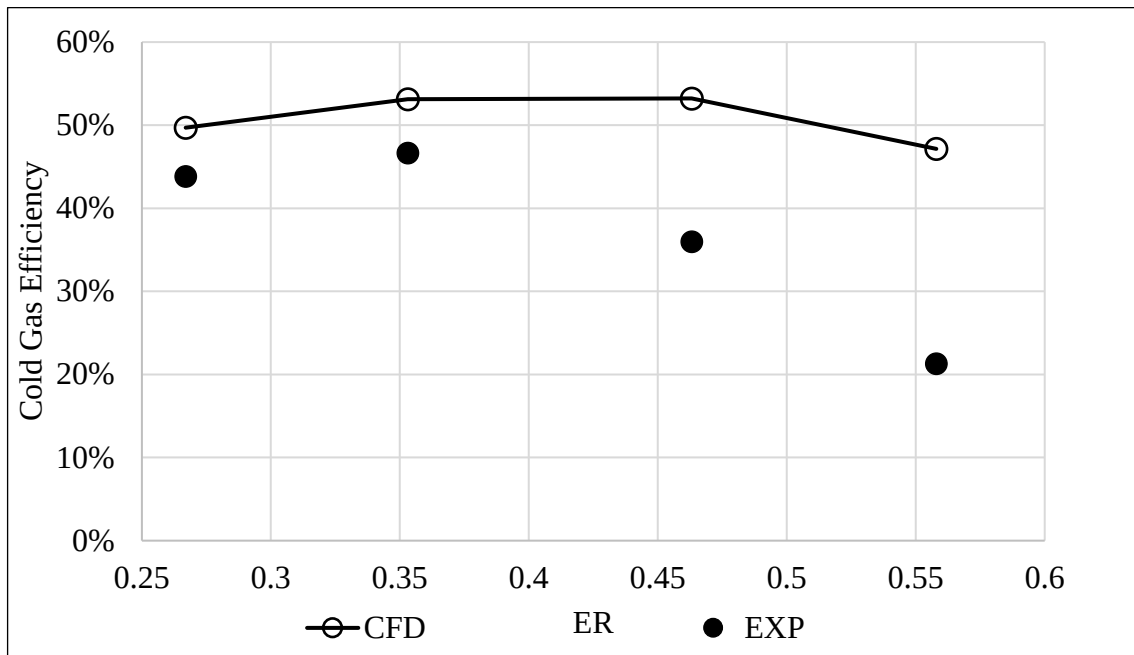


**Figure 3.24** Cold gas efficiency of the gasification process of Sample-3

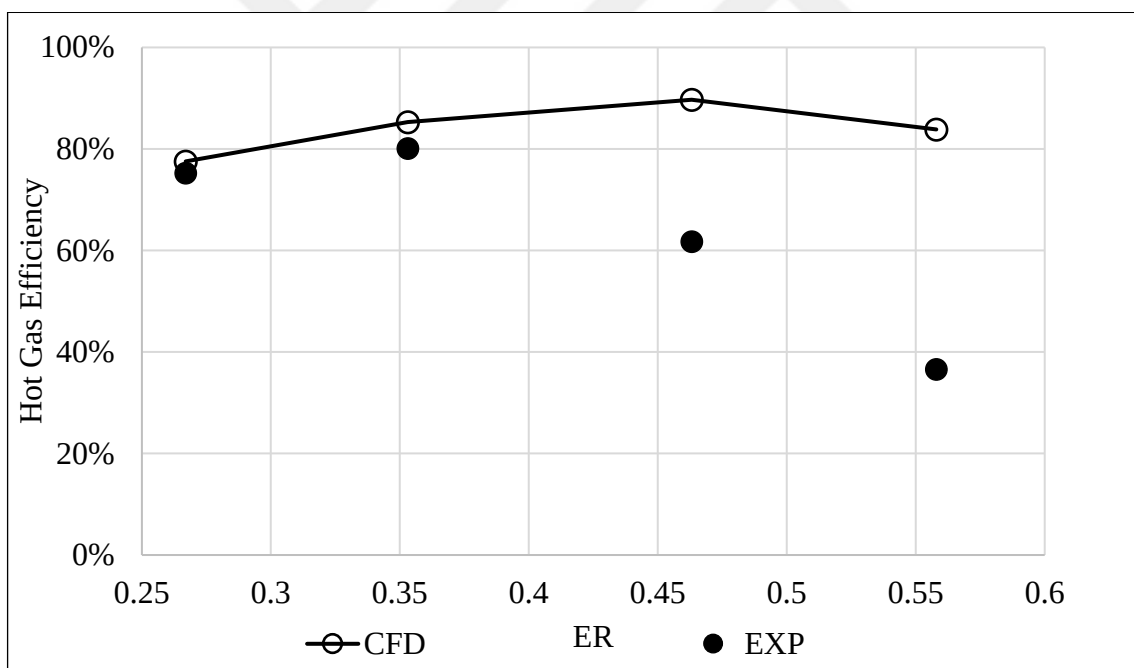


**Figure 3.25** Hot gas efficiency of the gasification process of Sample-3

## **4. DESIGN OF 10 MW<sub>th</sub> INPUT CFBG AND NUMERICAL ANALYSIS OF CFB HYDRODYNAMICS**

In the second part of the thesis study, design of a 10 MW<sub>th</sub> CFB gasification system is carried out for scale-up purposes. Firstly, the design details will be explained. Then the isothermal simulation of the designed system is presented.

### **4.1. Industrial Scale CFBG Design**

#### **4.1.1. Operating Conditions**

Thermal input of 10 MW, 900°C operating temperature, maximum superficial velocity of 6 m/s, usage of coal Sample-3, particle size distribution and ER value of 0.4 are the main inputs of the design of industrial-scale CFBG. By using these parameters and coal characteristics, actual and stoichiometric air flow rates, syngas output parameters, inputs for ash removal and sulfur capture processes are calculated.

In the design of industrial-scale gasifier, it is assumed that a start-up burner should supply a thermal power of approximately 10% of the total system thermal input. Design parameters and operating conditions of the CFBG system are given in Table 4.1.

An air pre-heater is used to increase the inlet temperature of the gasification agent by approximately 100°C due to the low-rank properties of chosen coal samples and to sustain steady-state operating temperature levels during the autothermal process.

Mass flow rate of the syngas is calculated by considering the experimental and numerical results and an assumption of 95% carbon conversion ratio. Five percent of unreacted carbon is removed with bottom ash from the reactor. Carbon conversion rate is an indicative parameter of a clean and efficient energy conversion system and should be maximized for all CFB reactors.

Most of the ash produced in a gasification process exits the reactor in a fly ash form in a circulating fluidized. It is assumed that the mass fraction of the fly ash is 80% of the total

ash produced in the reactor. A small amount of total ash called bottom ash remains in the bed region. Bottom ash passes through a conical section and ash removal pipe at the bottom of the reactor. A water-cooled screw removes the bottom ash from the system.

**Table 4.1** Design parameters and operating conditions of industrial-scale CFBG

| Properties                              | Symbol              | Value    | Unit                       |
|-----------------------------------------|---------------------|----------|----------------------------|
| Reactor nominal thermal power input     | $P_{R,th}$          | 10       | MW <sub>th</sub>           |
| Calculated Coal MFR                     | $\dot{m}_{c,calc}$  | 2244     | kg/h                       |
| Actual Coal MFR (max)                   | $\dot{m}_{c,act}$   | 2850     | kg/h                       |
| O <sub>2</sub> needed for Stoichiometry | $m_{O_2,sto}$       | 1.331    | kg-O <sub>2</sub> /kg-coal |
| Air needed for Stoichiometry            | $m_{a,sto}$         | 5.714    | kg-air/kg-coal             |
| Equivalence Ratio                       | $ER$                | 0.4      | -                          |
| Gasification air per unit mass of fuel  | $m_a$               | 2.286    | kg-air/kg-coal             |
| Total Air MFR                           | $\dot{m}_a$         | 6514     | kg/h                       |
| Total Air VFR                           | $\dot{V}_a$         | 5109     | Nm <sup>3</sup> /h         |
| Reactor bed design temperature          | $T_R$               | 900      | °C                         |
| Operating pressure                      | $p_R$               | 1.05     | bar                        |
| Air inlet temperature                   | $T_a$               | 130      | °C                         |
| Loop-seal air inlet temperature         | $T_{a,LS}$          | 20       | °C                         |
| Syngas density                          | $\rho_g$            | 0.311    | kg/m <sup>3</sup>          |
| Particle density                        | $\rho_p$            | 1400     | kg/m <sup>3</sup>          |
| Syngas viscosity                        | $\mu_g$             | 4.82E-05 | Pa.s                       |
| Maximum superficial velocity            | $U_{max}$           | 6        | m/s                        |
| MFR of syngas                           | $\dot{m}_g$         | 8992     | kg/h                       |
| VFR of syngas                           | $\dot{V}_g$         | 28914    | m <sup>3</sup> /h          |
| Total ash production rate               | $\dot{m}_{Ash}$     | 657      | kg/h                       |
| Bottom ash output                       | $\dot{m}_{Ash,bot}$ | 131      | kg/h                       |
| Fly ash output                          | $\dot{m}_{Ash,fly}$ | 525      | kg/h                       |
| Fly-ash in syngas                       | $f_{Ash,g}$         | 18.2     | g-ash/m <sup>3</sup>       |
| Coal particle size range                | $d_p$               | 1-3      | mm                         |
| Silica sand mean particle diameter      | $\bar{d}_{Si}$      | 0.5      | mm                         |
| Start-up burner thermal power           | $p_{B,th}$          | 1.1      | MW <sub>th</sub>           |
| Start-up gas temperature                | $T_{SU,g}$          | 700      | °C                         |
| Start-to-feed reactor temperature       | $T_{SU,feed}$       | 600      | °C                         |
| Limestone mean particle diameter        | $\bar{d}_{Ca}$      | 1        | mm                         |
| MFR of limestone                        | $\dot{m}_{Ca}$      | 285      | kg/h                       |

By aiming to minimize the sulfur content of the syngas, limestone feeding method is used to capture  $\text{H}_2\text{S}$ ,  $\text{SO}_x$  and  $\text{COS}$  species in the fluidized bed region. Limestone is mostly composed  $\text{CaCO}_3$  mineral which is a very effective and low-cost desulfurization method. The mass flow rate of the limestone is obtained according to the ultimate analysis of chosen coal sample.

Design parameters and dimensioning of CFBG system components and assembly technical drawing of these components are given in the following sub-sections.

#### **4.1.2. Dimensioning of The Reactor**

Inner diameter of the reactor is the first dimension to be determined during design calculations. Inner diameter of a CFB reactor can be calculated by known fuel, thermal input, ER, and superficial velocity values. Although various analytical approaches exist in order to determine dynamic bed height in both circulating and bubbling fluidized bed systems, there is not any calculation method available for reactor heights. However, height of the free-stream (free-board) region is taken between 1-2 m for bubbling fluidized bed systems, and it varies between 10-30m for circulating fluidized bed systems in practice [12]. In this design, total reactor height is determined by 17.5 m by considering superficial velocity value, residence time of coal particles obtained from numerical calculations and this range given in the literature.

Coal and limestone feeding levels are determined by considering static and dynamic bed height values. Feeding channels with  $45^\circ$  slope angle in the vertical direction are located slightly above the fluidized bed region. The channel between loop-seal and reactor is also located on the same level as shown in Table 4.2.

Section of syngas outlet channel is determined by the design calculations of the cyclone located on the upper regions of the reactor. Coal and limestone feeding channel diameters are obtained by using the calculated mass flow rates from thermal input and maximum particle velocity values.

**Table 4.2** Main inner dimensions of the gasification reactor [mm]

| Dimension                                      | Symbol | Value    | Geometry |
|------------------------------------------------|--------|----------|----------|
| Inner Diameter                                 | D1     | 1300     |          |
| Total Height                                   | H1     | 17500    |          |
| Bottom Diameter                                | D2     | 1300     |          |
| Static Bed Height                              | H3     | 900      |          |
| Fuel, Bed Material Feeding and Recycling Level | H5     | 1700     |          |
| Secondary Start-up Burner Level                | H6     | 2100     |          |
| Coal Feeding Channel Diameter                  | D3     | 200      |          |
| Recycling Channel Diameter                     | D4     | 300      |          |
| Channel Diameter for Secondary Start-up Burner | D5     | 200      |          |
| Bed Material Feeding Channel Diameter          | D6     | 100      |          |
| Sparger Pipe Diameter                          | D7     | 125      |          |
| Ash Removal Pipe Diameter                      | D8     | 150      |          |
| Syngas Exit Channel Height                     | H2     | 16500    |          |
| Syngas Exit Channel Cross Section              | S1     | 730 x730 |          |
| Syngas Exit Channel Slope                      | A1     | 77°      |          |

Three-stage insulation is applied for the reactor, cyclone, stand-pipe and loop-seal components in the design. An inner refractory layer (Hycast 85) with 100mm thickness which has high corrosion and high temperature resistance properties is used. This layer is directly in contact with the solid particles and gaseous products in the reactor. Next layer is an insulating refractory layer (Hyisolate 110) with 130 mm thickness and has a

relatively lower thermal conductivity value when compared to the inner layer. A ceramic wool layer with 100mm thickness is preferred as outer insulation material. Ceramic wool is a very low-thermal conductive material and is commonly preferred for commercial and academic CFB and BFB applications due to its relatively low costs. All layers are fixed and assembled on an outer steel shell structure via anchors and anchor bricks in the design. By using one-dimensional heat transfer calculations outer shell temperature is calculated to be 63°C for steady-state operating conditions (Appendix-D).



#### 4.1.3. Cyclone Design

The dense particulate gas flow exit from the gasification reactor is directed to the recirculation cyclone in order to separate relatively large-diameter particles including unreacted carbon and feed them back into the reactor. Mechanically captured particles in the cyclone are transferred to the loop-seal through a stand-pipe installed between cyclone and loop-seal components.

There are various types of cyclone designs for high temperature CFB applications available in the literature [104]. While some of those cyclone designs are used for high particulate flow rate applications, some of them have high-efficiency particulate matter capturing capabilities in the case of low particulate density flows. In the present design, a modified Lapple type cyclone model is used as a general-purpose approach which yields moderate levels for both separation efficiency and particulate matter capturing rate.

Theoretical cut-off size and 50% cut-off size are two critical parameters for the design of a cyclone. Theoretical cut-off size is an indicator that a cyclone is capable to separate 100% of the solid particles that have a larger diameter than this value. 50% cut-off size yields 50% separation efficiency at a given particle size. For lower particle diameters, lower separation efficiency values are seen. Dewil et al. presented some alternative analytical approaches in order to determine these parameters [104]. Calculated theoretical and 50% cut-off size values are given in Table 4.3.

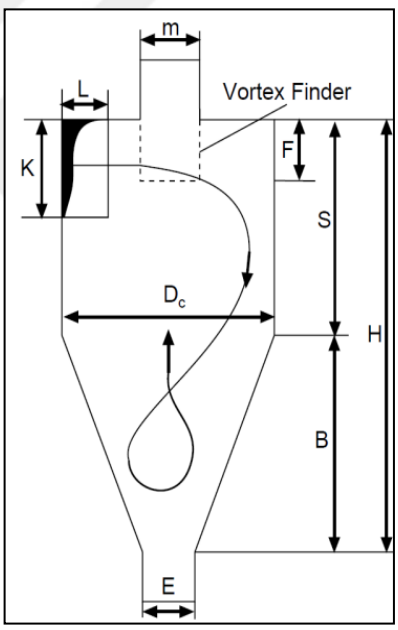
Inlet sectional area is calculated by assuming the maximum inlet gas velocity is 15 m/s in the inlet channel. Although a standard Lapple type cyclone involves a rectangular section ( $K/L=2$ ), it is preferred to use a square section in the modified geometry as shown in Table 4.3 in order to ease the application of the refractory liners and other insulation layers at the inlet region.

The channel connecting the cyclone and the reactor was considered horizontal in the preliminary design studies. As a result of the studies on the manufacturing details, it is decided that this channel should be inclined, considering that the horizontal nature of this channel may cause particle accumulation in the channel for long operating periods and

clogging of the channel. The channel is designed to make an angle of  $17^\circ$  with the horizontal plane.

Outer dimensions of the cyclone are determined by the insulation design given in the previous sub-section. All inner dimensions and design parameters of the cyclone can be seen in Table 4.3.

**Table 4.3** Design parameters and dimensions of the cyclone

| Property               | Symbol              | Value    | Unit                   | Geometry                                                                            |
|------------------------|---------------------|----------|------------------------|-------------------------------------------------------------------------------------|
| VFR of the gas         | $\dot{V}_g$         | 28914    | $\text{m}^3/\text{h}$  |  |
| Cyclone inlet velocity | $U_{\text{exit}}$   | 15       | m/s                    |                                                                                     |
| Inner Dimensions       | $D_c$               | 1.610    | m                      |                                                                                     |
|                        | K                   | 0.730    | m                      |                                                                                     |
|                        | L                   | 0.730    | m                      |                                                                                     |
|                        | M                   | 0.810    | m                      |                                                                                     |
|                        | F                   | 1.010    | m                      |                                                                                     |
|                        | S                   | 3.220    | m                      |                                                                                     |
|                        | H                   | 6.440    | m                      |                                                                                     |
|                        | E                   | 0.400    | m                      |                                                                                     |
| Gas density            | $\rho_g$            | 0.311    | $\text{kg}/\text{m}^3$ |                                                                                     |
| Gas dynamic viscosity  | $\mu_g$             | 4.82E-05 | Pa.s                   |                                                                                     |
| Particle density       | $\rho_p$            | 1400     | $\text{kg}/\text{m}^3$ |                                                                                     |
| Centrifugal force      | $F_{\text{cent}}$   | 5.69E-07 | N                      |                                                                                     |
| Spiral #               | $N_{\text{spiral}}$ | 4.8      | -                      |                                                                                     |
| Theoretical cut-off    | $d_{p,\text{th}}$   | 100      | $\mu\text{m}$          |                                                                                     |
| 50% cut-off diameter   | $d_{50}$            | 18       | $\mu\text{m}$          |                                                                                     |

#### 4.1.4. Loop-seal Design

The operating conditions of the LS are determined in the scope of the design phase. Detailed calculations and dimensioning studies are conducted in order to control the fluidized bed structure in this component. The operating conditions of the LS are given in Table 4.4.

**Table 4.4** Design parameters of the loop-seal

| Property                              | Symbol                         | Value     | Unit                |
|---------------------------------------|--------------------------------|-----------|---------------------|
| Void fraction                         | $\epsilon_g$                   | 0.5       | -                   |
| Standpipe particle velocity           | $U_{SP}$                       | 0.1       | m/s                 |
| Particle density                      | $\rho_p$                       | 1400      | kg/m <sup>3</sup>   |
| Standpipe diameter                    | $d_{SP}$                       | 0.4       | m                   |
| Analytical particle SCR per unit area | $W_p$                          | 70        | kg/m <sup>2</sup> s |
| Analytical particle SCR               | $G_p$                          | 8.8       | kg/s                |
| Horizontal particle velocity in LS    | $U_h$                          | 0.15      | m/s                 |
| Gas density                           | $\rho_g$                       | 0.311     | kg/m <sup>3</sup>   |
| Air feeding temperature in to LS      | $T_{a,LS}$                     | 20        | °C                  |
| Air density                           | $\rho_{a,LS}$                  | 1.575     | kg/m <sup>3</sup>   |
| Fluidization velocity                 | $U_{LS}$                       | 0.2-0.7   | m/s                 |
| VFR of the gas in LS                  | $\dot{V}_{LS}$                 | 0.2-0.35  | m <sup>3</sup> /s   |
| MFR of the gas in LS                  | $\dot{m}_{LS}$                 | 224-392   | kg/h                |
| Air Flow Rate ratio of LS to total    | $\dot{m}_{LS}/\dot{m}_{a,act}$ | 3.44-6.02 | %                   |

A three-dimensional model is developed for the loop-seal component and it is examined whether there would be any problems in terms of manufacturing. Previously, it was thought that an inclination angle of 60 degrees for the return leg channel would facilitate and accelerate the solid circulation rate. However, in the examinations made on the three-dimensional model, it was seen that the 60-degree slope would cause some problems in terms of manufacturing and it was concluded that the 45-degree slope would be appropriate. Figure 4.1 shows 3D geometric model and technical drawing created for the loop-seal.

During research and design studies, it was noticed that there is not any detailed study available in the literature on the operating conditions of the loop-seal component and the

effects of the aeration rates on the solid recirculation rate of a CFB system. This topic is another focus of this chapter evaluated in the following sections.

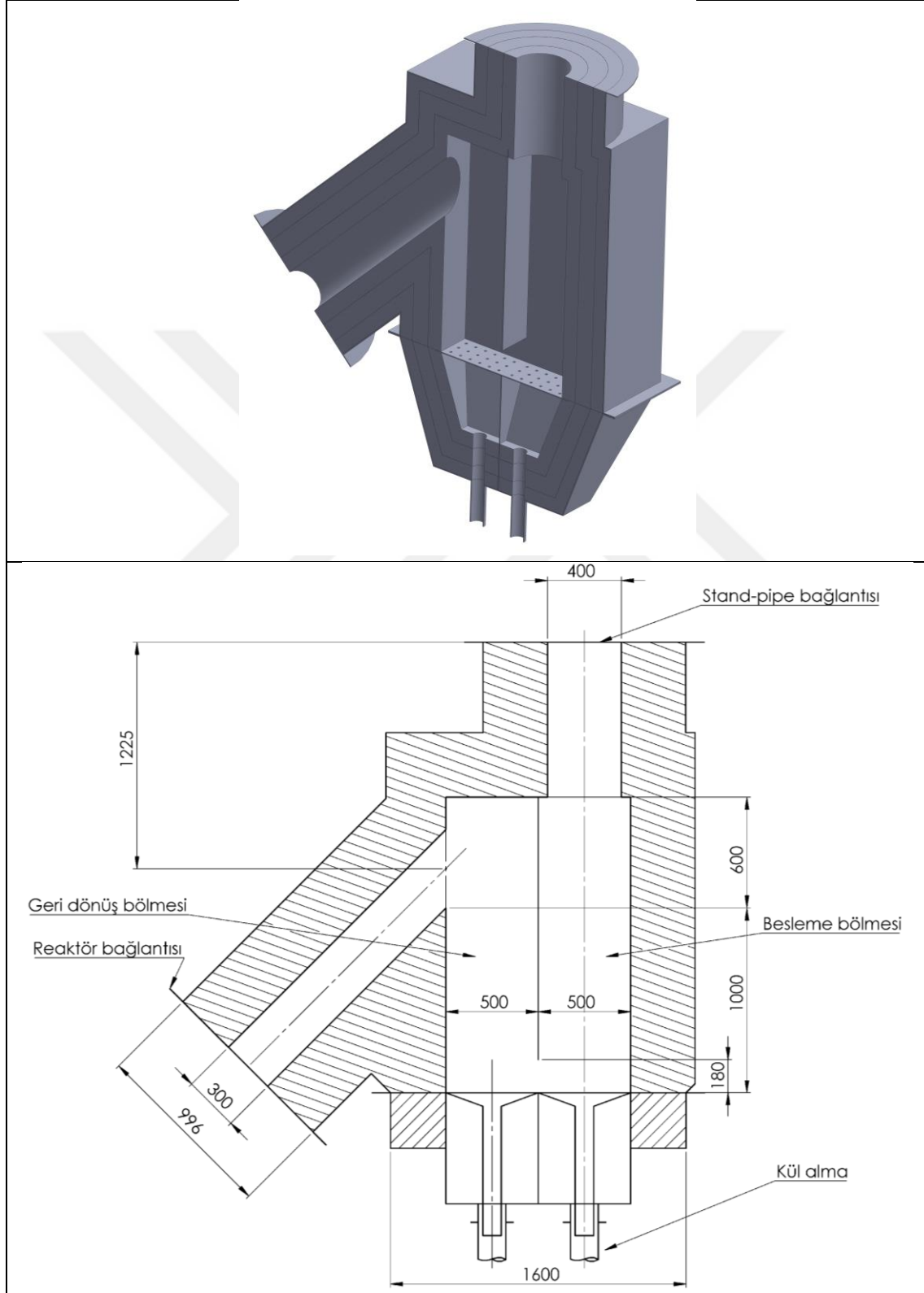


**Figure 4.1** 3D geometrical model and technical drawing of the loop-seal

#### 4.1.5. Start-up Burner and Combustion Chamber Design

Along with the studies carried out during design, it is determined that the minimum thermal capacity of the start-up burner should be 1 MW in order to increase the bed temperature above 600°C during the start-up process of the CFBG system. A diesel-fuel burner is chosen and appropriate fuel tank capacity, feeding systems and safety equipment are determined by considering thermal capacity, operating periods and fuel type.

The reactor operates in a bubbling fluidized bed regime by the supplied flue gas from start-up burner (superficial velocity in the bed is calculated 1.5 m/s). With the gas at 700°C to be supplied to the system, the temperature of silica sand in the bed is increased from ambient temperature to 600°C, which is the minimum temperature level required for the coal gasification process. According to the calculations, it is predicted that the time required for the system to reach the desired operating fluidized bed temperature is 6 hours.

The start-up burner has the following features:

- Thermal capacity: 1100 kW
- Fuel: Diesel
- Operating pressure: 180 mbar

The gas temperature of 700°C is obtained by mixing the flue gas produced from the burner and the clean air coming from the main forced draft fan in appropriate proportions in a mixing chamber. Since the flue gas produced by the start-up burner and the fresh air in the mixing chamber might cause a cross-flow in a mixing chamber, it is foreseen that the two flows can block each other. According to the final design study, the flue gas flow and air flow are parallel to each other and in the same direction as seen in Figure 4.2. In the mixing chamber located at the end of the combustion chamber, these two flows are mixed with a minimized pressure drop and fed to the reactor.

Supplying fresh air from outside the combustion chamber will both protect the refractory liner and reduce the refractory thickness required for the combustion chamber. Before the start-up process, N<sub>2</sub> gas should be supplied into the system in order to purge the reactor and clean the channels from combustible gaseous products and solid particles. This

Technical drawing of a furnace assembly showing a cross-section of the furnace body, a burner, and a gas connection. The drawing includes dimensions for the furnace body (1850x1400), the burner (1300x1450), and the gas connection (1500x1350). The furnace body is made of Hycast 85 and Hycast 110. The burner is made of Brülör. The gas connection is made of Sparger bağlantısı. The drawing also shows the air inlet (Hava girişi) and the burner (Brülör).

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#### **4.1.6. Air Feeding System Design**

The air feeding system is one of the most important components of a CFB system which significantly affects the fluidization characteristics, char conversion rates and auxiliary equipment properties. The appropriate air feeding system yields lower pressure drop and homogenous air supply into the reactor.

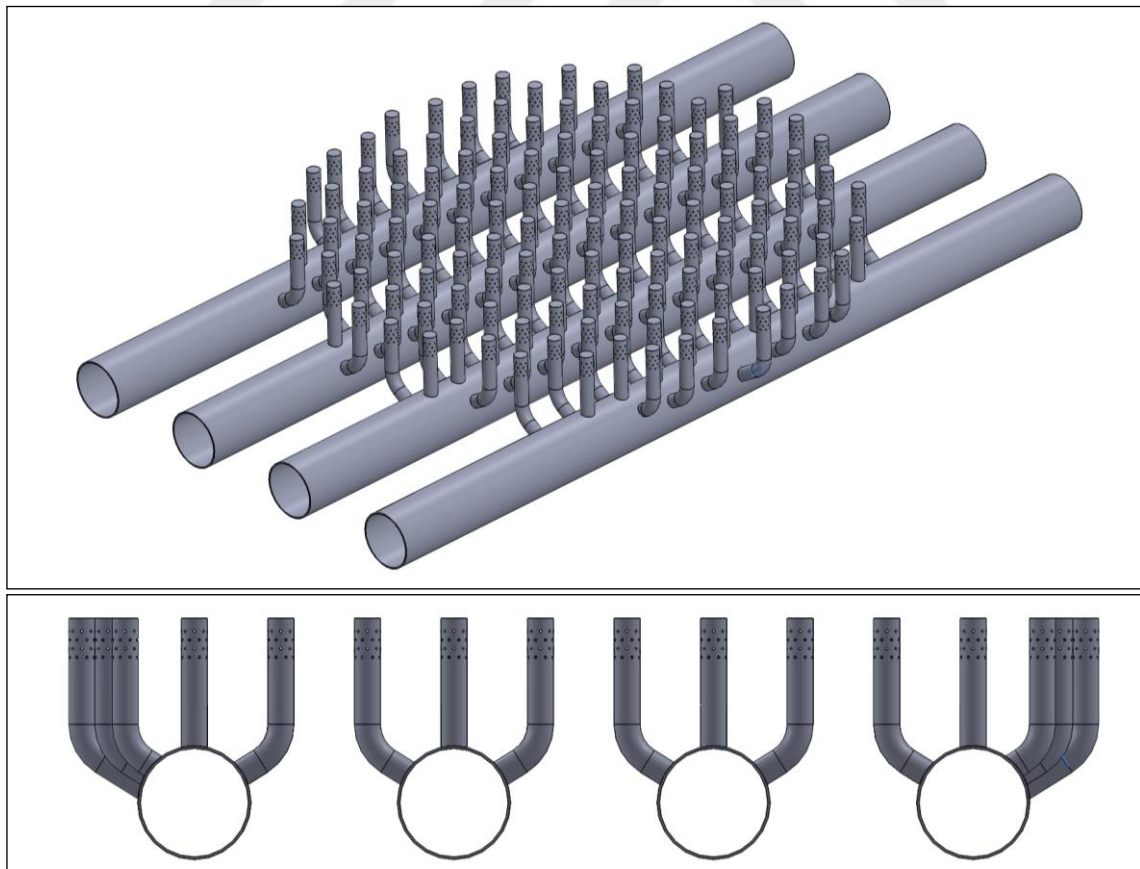
Various types of air feeding systems can be found in the literature. However, these systems can be grouped into two design types: Distributor plate and Sparge pipe designs. In the preliminary design, the main air supply to the reactor by using distributor plate including bubble-caps is used. However, as a result of the design studies, it is concluded that sparge-pipe option is more appropriate solution for the present design by considering the total weight of ash and solid material in the fluidized bed and dynamic bed height. In addition, it is thought that the thermal stresses might cause very high displacement and considerable change in the shape of distributor plate surface. Thermal expansion effects are minimized by the usage of sparger-type air feeding system.

During design calculations, 40 m/s maximum air velocity is considered in the nozzle feeding pipes in the reactor. Maximum air exit velocity from the nozzle holes is taken 70 m/s as a practical value.

The design parameters of the air feeding system are shown in Table 4.5. 3D geometrical model of the sparger feeding system is given in Figure 4.3.

**Table 4.5** Design parameters of the sparger type air feeding system

| Property                              | Symbol            | Value    | Unit                   |
|---------------------------------------|-------------------|----------|------------------------|
| Total air MFR                         | $\dot{m}_{a,act}$ | 6514     | kg/h                   |
| Air MFR supplied to the sparger       | $\dot{m}_{a,R}$   | 5993     | kg/h                   |
| Reactor inner diameter                | $d_R$             | 1.3      | m                      |
| Air exit velocity from holes          | $U_h$             | 70       | m/s                    |
| Air density at nozzles                | $\rho_N$          | 0.846    | kg/m <sup>3</sup>      |
| Nozzle hole diameter                  | $d_h$             | 3        | mm                     |
| Total exit area                       | $\sum a_h$        | 2.811E-2 | m <sup>2</sup>         |
| Total hole number                     | $\sum N_h$        | 3980     | -                      |
| Holes per nozzle                      | $N_h$             | 32       | 4x8 holes              |
| Number of the nozzles - calculated    | $N_{N,calc}$      | 124      | -                      |
| Number of the nozzles - design        | $N_{N,des}$       | 132      | -                      |
| Hole number per area                  | $\lambda_h$       | 2999     | holes/m <sup>2</sup>   |
| Nozzle number per area                | $\lambda_N$       | 93       | nozzles/m <sup>2</sup> |
| Wall thickness of nozzle              | $t_N$             | 4        | mm                     |
| Height of the hole from feeding level | $h_h$             | 100      | mm                     |
| Nozzle outer diameter                 | $d_{N,out}$       | 30       | mm                     |

**Figure 4.3** 3D geometrical model of the sparger type air feeding system

A technical drawing of the geometrical model of system assembly is created at the end of the design studies. Sectional view of the technical drawing of the CFBG is shown in Figure 4.4. The top view and 3D model of the assembly can be seen in the figures given in Appendix-E.



## 4.2. Background of The Studies About Solid Circulation in a CFB Reactor

A CFB system mainly consists of a riser, a cyclone, a standpipe and a loop-seal. A loop-seal (LS) is a non-mechanical valve, which may be defined as the heart of a CFB system. The main function of LS is to deliver unburnt fuel and other particles captured from the cyclone back to the reactor bed and to provide a circulation loop by using a relatively high-pressure fluidizer. It also provides sealing for the synthetic gas leakage produced in gasification reactors.

Understanding the operation of this device is very difficult due to the complexity of its geometry and the associated physical phenomena. The solid circulation rate (SCR) is a significant factor for the operation of a CFB system. SCR cannot be directly measured on a commercial high temperature system due to operational safety. The SCR of a loop-seal can be controlled by appropriate aeration rate.

Most of the commercial CFB systems have a typical loop-seal with two stages at different aeration rates. In this chapter, design parameters of 10MWth CFB gasifier are presented. Moreover, the effect of the aeration rate on the circulation rate of a two-chamber loop-seal for a gasification system is examined using CPFD method. Pressure losses, pressure fluctuations and particle accumulation in the LS are also investigated numerically. In numerical analysis, Barracuda Virtual Reactor v17 has been used as a commercial software to model the whole CFB system with non-reacting isothermal flow assumption. Pressure fluctuations in LS and the particle height in the standpipe are also examined by processing the simulation results.

Several methods exist in the literature to determine the solid circulation rate (SCR) including experimental, analytical and numerical methods. Direct measurement is the most common method based on recording the time required to fill a known volume with solids after closing the butterfly valve located at the top of the standpipe [107]-[110]. Other measuring methods such as the impact plate [111] and the rotating fiberglass spiral in standpipe [112] are also used for experimental investigations of particle circulation characteristics in a CFB system. There are also analytical models using the pressure data to investigate the SCR. Markström et al. [113] experimentally evaluated the SCR in a

chemical looping combustor (CLC) system. The SCR calculated using the pressure drop measurements in the riser was 78% overestimated compared to the measured solid mass flux. Alghamdi et al. proposed an analytical method based on a kinematic model for the prediction of SCR by calculating the pressure drop at the top of the riser [114]. They have found that the prediction accuracy of SCR is improved by up to 60% compared to conventional analytical models utilizing the riser total pressure drop data.

The experimental techniques mentioned above are expensive. Moreover, they require relatively accurate calibration, systematic maintenance. On the other hand, analytical models give overestimated results for the SCR. There are few numerical studies in the literature for the investigation of the particle behavior in the LS of a CFB system. Particularly, no special work found in the literature on investigating numerically the effects of the operating conditions of the LS component of an industrial scale CFB system. Therefore, there is a lack of literature in both experimental and numerical studies about the operation of LS at industrial scale applications.

Peng et al. examined the SCR of a lab-scale CLC for cold flow conditions with DEM and compared the results with experiments [115]. They have reported that there is a linear relationship between SCR and the air flow rate in the fuel reactor within a certain range. Seo et al. investigated SCR and pressure drop of a dual CFB gasification unit by using Eulerian method and concluded that the SCR has to be maintained over a certain amount for a stable operation of the LS [116]. Li et al. compared the circulation characteristics for riser-only and full-loop simulations of a CFB system using MP-PIC method based on MFIX code [117]. They emphasized that a reasonable estimation of the SCR is a precondition for a riser-only simulation to predict properly the flow characteristics inside the riser. Xueyao et al. applied a modified EMMS based, 2D Eulerian model to calculate hydrodynamic properties in standpipe and L-valve type LS of a CFB and reported the relationship between the flow field, the aeration rate and the change in pressure drop fluctuations for different operating conditions [118]. Wang et al. investigated the effects of LS aeration rate, superficial velocity in the riser and solid particle inventory on SCR using CPFD Barracuda VR code in a single chamber LS and validated the results experimentally [119].

### 4.3. Working principle of a CFB system with Loop-Seal

Circulation loop of a typical CFB reactor consists of a riser, a cyclone, a standpipe, and a loop-seal along with the reactor. The cyclone, standpipe, and LS are the particle recycle system components of a CFB reactor. Solid fuel is usually fed by a screw conveyor into the reactor in the bed region. Air as a reactant is fed to the system by different methods. A common method is to feed air by a nozzle system integrated on a distributor plate. Most of the chemical reactions between the air and solid fuel are completed in the reactor bed and riser regions. However, some particles may exit from the riser of the reactor without completing the whole reaction steps. Unreacted fuel particles and relatively large ash particles captured in the cyclone are redirected to the LS through a standpipe. LS delivers back these particles into the reactor bed with the help of the aeration provided at the bottom of the LS. In addition, it increases the carbon conversion rate and decreases the unreacted particle fraction in the fly ash. As LS is connected to the CFB reactor through a pipe channel (i.e. downcomer), it is used to feedback the solid particles captured in the cyclone to the reactor. However, it provides an alternative flowing path for the unreacted gasses that may escape from the reactor. Therefore, it is required to block such gasses consisting of reactive ones such as  $\text{CH}_4$ ,  $\text{H}_2$ , and  $\text{CO}$ , and it may cause uncontrolled reactions and thereby instability in LS. To summarize, control of the LS is vital for the stable operation of a CFB system.

LS behaves, in fact, as a secondary fluidized bed in a CFB system. In most studies in the literature, the aeration rate value is selected to keep the LS in the bubbling fluidized bed regime. To our knowledge, there is no published research focusing on the working characteristics of an LS system in transition or turbulent regimes. The main objective of this study is to provide this information. Moreover, this work aims to investigate the influence of the fluidization regime on the SCR and the pressure in LS.

The minimum fluidization velocity is an indicative parameter for the initiation of the fluidization phenomena when the solid particles start moving in the bed. In the current study, the investigated total aeration rates in LS are chosen to be multiples of the fluidization velocity.

#### 4.4. Numerical Model and Simulation Parameters

##### 4.4.1. Geometrical model

Numerical simulations are performed for a 10 MWth capacity atmospheric CFB gasifier reactor. The riser of the reactor has an inner diameter of 1.3m and a height of 18 m. The CFB system has a Lapple type cyclone with an inner diameter of 1.68 m. The standpipe having a cylindrical shape with a diameter of 0.4m is located between the cyclone and the LS as shown in Figure 4.5.

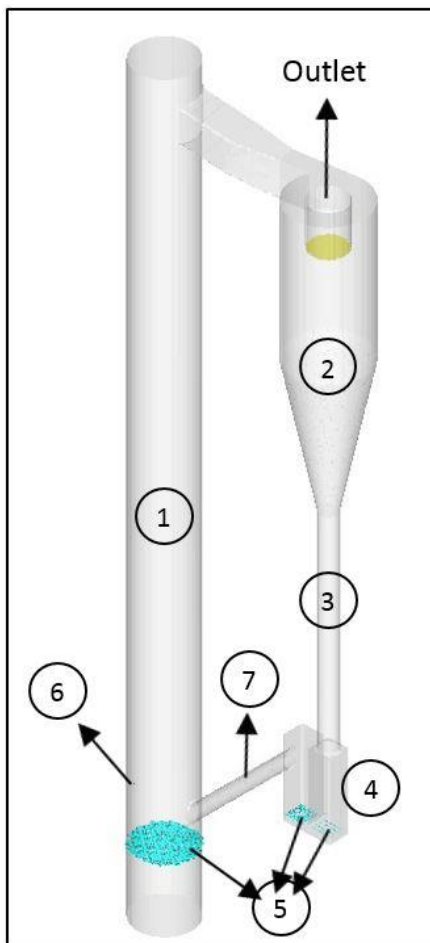


**Figure 4.5** Geometrical Model of the CFB system: Riser (1), cyclone (2), standpipe (3), loop-seal (4), air feed (5), solid particle feed location (6) and recycle pipe (7).

The details of the LS can be seen in the drawing given in Figure 4.6. Here, the LS is divided into two parts by a plate; a supply chamber (region under the standpipe) and a recycle chamber with the same cross-sectional area of 0.5 m x 0.5 m and a height of 1.78 m. These chambers are connected to each other by a gap (with a height of 0.18 m) located

at the bottom of the separator plate. The recycle chamber is connected to the reactor bed by a recycle pipe.

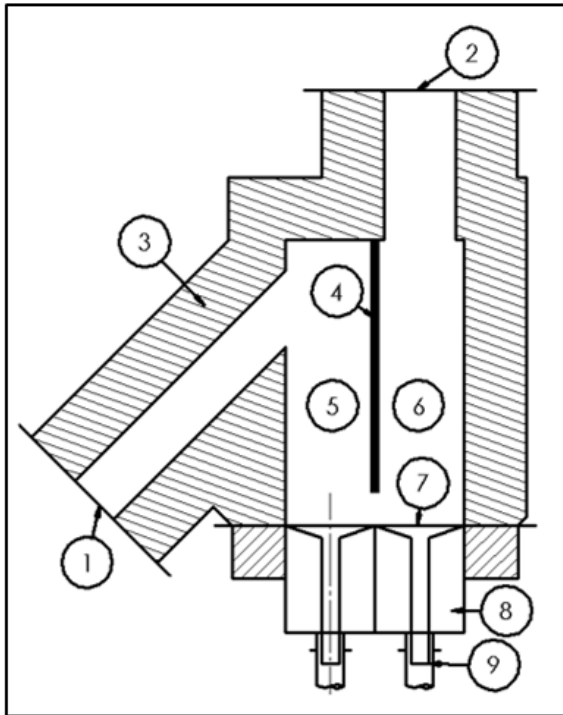


**Figure 4.6** Technical drawing of the loop-seal: Riser connection (1), standpipe connection (2), refractory wall (3), separator plate (4), recycle chamber (5), supply chamber (6), distributor (7), air-box (8), drain (9)

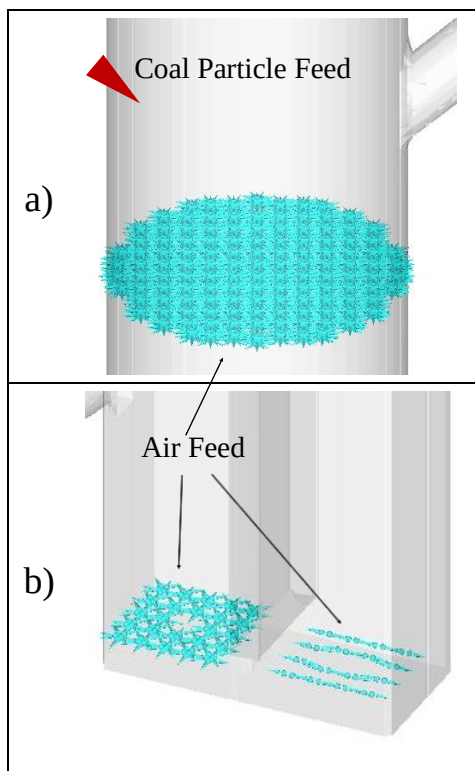


**Figure 4.7** Air and coal injection points in the reactor bed (a) and loop seal (b)

In the CFB system model, air is supplied to the riser through 128 nozzles, each having 32 holes, located on a sparger. Aeration of the loop-seal is provided with 16 nozzles, each with 2 holes, at the supply chamber and with 24 nozzles, each with 8 holes, at the recycle chamber. Nozzles in the reactor and the LS chambers are modeled as injection points to simplify the geometry for the simulations as seen in Figure 4.7. Similarly, the particle feed inlet is represented as a single injection point in the simulations.

#### **4.4.2. Computational Method**

The 3-D unsteady governing equations (continuity, momentum and turbulence) given in Chapter 2 are solved to obtain the continuous phase characteristics of the flow. The MP-PIC method described in the same section, which solves a transport equation for the particle distribution function is used to compute the discrete phase field. Isothermal non-reactive flow is assumed in the simulations since the objective of this study is to examine the SCR and the pressure variations in the system. Thus, the energy equation is not taken into account for numerical setup. Also, the mass transfer between phases is not considered for the computations.

#### **4.4.3. Operational Conditions**

In most of the experimental and numerical studies, SCR is investigated at cold flow conditions. However, in the present study, the particle circulation in the loop-seal part of the system has been investigated at an operating temperature of 900 °C. The gas-particle interactions and the circulation of particles are examined at operating temperature since the thermo-physical properties of the gas phase depend on vary with temperature. As the reactions are mainly completed in the fluidized bed and riser section of the gasification reactor, the temperature is supposed to remain constant, if it is well insulated, at the rest of the system components including cyclone, standpipe and LS. Moreover, in LS, solid particles are mainly the ashes captured from the cyclone. It can be concluded that the temperature level remains almost constant through whole system for the steady operating conditions. In addition, it is assumed that the CFB system walls are adiabatic so that there is no heat transfer to the surroundings by radiation, convection and conduction.

Liu et al. studied experimentally and numerically on a double bed pilot-scale biomass gasification system having 1 MWth capacity [46]. They observed that the temperature

level changes within 10% in the fluidized bed region of the system and about 5% in the second reactor region rest of the system. In the present work, isothermal non-reactive flow is assumed in the simulations since the objectives are to examine the SCR and the pressure variations in the system. The operating temperature is chosen as 900°C and the operating gauge pressure is 0.05 bar in all cases corresponding to common operating conditions. Air feed temperature is assumed to be the same as the operating temperature.

Total particle mass flow input in the reactor is kept constant as 1.8 kg/s for all cases. For total aeration of LS, 11 different aeration rates have been selected to obtain fluidization velocities between  $1 \times U_{mf}$  and  $48 \times U_{mf}$ . Moreover, the ratio of the aeration rate of the recycle chamber to that of the supply chamber, RS, is defined to be another parameter to be studied. Four different RS values are examined for each LS aeration rate (LAR) parametrically. LAR and RS values for simulation cases are listed in Table 4.6. In total, 44 cases of different aeration conditions which refer to the aeration rate scale for all possible fluidization regimes are investigated numerically. With analysis of all these cases, the fluidization behavior of LS is examined for various gas velocities corresponding to different bed structures.

As a boundary condition for discrete phase, the solid particles that exit from the cyclone outlet are assumed to be fed back into the reactor; therefore, the total solid inventory in the domain is held constant for all simulation cases.

**Table 4.6** LAR and RS values

| #                                       | 1    | 2    | 3     | 4     | 5     | 6     | 7     | 8     | 9      | 10     | 11     |
|-----------------------------------------|------|------|-------|-------|-------|-------|-------|-------|--------|--------|--------|
| <b>LAR</b><br><b>[U/U<sub>mf</sub>]</b> | 1    | 2    | 4     | 8     | 12    | 18    | 24    | 30    | 36     | 42     | 48     |
| <b>LAR</b><br><b>[g/s]</b>              | 3.82 | 7.64 | 15.28 | 30.56 | 45.84 | 68.76 | 91.68 | 114.6 | 137.52 | 160.44 | 183.36 |
| <b>RS [-]</b>                           | 3    | 5    | 10    | 15    |       |       |       |       |        |        |        |

#### 4.4.4. Mesh Independency Study

Mesh independency is studied to determine the optimum computational grid size without losing the accuracy of numerical results. Four different mesh sizes are tested as shown in Figure 4.8. All cases are calculated for a physical time of 100s. The results show that all cases reached statistically steady-state flow regime approximately in 30 seconds.

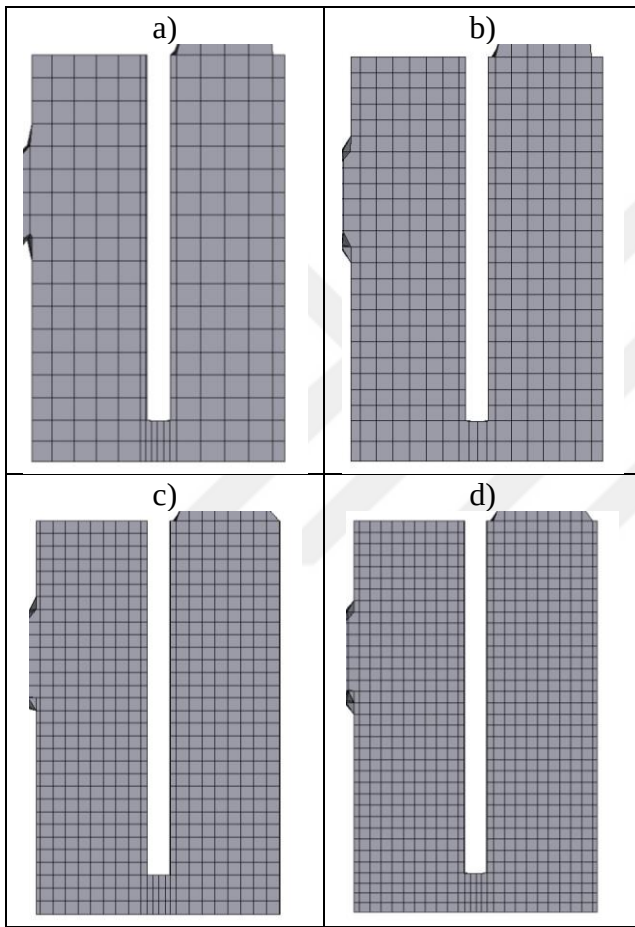


**Figure 4.8** Cartesian mesh sizes prepared for mesh independency study: a) Mesh1: 48k, b) Mesh2: 108k, c) Mesh3: 217k and d) Mesh4: 316k

The mesh size did not have a significant effect on pressure results at the reactor bed as seen in Figure 4.9-a. However, it affects the pressure data obtained from the bottom of the recycle chamber. Pressure fluctuations could not be captured for coarser mesh sizes as seen in Figure 4.9-b.

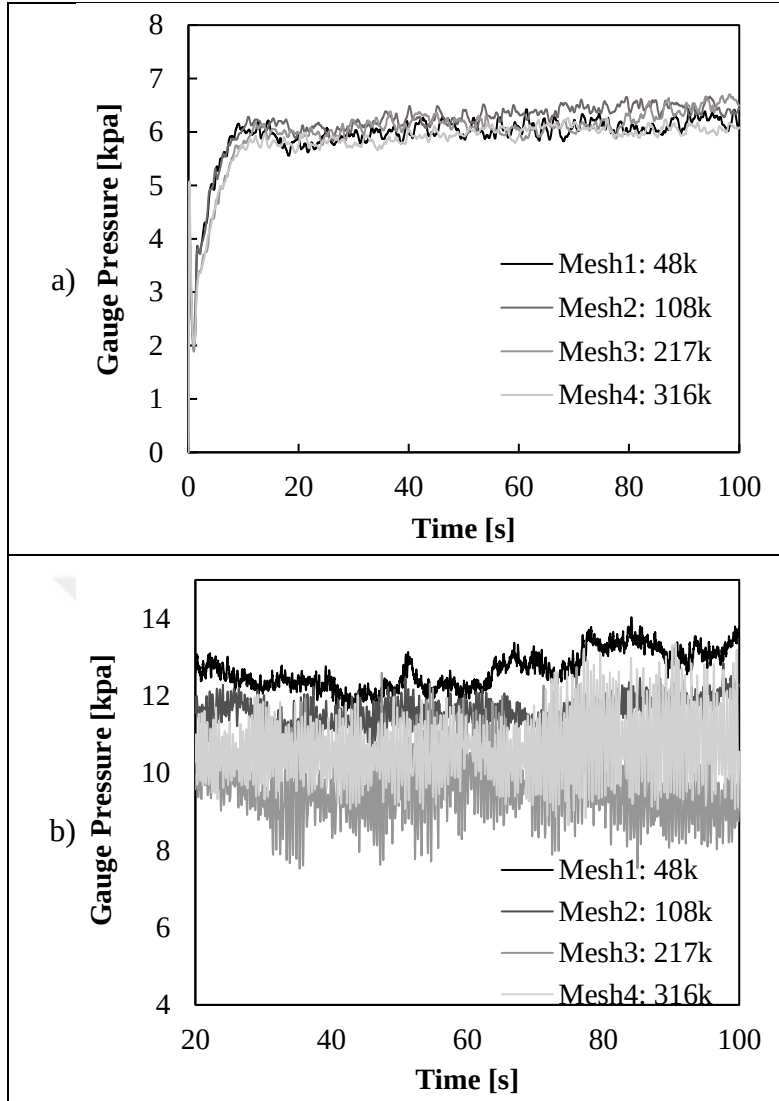


**Figure 4.9** Mesh independency study: (a) unsteady pressure data at the bottom of the reactor bed and (b) in the recycle chamber.

In addition, SCR values considerably depend on the mesh size and coarser grids give higher SCR values as shown in Figure 4.10-a&b. The SCR value is obtained from the slope of the curve in Figure 4.10-a and the same values are observed for Mesh3 and Mesh4. Thus, Mesh3 with 217k elements is considered to be suitable for the simulations.

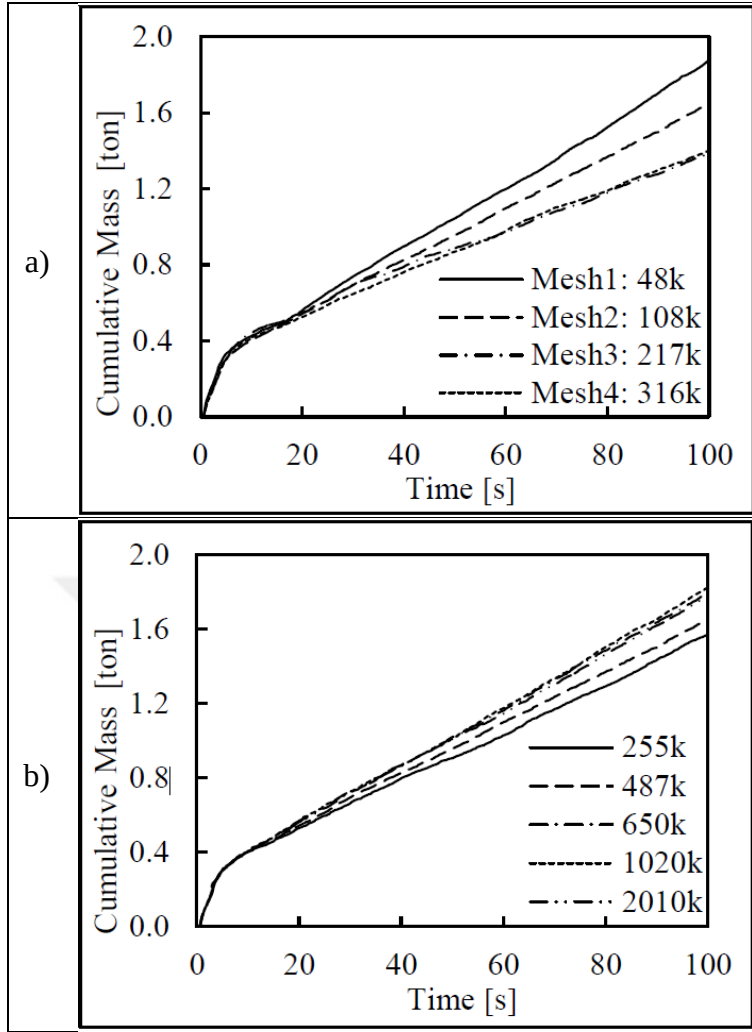


**Figure 4.10** a) Mesh independency and b) Cloud independency study in terms of cumulative recycled solid particle mass.

#### 4.4.5. The determination of the proper computational particle cloud size

Solid particles are represented as computational points, which are called particle clouds in simulations. Each particle cloud has a certain number of physical particles with the same diameter defined as number density. The cloud size can be varied using this parameter, i.e. the higher the number density the larger the cloud size, and vice versa.

The optimum initial cloud size should also be determined for the simulations. For this purpose, five different cloud sizes are selected and the results of simulations have been evaluated. In Figure 4.10-b, it is seen that the lower cloud sizes give lower SCR values. For the cloud sizes larger than 650k, SCR remains constant. Moreover, the unsteady pressure curves for cloud sizes are also examined and it is observed that the cloud sizes

larger than 650k are very close to each other. Therefore, it can be concluded that the initial cloud size of 650k is precise enough for modeling studies of the system.

#### 4.4.6. Other simulation parameters

The particle physical properties and simulation parameters set for the multiphase solver are shown in Table 4.7. Particle size distribution (PSD) for solid fuel and ash particles used in simulations can be seen in Figure 4.11.

**Table 4.7** Simulation parameters

| Property                                                | Unit              | Value |
|---------------------------------------------------------|-------------------|-------|
| Particle Density                                        | kg/m <sup>3</sup> | 1300  |
| Packed Bed Volume fraction                              | -                 | 0.6   |
| Particle-wall tangential momentum retention coefficient | -                 | 0.3   |
| Particle-wall normal momentum retention coefficient     | -                 | 0.99  |
| Max. Momentum redirection from collision                | %                 | 40    |
| Maximum time step                                       | s                 | 0.01  |
| Total physical time                                     | s                 | 100   |

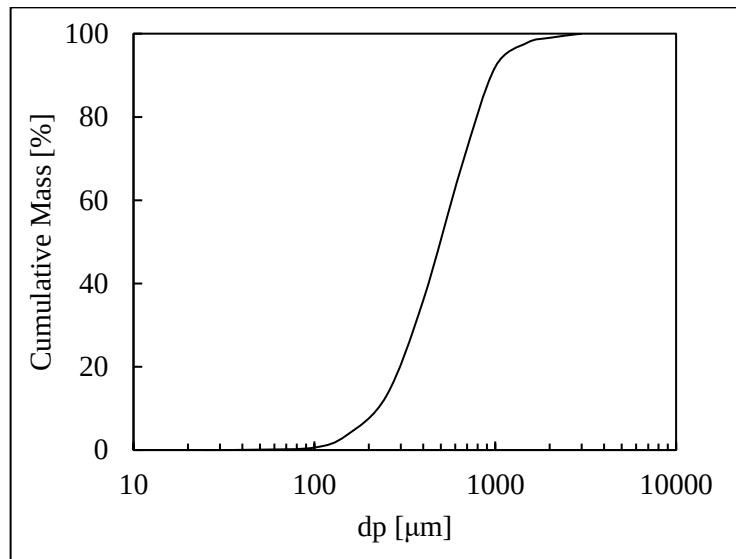


**Figure 4.11** PSD of fuel and ash particles

For the initialization of the discrete phase computations, solid particles are filled into the reactor bed up to a height of 1.8 m from the bottom and into the LS chamber until a height of 3.25 m, as seen in Figure 4.12.

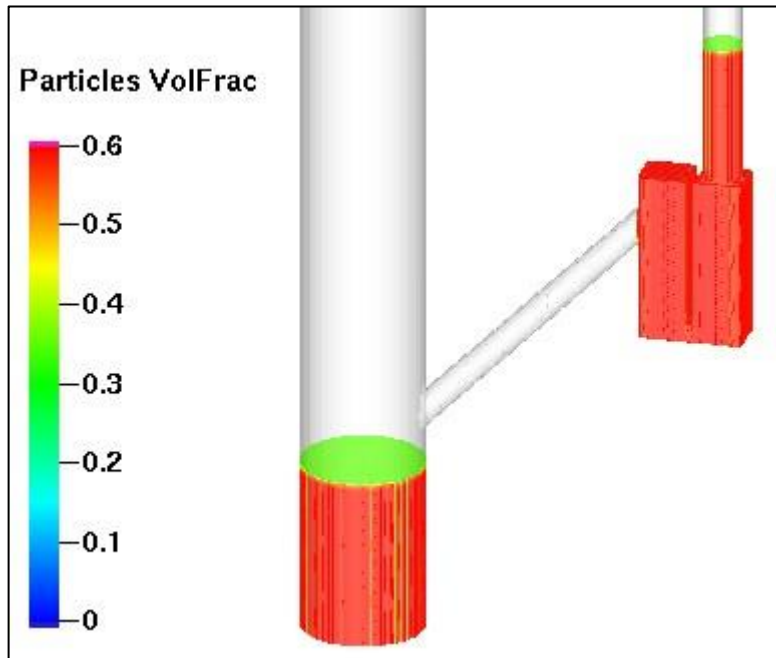


**Figure 4.12** The initial particle distribution

Five pressure monitoring points are defined to observe unsteady pressure distributions along a loop in the simulations (Figure 4.13). They are located at the bed, cyclone inlet, inlet of the standpipe, bottom of both supply chamber and recycle chamber. A flux plane is defined in the recycle pipe to acquire unsteady particle mass flow rate data.

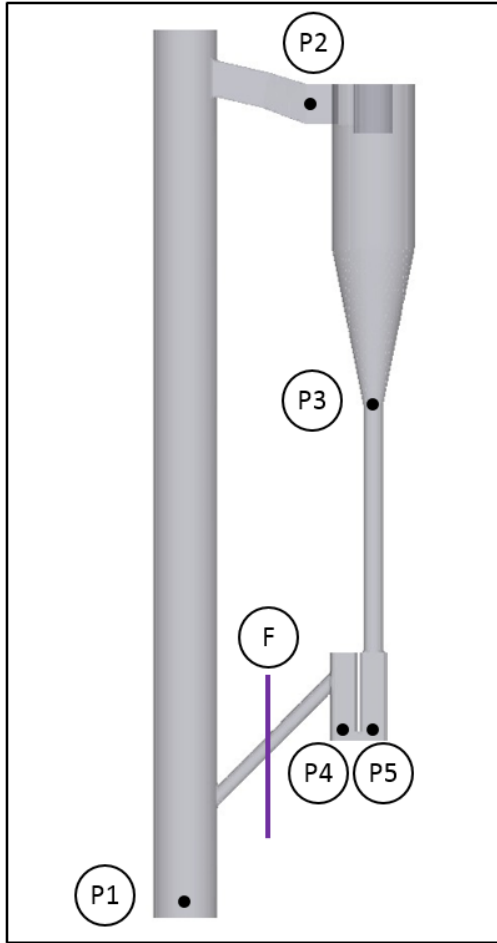


**Figure 4.13** Transient pressure data points for the reactor bed (P1), cyclone inlet (P2), top of standpipe (P3), supply chamber (P4), recycle chamber (P5) and flux plane (F)

#### 4.5. Numerical Results and Discussion

The computations are performed on a workstation by GPU based parallel computing method. The computation time was approximately 60 hours for each case. Figure 4.14 reports the contours of static pressure and particle volume fraction distribution along the CFB system for total aeration rate of LAR#5 and RS value of 10. It is found that the pressure value at the loop-seal is higher than that of riser bed section. Additionally, it is observed that there is not any particle accumulation in the stand-pipe which means operational parameters are convenient to provide a good particle recirculation in the CFB system.

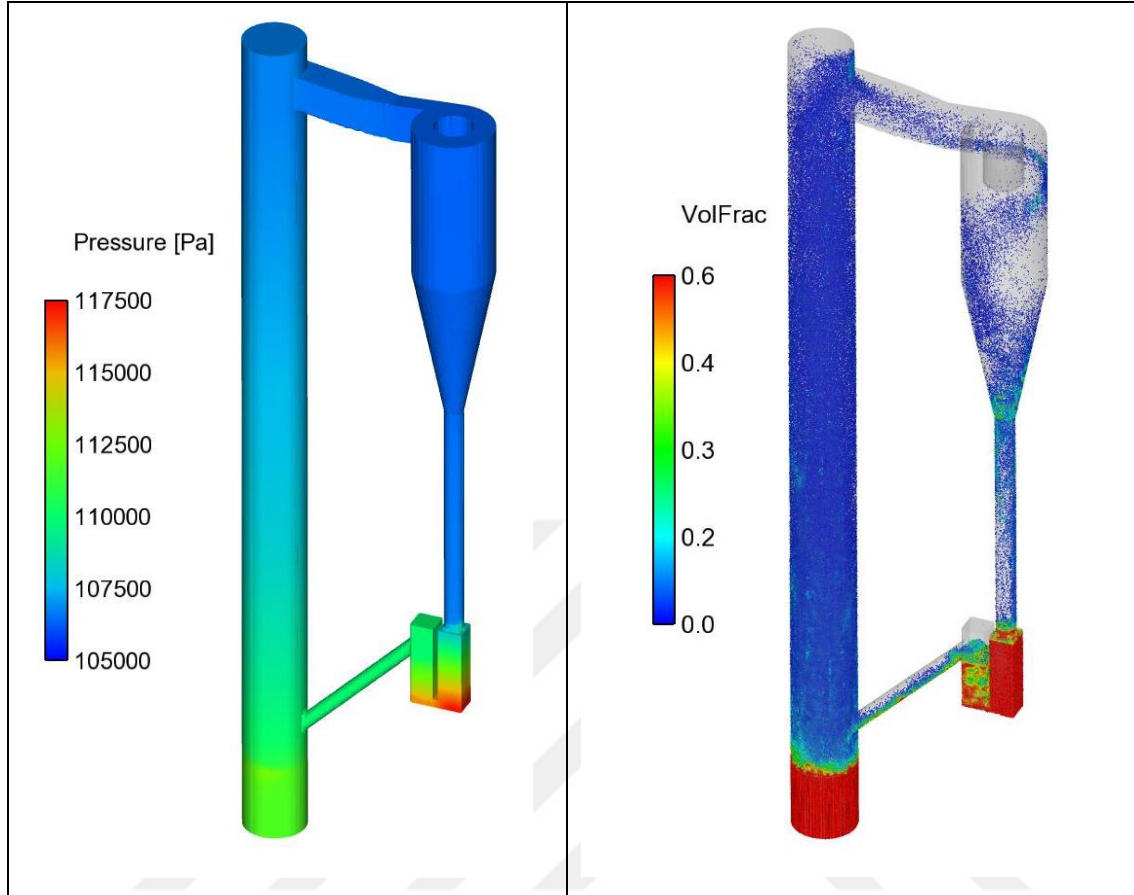


**Figure 4.14** Pressure (a) and particle volume fraction (b) distributions on whole the system for LAR#5 & RS=10

#### 4.5.1. Effect of LS aeration on circulation

The calculated SCR and fluid mass flow rate, represented by  $G_p$ , through the recycle pipe can be seen in Figure 4.15-a&b. Figure 4.15-a shows that SCR is increasing suddenly while changing the total aeration rate in the loop-seal between  $1xU_{mf}$  and  $8xU_{mf}$ . Peak value of the circulation is shifted to the right, i.e. at higher aeration rates, at higher RS values. In addition, at elevated aeration rates SCR reaches about a constant value of 12 kg/s and it is almost independent of the RS value. For all RS cases, a similar distribution of initially a sudden increase, after reaching a peak value a decrease and finally a region of almost constant SCR value has been observed. In addition, it can be reported that RS parameter is highly effective on SCR especially for aeration rates up to  $18xU_{mf}$ . However, this effect is very complex between  $8xU_{mf}$  and  $18xU_{mf}$ . In case of higher RS values with lower aeration rates, fluidization in the supply chamber may not be enough to circulate

the particles in the loop. In Figure 4.15-b, it is shown that there is almost a linear relationship between the aeration rate of LS and the air mass flow through recycle pipe. However, for higher RS values at higher aeration rates, there is a decrease in mass flow rate indicating that more air passes through the standpipe. This situation is undesirable for combustion, gasification and chemical-looping systems since higher O<sub>2</sub> concentration rates will be observed at the exit of the reactor.

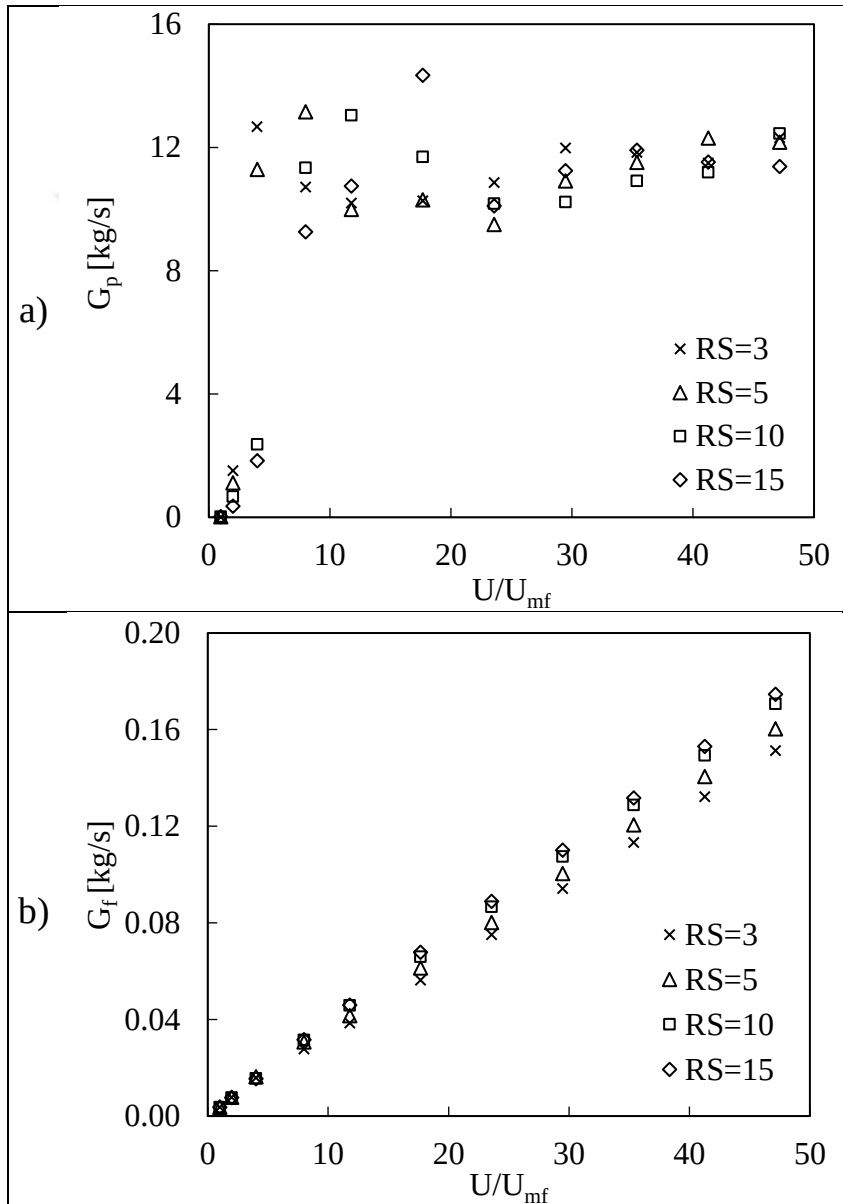


**Figure 4.15** Effect of the total aeration rate of LS on SCR (a) and fluid mass flow rate in recycle pipe (b) for different RS values

Particle distributions in the loop-seal for different aeration rates at a constant RS value, 10, are given in Figure 4.16.

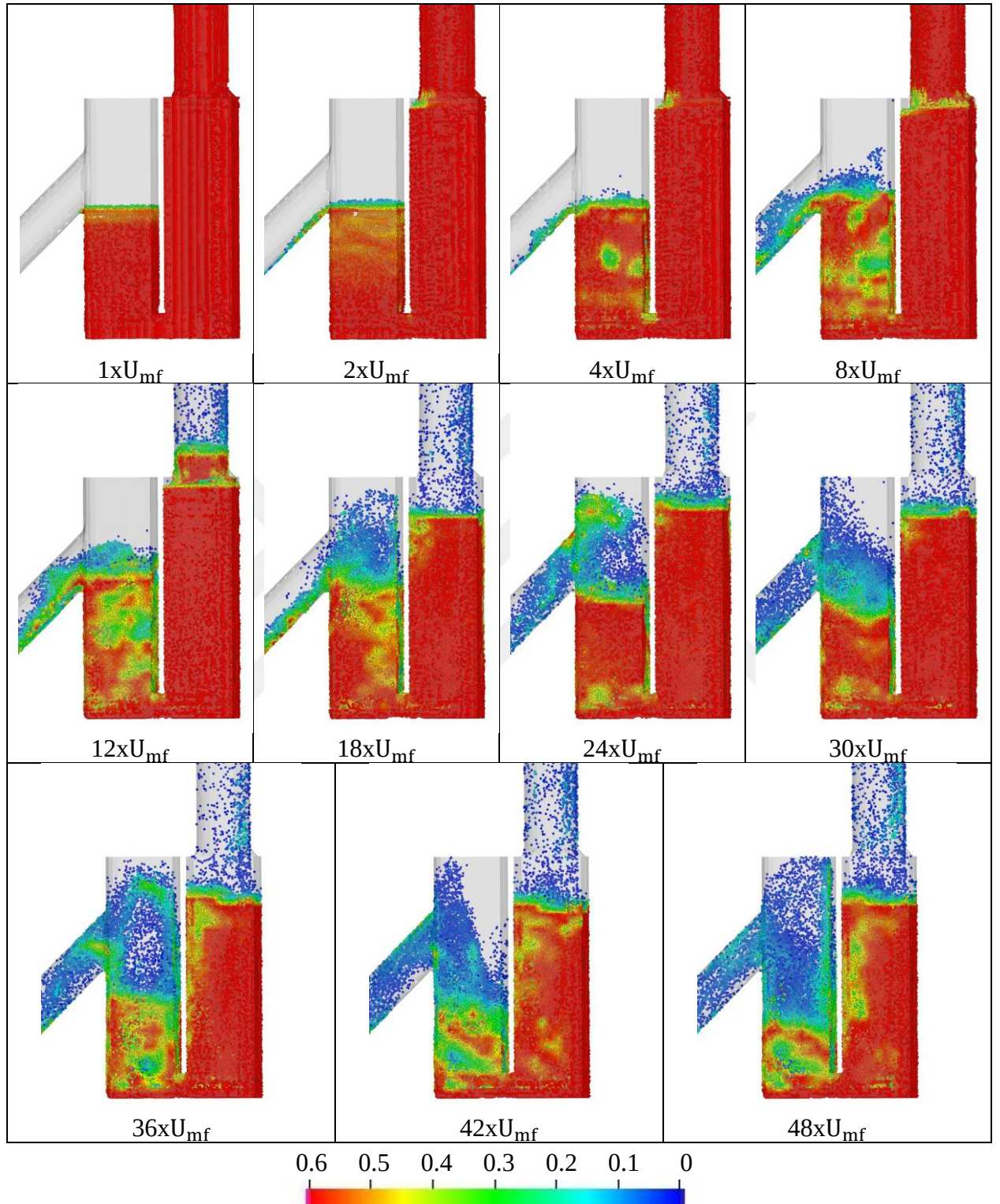


**Figure 4.16** Particle volume fraction distribution at loop-seal for different total aeration rates and for a constant RS value (RS=10)

It can be easily said that fluidization starts at  $1xU_{mf}$ , although no recirculation occurs. The particle recirculation starts at  $2xU_{mf}$  as seen in figure. Bubble formation starts at aeration rate of  $4xU_{mf}$  in the LS. The bubbles are getting coarser with the increase in the aeration rate. On the other hand, bed structure of the loop-seal almost disappears at higher aeration rates (above  $30xU_{mf}$ ), especially in the recycle chamber, which may cause back flow gas leakage from reactor bed to the loop-seal.

#### **4.5.2. Effect of LS aeration on mean pressure and pressure fluctuations of LS aeration on circulation**

The effect of LS aeration rate on pressure has been also examined. It is observed that the mean static pressure at the supply chamber increases rapidly for aeration rates up to  $4xU_{mf}$  in line with the increase in SCR as seen in Figure 4.17-a. However, at higher aeration rates, this parameter decreases continuously. Moreover, it is found that for aeration rates up to  $4xU_{mf}$  lower RS values result in higher pressure data at the supply chamber. This is one of the significant results of this study.

A sudden increase in the mean static pressure at the recycle chamber can be seen in Figure 4.17-b for aeration rates up to  $8xU_{mf}$ . A continuous decrease is observed at higher aeration rates as in the supply chamber. The RS parameter has been found relatively more effective on pressure data of the recycle chamber compared to pressure data of the supply chamber for all aeration rates. It can be clearly stated that lower RS values yield more static pressure in the recycle chamber up to the aeration rate of  $18xU_{mf}$ . A similar behavior is observed for supply chamber as well.

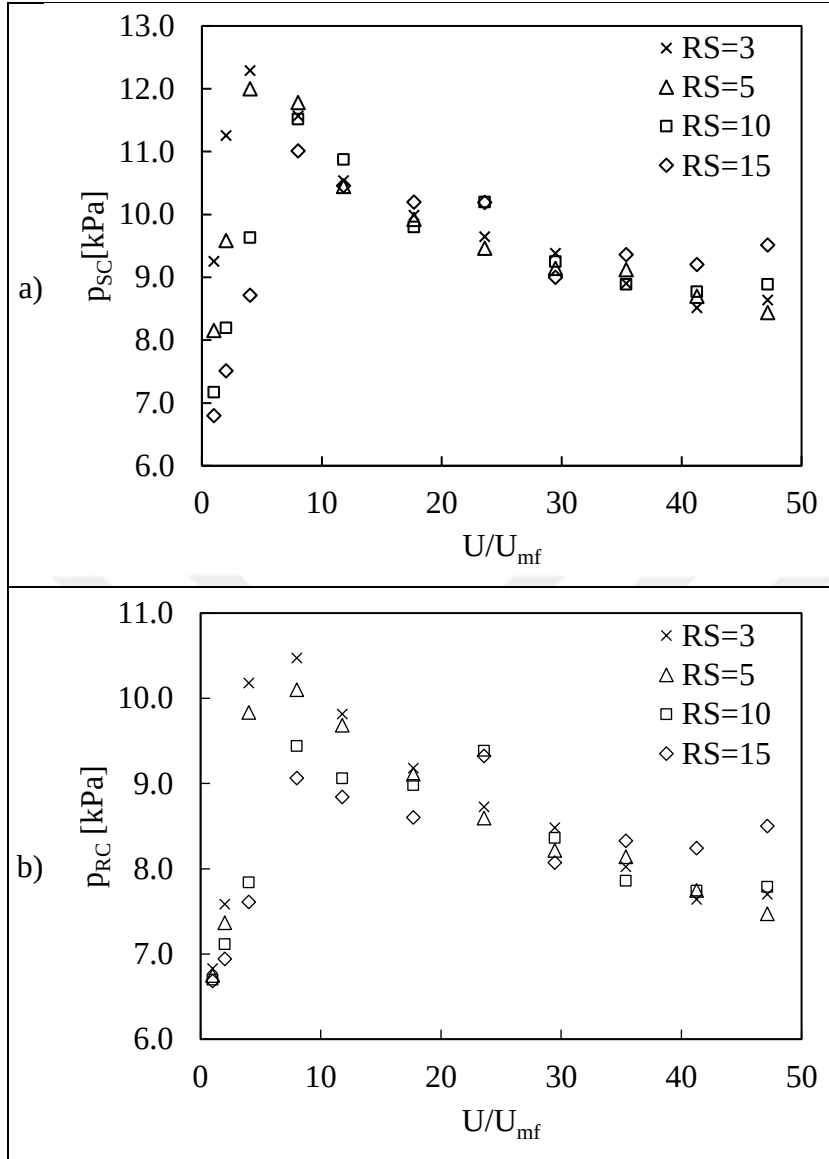


**Figure 4.17** The relationship between the total aeration rate of LS and the mean static pressure at LS chambers for different RS values: Supply chamber (a) and recycle chamber (b)

Sobrino et. al. investigated the behavior of the standard deviation of pressure fluctuations,  $\sigma_p$ , in fluidized beds for Geldart group-B particles and showed the relationship between the minimum fluidization velocity and pressure fluctuations [120]. It is known that the fast motion of gas bubbles and pressure gradients inside the reactor produce fluctuations of relatively higher amplitudes. Pressure fluctuations cause operational noise and erosion on refractory walls in CFB systems. Figure 4.18-a&b shows the numerical results for pressure fluctuations, in terms of standard deviations, both in supply and recycle chambers. It is observed that the amplitude of pressure fluctuations increases for the aeration rates up to  $18xU_{mf}$ . Peak pressure fluctuations for aeration rate values between

$18xU_{mf}$  and  $30xU_{mf}$  are observed for all RS values, which stand for rough running of LS. Here, the sudden change in fluctuations indicates a change in the fluidization regime. It can be concluded that this range represents the transition region from bubbling regime to turbulent regime of fluidization.

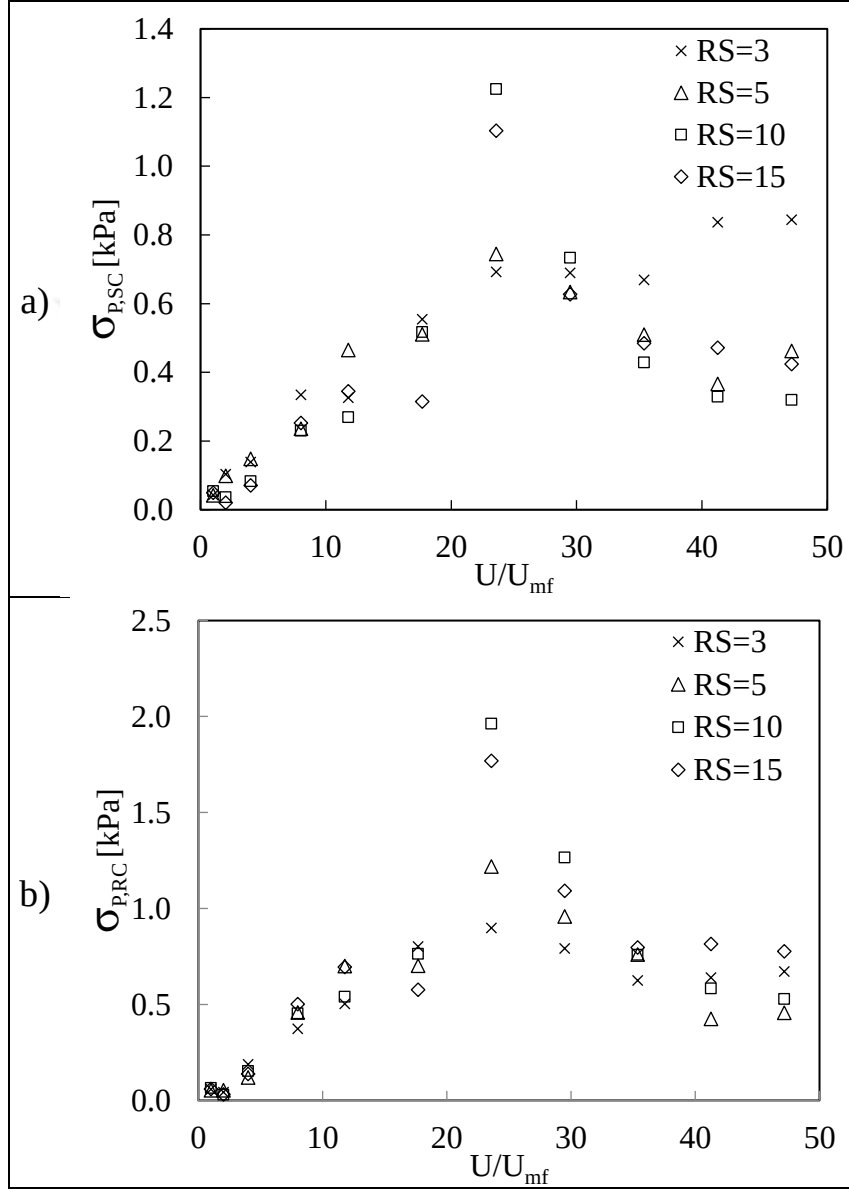


**Figure 4.18** Influence of the total aeration rate of LS on pressure fluctuations at LS chambers for different RS values: Supply chamber (a) and recycle chamber (b)

It is observed that the amplitude of pressure fluctuations increases for the aeration rates up to  $18xU_{mf}$ . However, a peak fluctuation for aeration rate values between  $18xU_{mf}$  and  $30xU_{mf}$  is observed for all RS values, which stand for rough running of LS. Here, the sudden change in fluctuations indicate a change in the fluidization regime. It can be

concluded that this range represents the transition region from bubbling regime to turbulent regime of fluidization.

The relationship between SCR and differential pressure data obtained from LS chambers has also been investigated for different fluidization conditions. When all cases are plotted in terms of circulation rate and differential pressure, it is seen that they can be grouped into three regions as seen in Figure 4.19. Region-I represents aeration rates less than  $4xU_{mf}$ . It is also found seen that lower fluidization occurs at lower SCR values in this region. Region-II represents mostly aeration rates that produce turbulent and transition regimes. In this region differential pressure is low, however, SCR is high. Region-III corresponds to aeration rates higher than  $4xU_{mf}$  and below the transition region. Maximum SCR values are achieved in this region accompanied with relatively higher differential pressures.

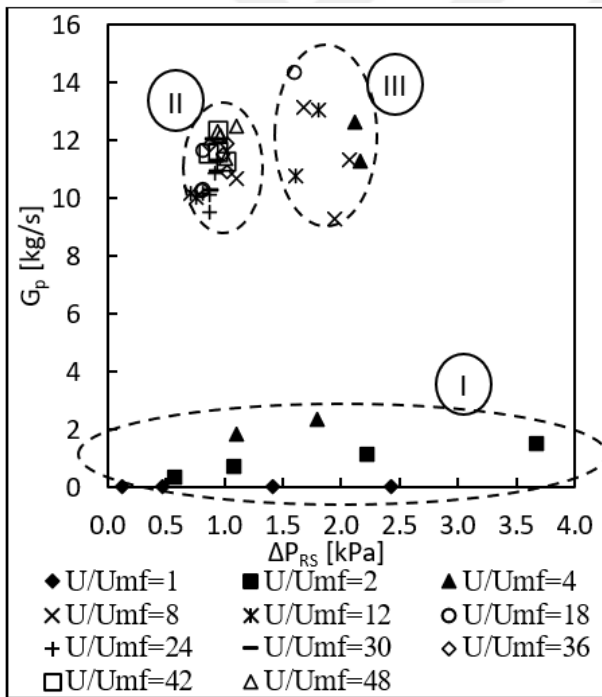


**Figure 4.19** Relationship between SCR and differential pressure at LS chambers

#### 4.5.3. The effect of the aeration rate on particle accumulation height in the standpipe

The effect of the aeration rate on the particle accumulation height in the standpipe is examined in this part. The particle accumulation height in the standpipe from the bottom of the supply chamber is obtained by processing particle volume fraction data on the centerline of the standpipe. In the calculations of the unsteady height data, 0.05 is taken as a cut-point reference for volume fraction on the centerline. Then, the mean height, seen in Figure 4.20, is calculated by time averaging. It is found that there is no particle accumulation in the standpipe for the LS aeration rates above  $18xU_{mf}$  indicating an “over-circulation” for the CFB system. Solid circulation is observed as insufficient to keep the system at the steady state regime for aeration rates below  $4xU_{mf}$ . Additionally, RS parameter has an effect on particle height only for aeration rates between  $4xU_{mf}$  and  $18xU_{mf}$ . It is also found that the particle height decreases for lower RS values and this stands for higher SCR at these conditions. Therefore, for a stable operation, the aeration rate is recommended to be higher than  $4xU_{mf}$  and lower  $18xU_{mf}$  for all RS cases of studied system. Moreover, at higher RS values more accumulation of particles is observed in the standpipe as seen in Figure 4.20.

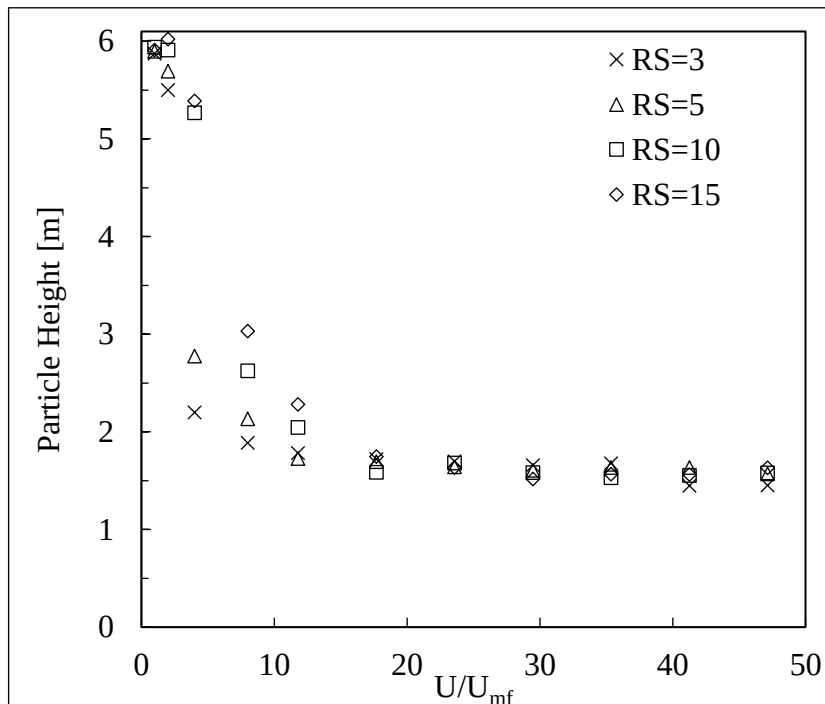


**Figure 4.20** Change in time-averaged particle height at standpipe with increasing total aeration rate of LS

Time-dependent particle accumulation height data for all cases are given in Figure A.17 in Appendix-F. As seen in the figures, the particle height increases continuously with time for aeration rates of  $1xU_{mf}$  and  $2xU_{mf}$ . The SCR values that indicate a steady-state regime are obtained first at an aeration rate of  $4xU_{mf}$  and RS values of 3 and 5. Steady state regime is observed for all cases having aeration rates higher than  $8xU_{mf}$ . In addition, high fluctuations in particle height in the stand-pipe at higher aeration rates (above  $24xU_{mf}$ ) but lower RS values (for 3 & 5) indicate the existence of large bubbles or pressure waves which causes unstable operational conditions. Therefore, for the present study the stable operational conditions may be recommended to be aeration rates of higher than  $8xU_{mf}$  and less than  $12xU_{mf}$  and RS values of higher than 10.

## 5. CONCLUSIONS AND RECOMMENDATIONS

In this dissertation study, it is fundamentally aimed to conduct an extensive investigation of the domestic low-rank coal resources in the fluidized bed gasification process by using experimental and numerical methods in order to make a comprehensive scientific contribution to the development of clean and efficient fluidized bed technologies.

In the second chapter, the physical and chemical characteristics of low-rank coal samples received from the coal site at Tekirdag are determined by using experimental methodologies. Different sample characteristics are obtained by applying different physical pre-processes such as washing and drying. Ash, moisture, volatile and fix carbon compositions are obtained from proximate analyses, while elemental distributions are specified by ultimate analyses. Heating value and particle size range are also obtained for the samples. Structural details and operating conditions of the experimental setup involving an atmospheric BFB gasifier in TÜBİTAK Marmara Research Center are clearly described. The experimental procedure and limitations of the lab-scale gasification system are also defined. Moreover, equations used during experiments and for the post-process of experimental results are given. Following concluding remarks can be drawn for the characterization of the coal samples:

- Very high ash (36.36%) and moisture (34.24%) contents are observed for the coal sample at as-received conditions (Sample-1). LHV of this sample is also calculated at very low level (1028 kcal/kg).
- Each physical pre-processes (washing & drying) improves LHV values of the samples by decreasing ash and/or moisture concentration. Calorific value of Sample-4 obtained by the application of two pre-processes on Sample-1, is determined 350% higher than as-received sample.
- While applied pre-processing improves the LHV content of the samples, a considerable increase in the elemental fraction of sulfur content in the coal samples is observed by considering the results of original basis ultimate analyses. The sulfur content of Sample-1 is obtained 2.85%, while it is measured 4.07% for Sample-4 in dry basis. It is needed to consume higher amounts of sulfur capturing material per unit fuel in the case of utilization of these samples in a commercial application.

The followings are recommended for the improvement of the experimental setup:

- Syngas exit temperature should be measured to evaluate the experimental results in detail.
- Activation period of the electric furnaces should be recorded during experiments to calculate the heat flux supplied into the gasification reactor.
- PID controller should be re-programmed by adding an option to conduct the experiments in quasi-adiabatic conditions. Time-dependent wall temperature data should also be collected during the experiments for the adiabatic case to ensure the appropriate steady-state conditions.
- H<sub>2</sub>O concentration in syngas should be measured in order to obtain detailed syngas composition, especially for high-moisture content coal samples and to make a more accurate comparison with numerical results.
- Dynamic bed height should be calculated by measuring the transient pressure data and using a correlation obtained from experimental studies.

The definition of the experimental setup is followed by the numerical methods used in the computational studies in Chapter 2. MP-PIC method is used for the numerical analysis. Governing equations, drag model, turbulence model as well as the physical interactions between discrete particle phase, continuous phase and boundaries of the computational domain in terms of heat, mass and momentum transfer are also presented in detail.

The scientific and technical background of gasification chemistry and alternative computational approaches to simulate the gasification process is another focus of Chapter 2. Main gasification reactions are defined as well as the combustion reactions for both heterogeneous and homogeneous phases. Alternative numerical solution methodologies in order to solve the chemistry of solid char conversion are investigated. In addition, two different devolatilization model is evaluated for specified coal samples and operating conditions: MGAS and CPD models. Kinetic rates of the reactions used in the numerical analysis are also defined.

In the third chapter, experimental results of the gasification of different coal samples are presented for different cases. Volumetric fractions of the species in product gas species

and reactor temperatures are measured during experiments. The effect of the ER parameter on the gasification process is evaluated for each sample. The results can be evaluated by the following comments in summary:

- As a general trend, LHV of the syngas decreases with increasing ER values. 2.05 MJ/Nm<sup>3</sup>, 2.67 MJ/Nm<sup>3</sup>, 3.30 MJ/Nm<sup>3</sup> and 3.79 MJ/Nm<sup>3</sup> of syngas LHV are calculated for ER values of 0.558 (ER2), 0.463 (ER3), 0.353 (ER4) and 0.267 (ER5), respectively.
- There is not any optimum value in the molar concentrations of CO, CH<sub>4</sub>, and H<sub>2</sub> species observed for any ER case according to the comparison of all experimental data. A similar decrease trend is also obtained for the molar concentrations of these species that affect LHV of syngas.
- It is recommended that the effect of relatively lower ER values should be investigated for further studies in order to determine optimum ER values for the gasification process.
- Despite this, an optimum ER value is obtained for the thermal gasification process efficiency. It is determined that ER value of 0.353 maximizes both hot and cold gas efficiency of the process.
- Wall temperature values are measured very close to the desired operating temperature of 825°C in every ER case for different coal samples. Since the total MFR of the coal is kept constant, it's evaluated that PID controller was activated furnaces especially for coal samples with relatively low calorific values and for lower ER values. It should be taken into consideration that activation of furnaces changes the boundary conditions of the experimental setup.

3D simplified geometrical model of the gasification reactor, its boundary conditions and solver parameters are defined for the numerical setup. Coal Sample-3 and MGAS devolatilization model are selected for the numerical analyses in order to validate the results of experimental studies. Numerical results of the syngas compositions and LHV data are obtained for different ER cases and compared to the experimental data. Effects of the devolatilization model, initial bed height and thermal wall boundary condition are also investigated in numerical analyses. The results of the numerical studies conducted for the lab-scale BFB can be summarized by the following items:

- Particle volume fraction distributions are obtained for each ER value. A significant increase in bed height is observed for ER3 and ER2 with increasing superficial velocities.
- A small change (max. 10%) in static pressure values through the reactor axis is obtained with increasing ER values. According to the numerical results, higher ER values yield relatively higher static pressure in the bed region.
- It is obtained that ER parameter has a significant effect on the temperature values in both fluidized bed and free board regions. Mean reactor temperature increases with increasing ER values. Time and space averaged reactor temperature values in ER5, ER4, ER3 and ER2 cases are calculated 989 K, 1024 K, 1088 K and 1146 K, respectively.
- Although the reactor wall temperature is kept constant during experiments, it is not possible to keep every single point at equal temperature levels in reality. Numerical results show a considerable increase in temperature levels in the bed region for all cases due to the combustion reactions of the char particles. Also, a sudden decrease in temperature is determined in the upper region of the bed where the coal and nitrogen are fed at atmospheric temperatures. A decreasing trend in temperature levels is observed for all ER cases due to the effect of endothermic gasification reactions.
- A good agreement between numerical and experimental data for the molar concentration values is obtained for ER5 and ER4 cases by using MGAS devolatilization model. However, significant differences is observed especially for CO and H<sub>2</sub> species at higher ER cases. These differences affect both LHV of syngas and the thermal efficiency of the gasification process; consequently, similar gaps are also observed for these parameters. While the difference between experimental and numerical data for LHV is obtained 12% for ER5 it reaches up to 55% for ER2.
- In order to understand the reason of discrepancy in molar concentrations in ER3 and ER2 cases, firstly, the volumetric concentration distributions of CO and H<sub>2</sub> species are evaluated. It is observed that major amount of H<sub>2</sub> is formed in the free-board region where a specific decrease occurs in methane concentration. Additionally, a small increase in CO concentration is observed in the same regions. The combination of these evaluations refers to the effect of the methane

reforming reaction. Hence, the effect of this reaction is investigated for all ER cases. At the end of the computations, a consistency between experimental and numerical analyses is obtained for high ER values in the case of excluding methanation reaction. Nevertheless, this numerical approach causes a significant difference between numerical and experimental results for molar concentration values of methane, carbon monoxide and hydrogen species for lower ER values. As a result, it is recommended that advanced kinetic models should be used for methane reforming reaction including operating conditions such as local temperature and concentration of other species in the flow field.

- In the scope of numerical studies, the effect of the devolatilization model is also investigated for a single ER case. Numerical results show that CPD model predicts lower volumetric concentration values for CO, H<sub>2</sub> and CH<sub>4</sub> in syngas composition. This also yields smaller values for LHV of the syngas output. It is observed that MGAS devolatilization model predicts the molar concentration of species better than CPD model.
- The effect of initial bed height on syngas composition is also investigated in the numerical studies. It is determined that the molar concentration of CO<sub>2</sub> is not affected by the change in initial bed height. On the other hand, there is a considerable effect on the molar concentration of H<sub>2</sub> (approximately 20%) in syngas due to the relative change in coal feeding level and the change in volumetric concentrations of oxygen in the bed with increasing initial bed height.

In the last chapter, a design concept of a 10 MWth thermal input capacity CFBG for specified operating conditions and coal sample is presented. The design parameters of each main component of the system are defined. Dimensioning methodologies with engineering assumptions are given in detail. A numerical study of the CFBG reactor is performed for several aeration cases for the LS of the system with the assumption of non-reactive isothermal conditions. The following conclusions are drawn for the numerical analyses of the hydrodynamics of the CFBG:

- Maximum SCR is obtained in the total aeration rate range of the LS from  $8xU_{mf}$  to  $18xU_{mf}$ .

- For all RS cases, a similar distribution of initially a sudden increase then a peak value and finally a region of almost constant SCR has been observed. RS parameter is found not effective on SCR for aeration rates higher than  $18xU_{mf}$ .
- There is almost a linear relationship between the aeration rate of LS and the air mass flow through recycle pipe. However, for higher RS values at higher aeration rates, there is a decrease in mass flow rate of gas indicating that more air passes through the standpipe.
- The bed structure has been examined in terms of particle plots at a chosen RS value of 10. It is observed that the bed structure is almost disrupted in the loop seal for the aeration rates of higher than  $30xU_{mf}$ . This may cause leakage of synthetic gas produced in the reactor bed to LS and result in serious operational instabilities in the CFB system.
- The mean static pressure at the supply chamber increases rapidly for aeration rates up to  $4xU_{mf}$  due to the particle accumulation in the standpipe. However, at higher aeration rates, this parameter decreases continuously.
- A transition regime characterized by a peak pressure fluctuation and a considerable decrease in SCR is identified at an aeration rate of  $24xU_{mf}$ .
- Three regions are revealed when the effect of differential pressure between two chambers of LS on SCR is depicted. It is observed that Region-III yields an optimum range for the operation with higher SCR and lower pressure fluctuations.
- There is no particle accumulation in the standpipe for the aeration rates above  $12xU_{mf}$  indicating an “over-circulation” for the CFB system. Insufficient solid circulation and high levels of particle accumulation in standpipe are obtained for aeration rates below  $4xU_{mf}$ . Therefore, it is not possible to keep the CFB system at a steady-state regime for these conditions. RS parameter has an effect on particle height only for aeration rates between  $4xU_{mf}$  and  $24xU_{mf}$ . It is also found that the particle height decreases for lower RS values and this stands for higher SCR at these conditions.

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## APPENDICES

### APPENDIX A – Comparison Of Experimental Results Of Various Gaseous Species And LHV of Syngas Obtained From Different Coal Samples in BFB Gasification Process

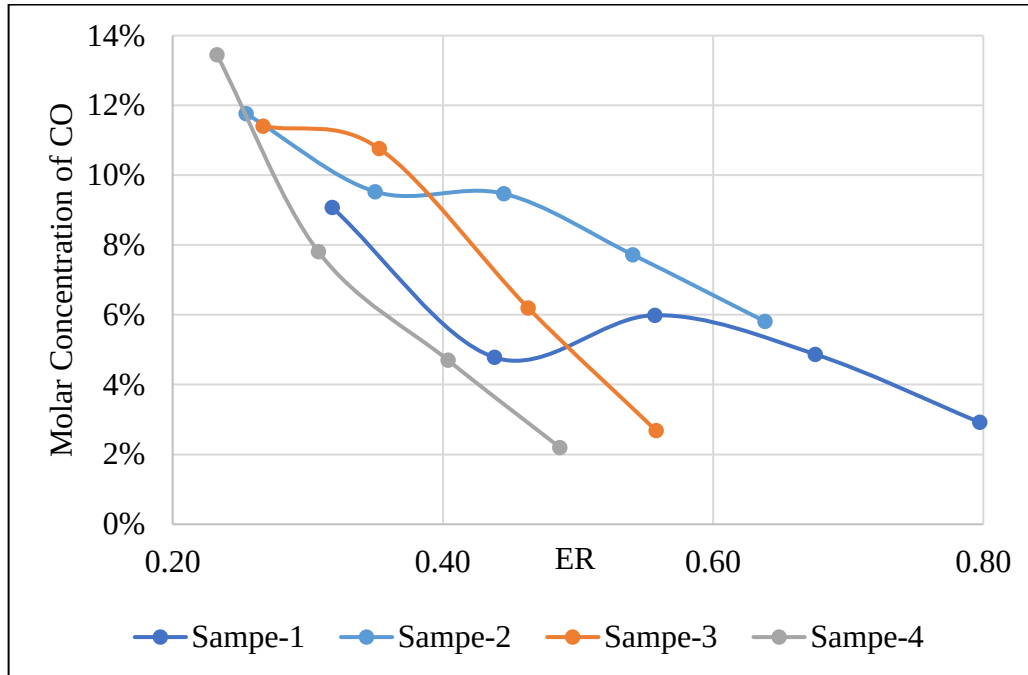


**Figure A.1** Comparison of the experimental results of molar concentrations of CO in syngas

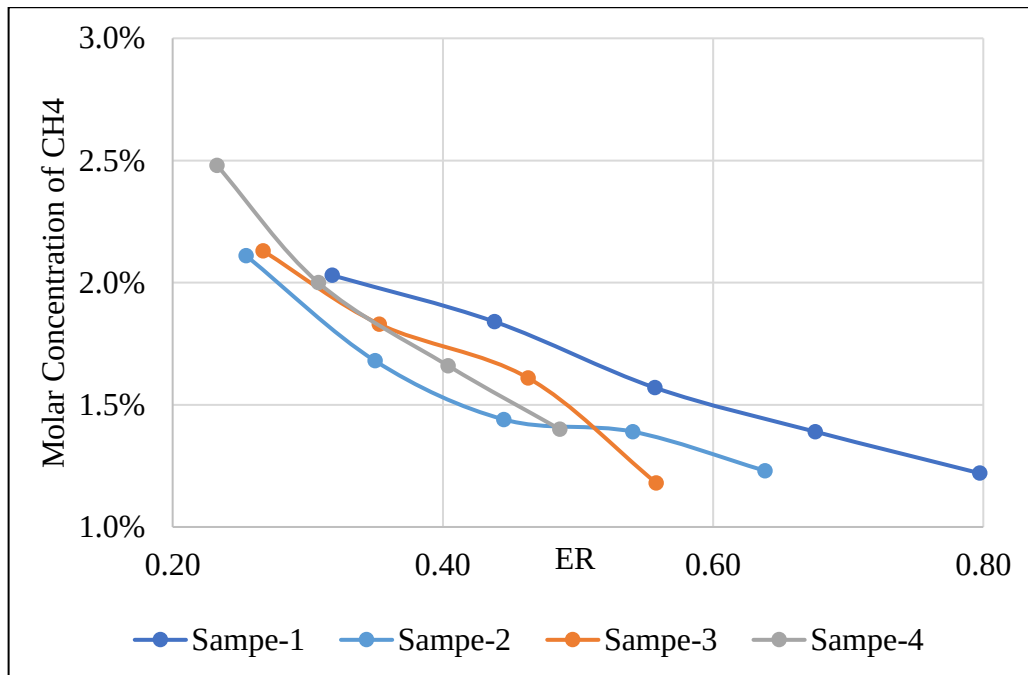


**Figure A.2** Comparison of the experimental results of molar concentration of CH<sub>4</sub> in syngas

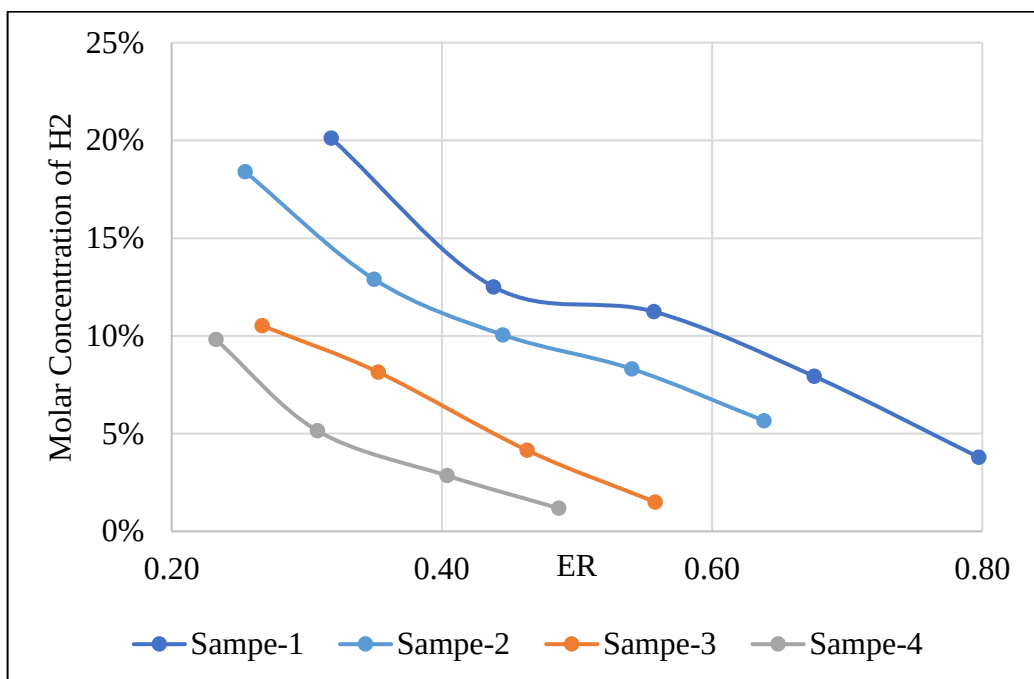


**Figure A.3** Comparison of the experimental results of molar concentration of H<sub>2</sub> in syngas

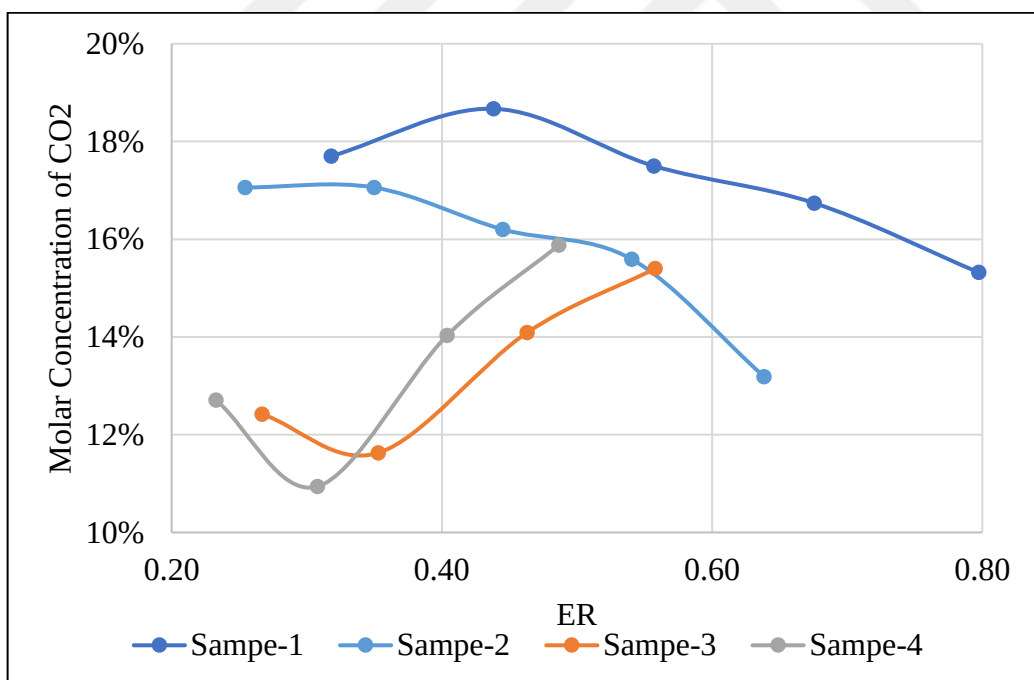


**Figure A.4** Comparison of the experimental results of molar concentration of CO<sub>2</sub> in syngas

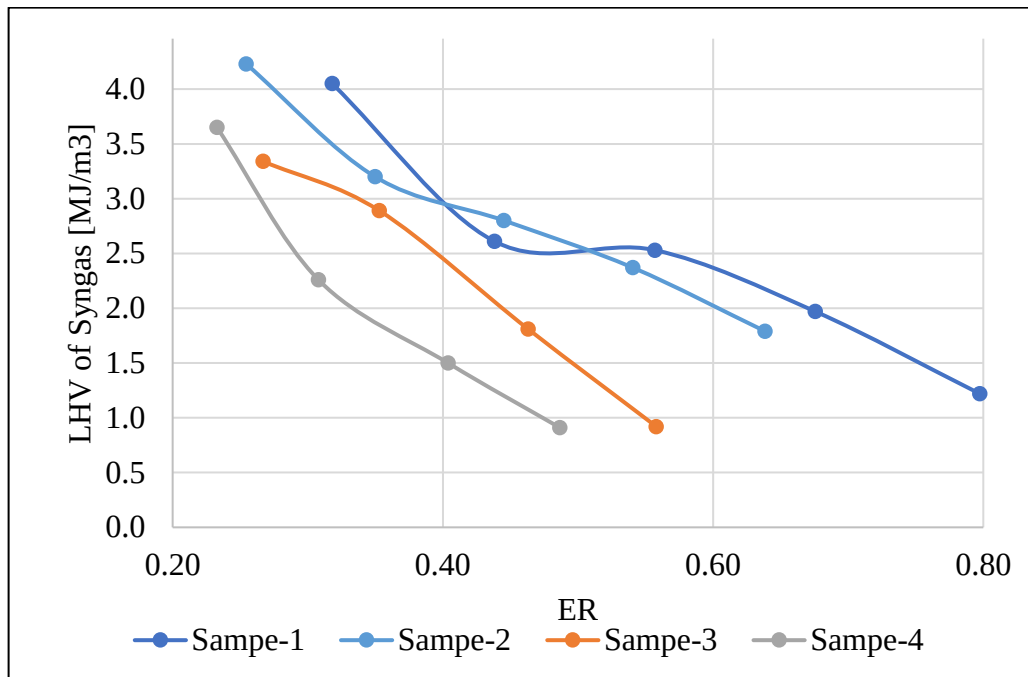


**Figure A.5** Comparison of the experimental results of syngas LHV for different coal samples and various ER values

## APPENDIX B – Contour Plots Obtained From The Numerical Studies At The Last Time Step For Sample-3

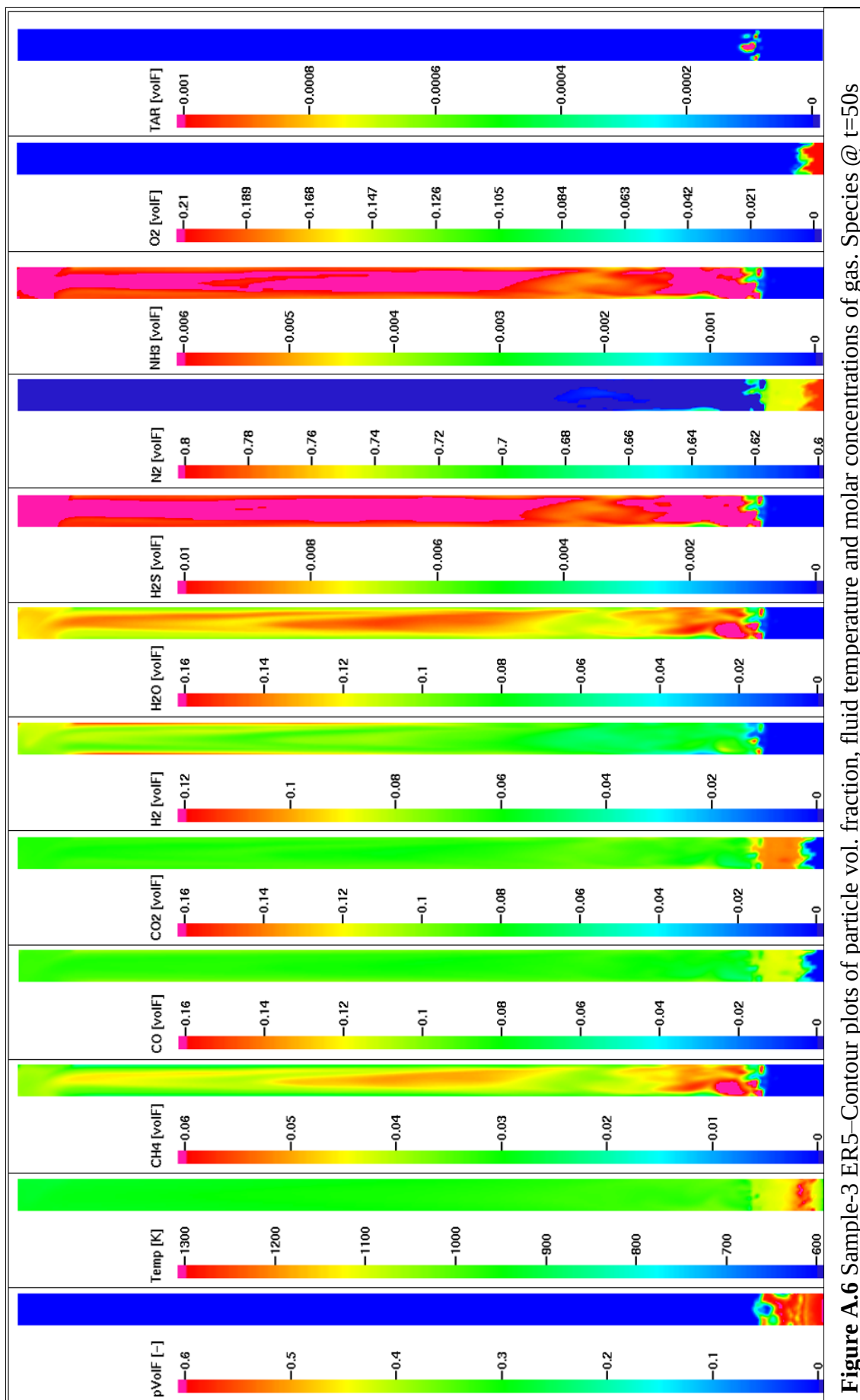


**Figure A.6** Sample-3 ER5—Contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas. Species @  $t=50s$

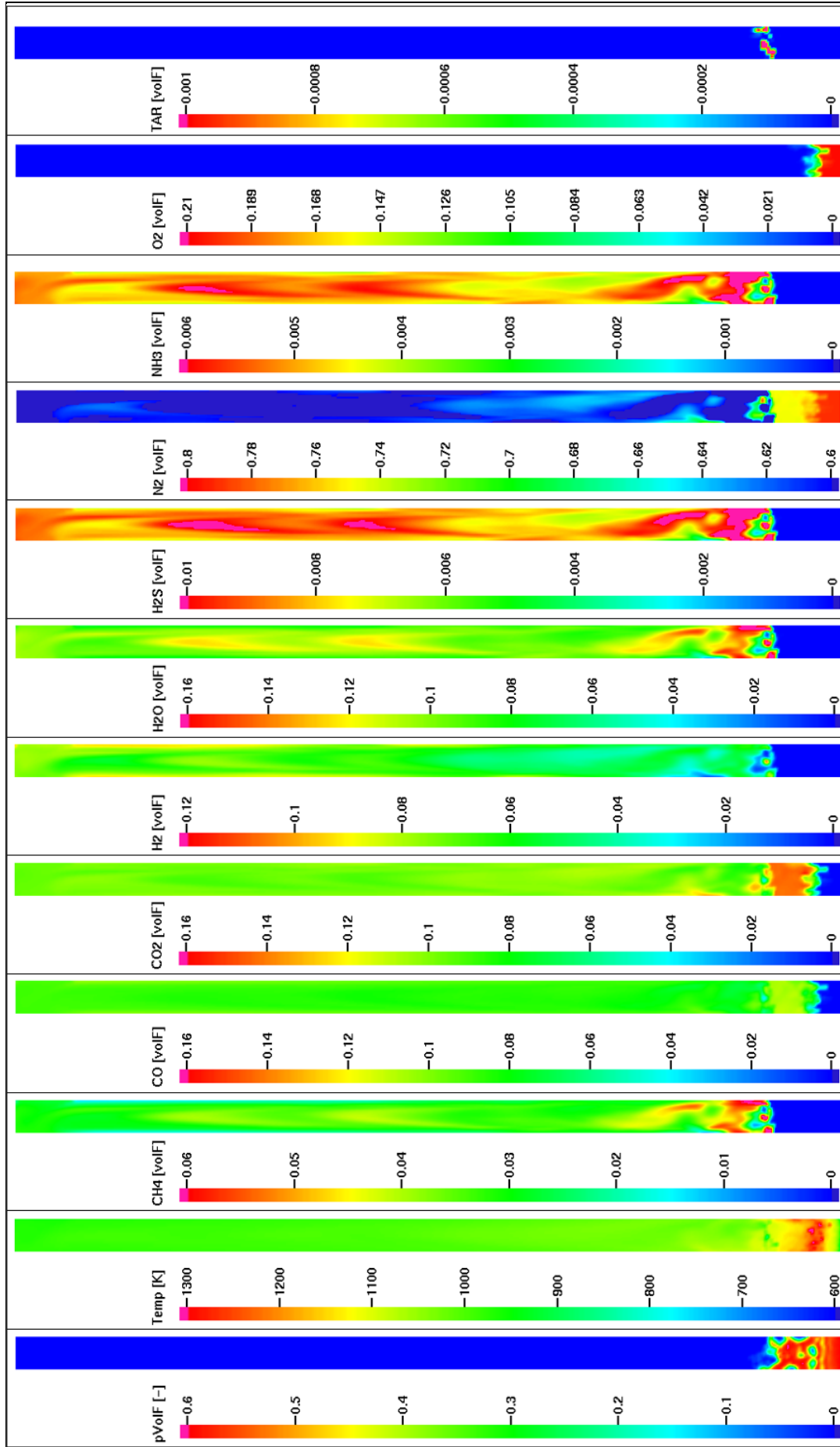


**Figure A.7** Sample-3 ER4–Contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas. Species @  $t=50s$

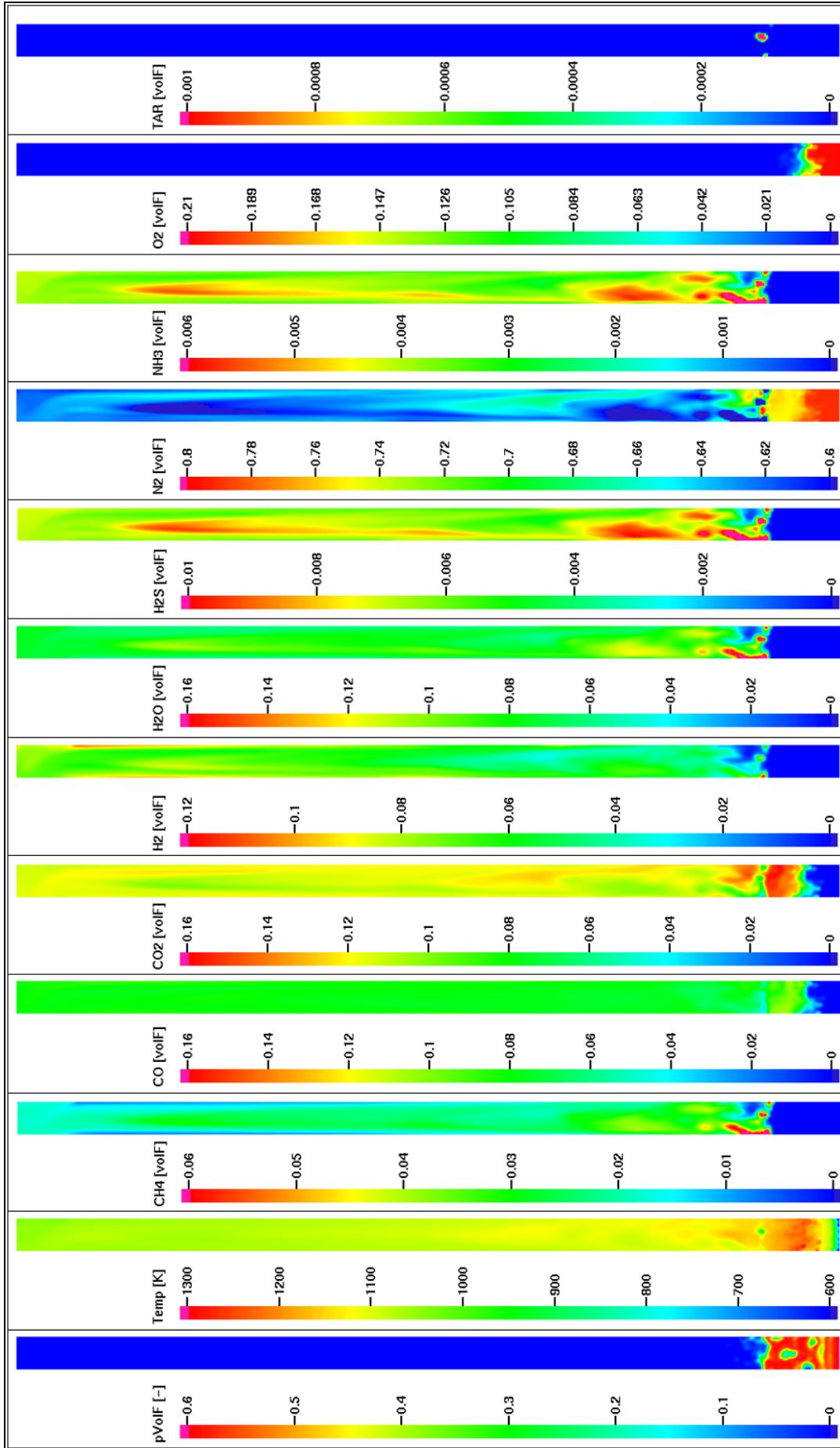


**Figure A.8** Sample-3 ER3—Contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas. Species @  $t=50s$

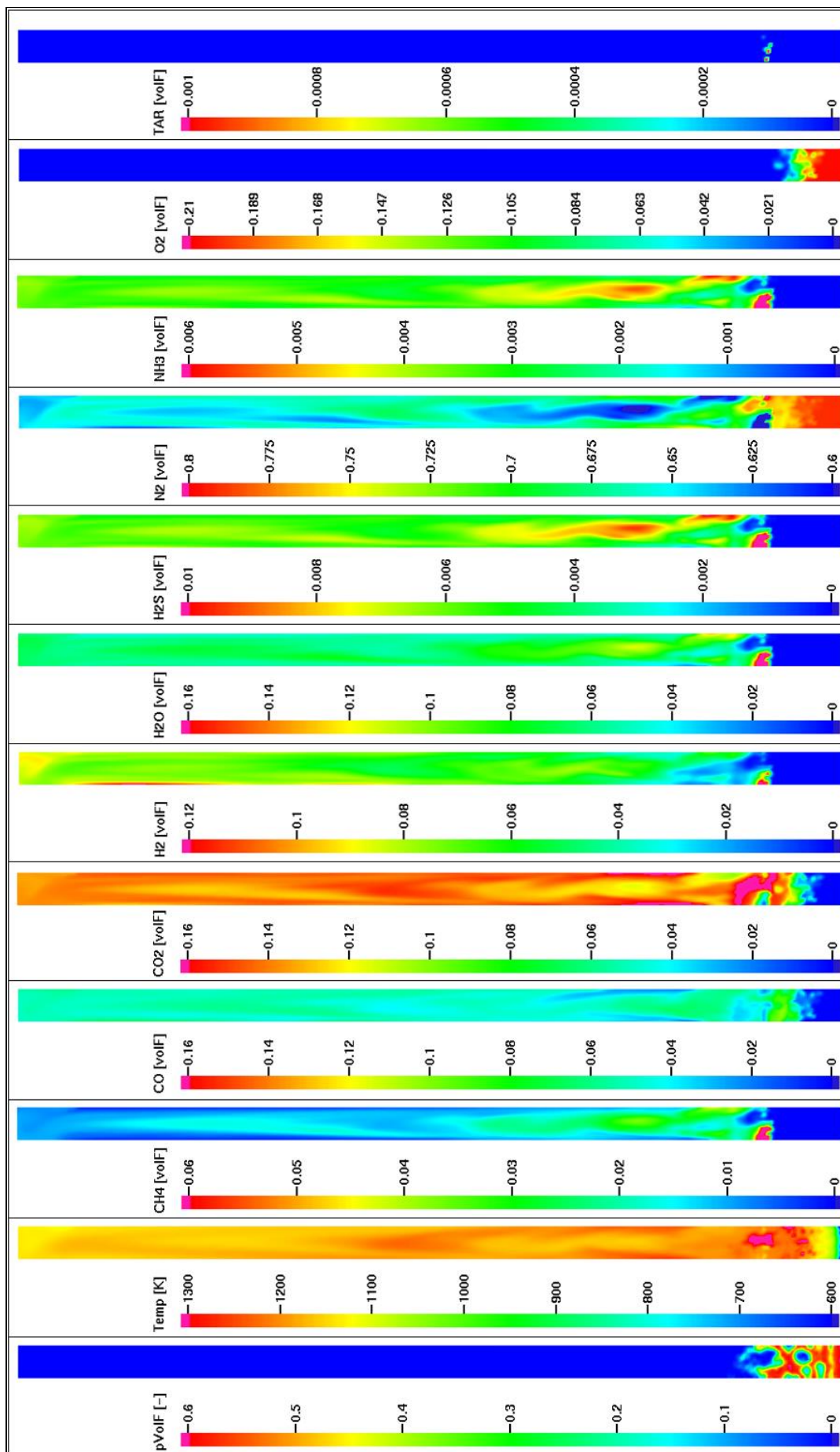


**Figure A.9** Sample-3 ER2–Contour plots of particle vol. fraction, fluid temperature and molar concentrations of gas. Species @  $t=50s$

## APPENDIX C - Time Dependent DTF Mole fractions at the Reactor Exit for Sample-3

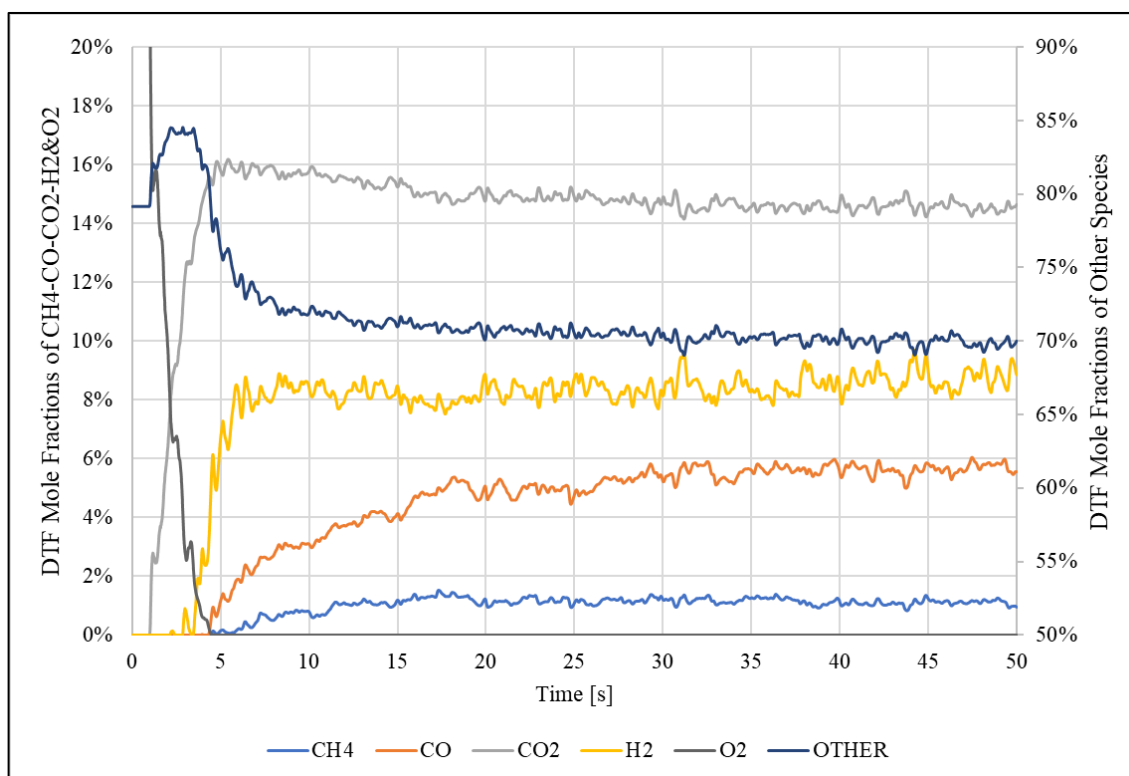


**Figure A.10** Sample-3 ER2–DTF Mole fractions

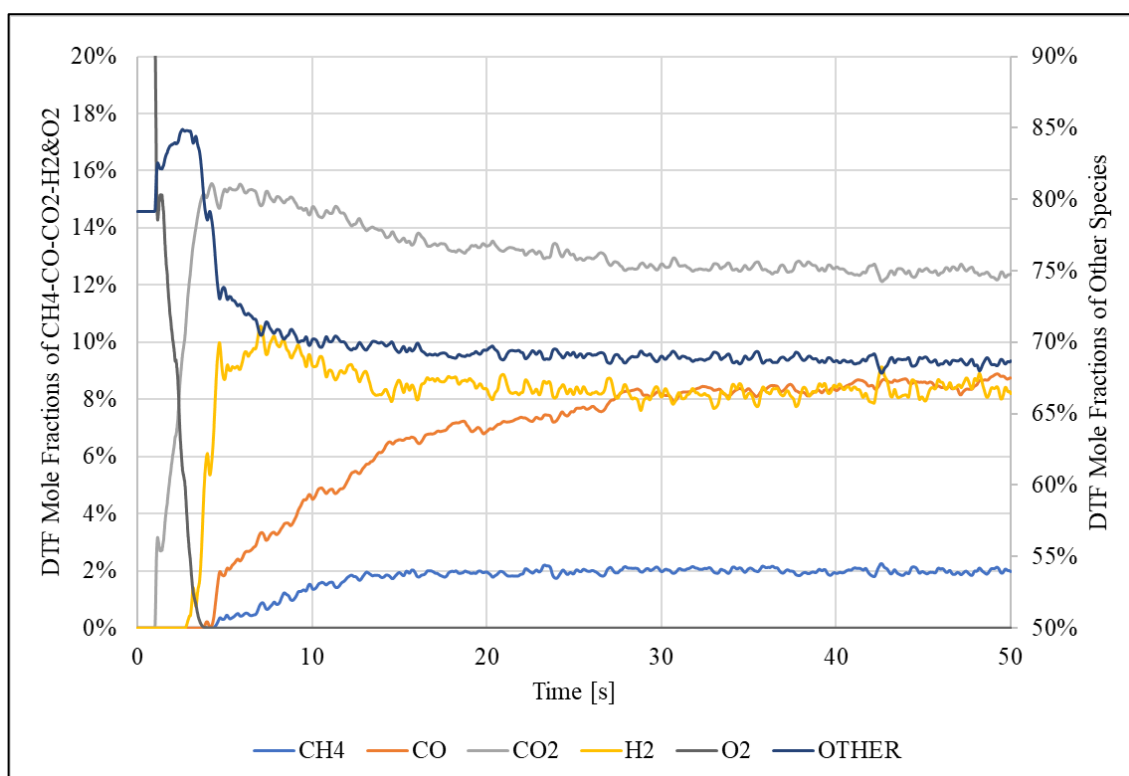


**Figure A.11** Sample-3 ER3–DTF Mole fractions

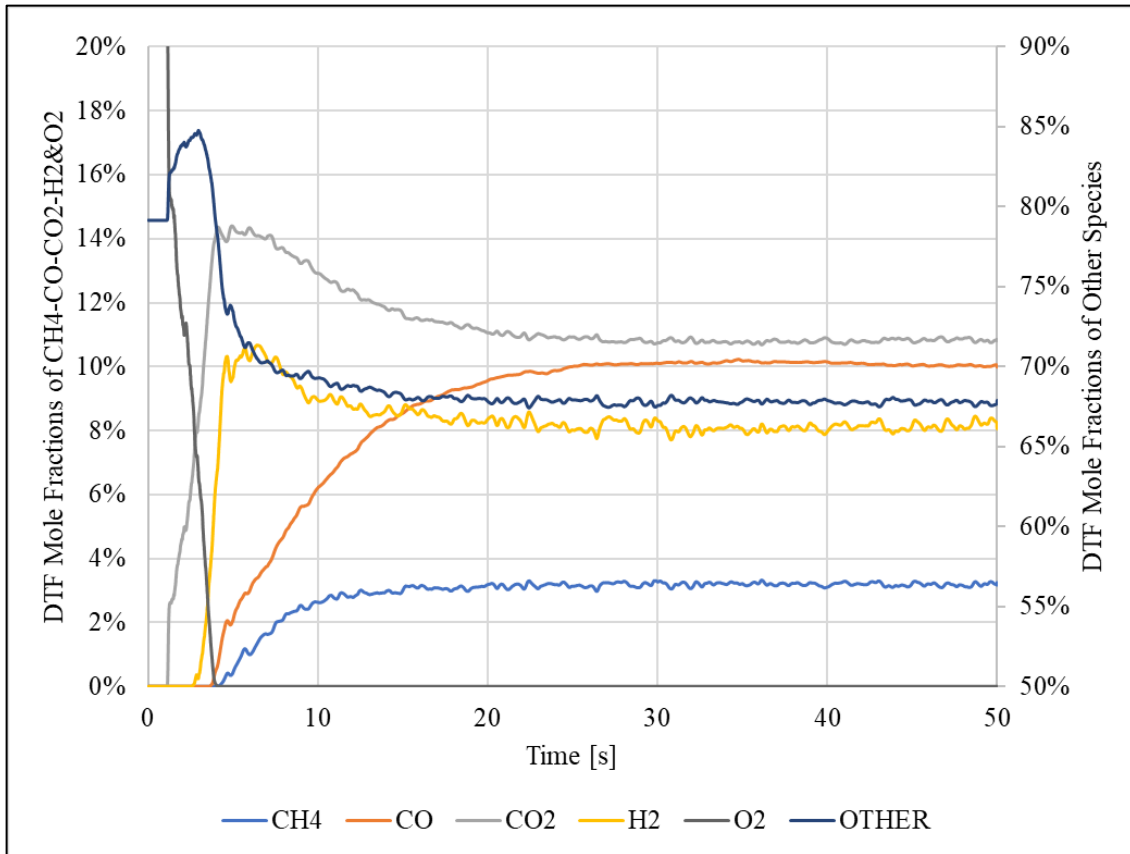


**Figure A.12** Sample-3 ER4–DTF Mole fractions

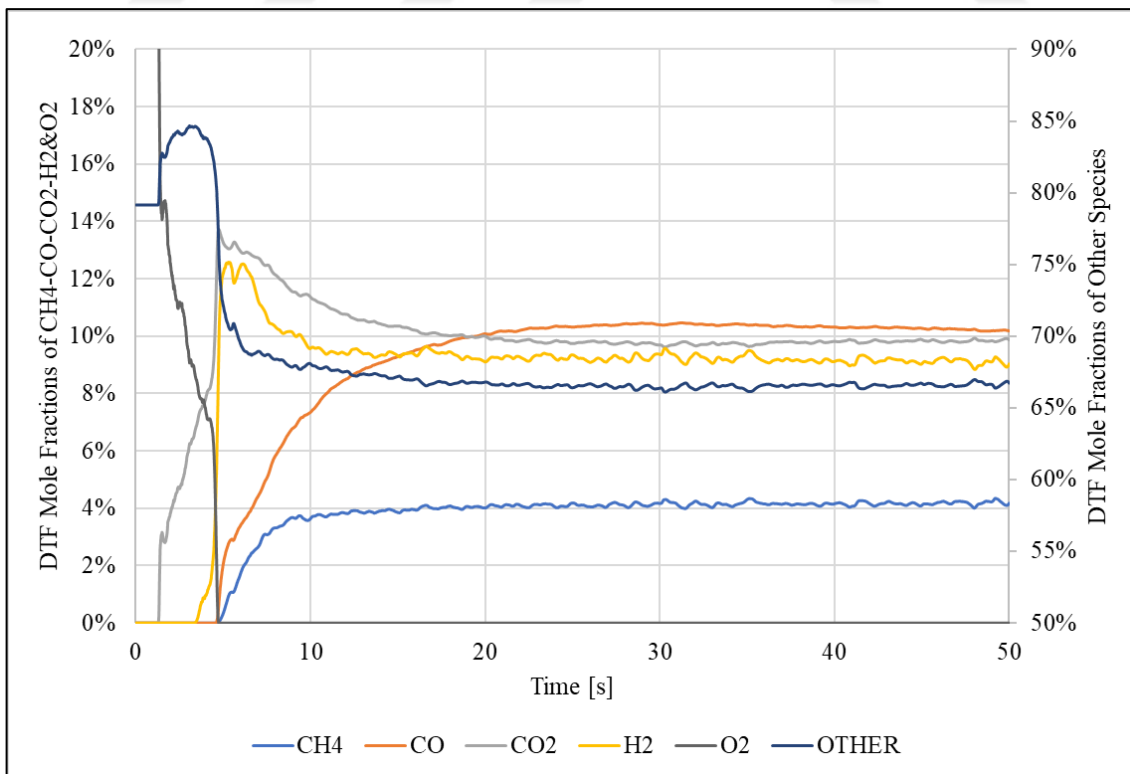


**Figure A.13** Sample-3 ER5–DTF Mole fractions

**APPENDIX D – One-dimensional Heat Transfer Calculations for Insulation Design of CFBG system**

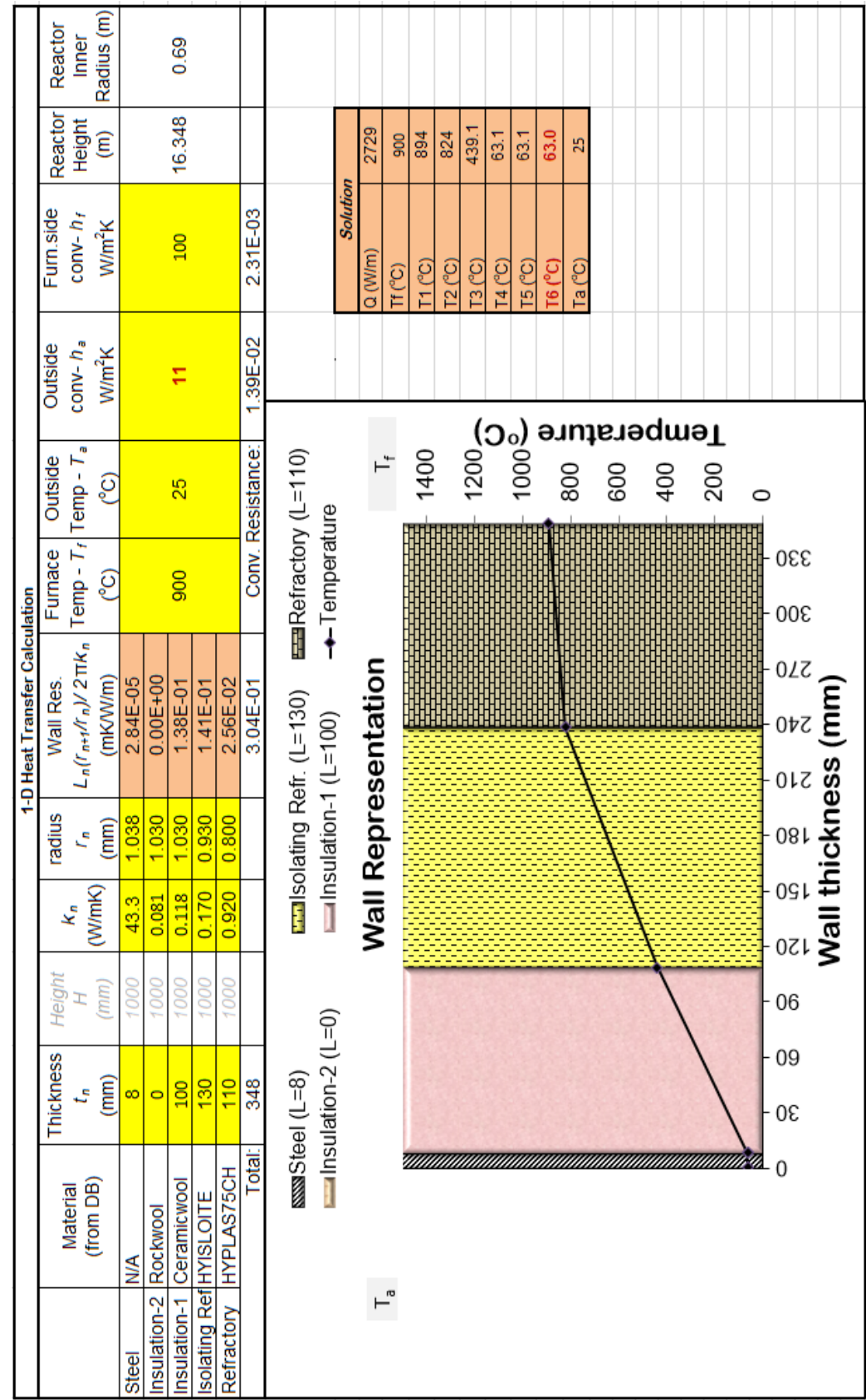


**Figure A.14** 1D Heat transfer calculations for insulation design of the reactor and gasification system components

## APPENDIX E – Assembly Details of the Industrial-Scale CFBG

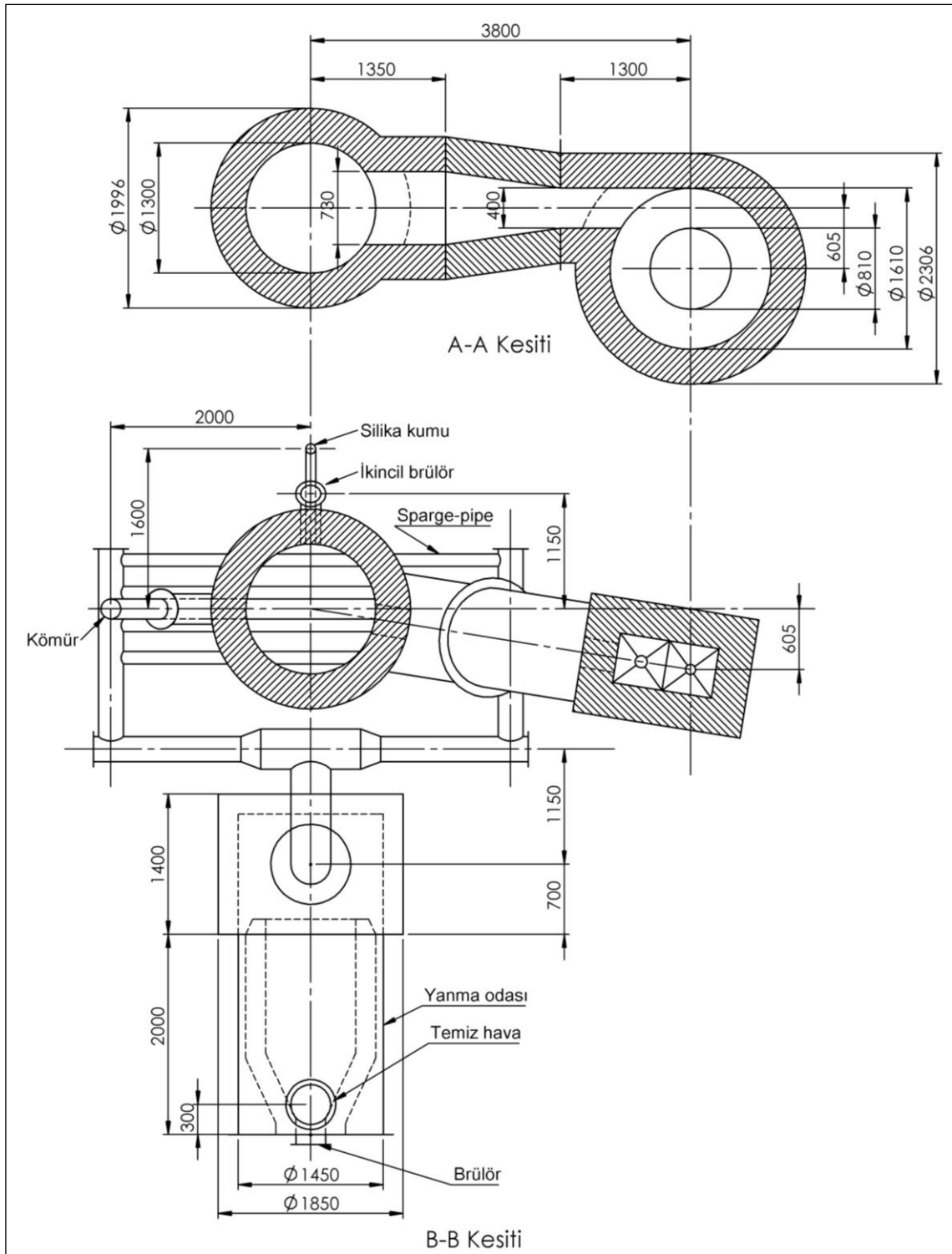


**Figure A.15** Technical drawing of the assembly of the system components – Top View

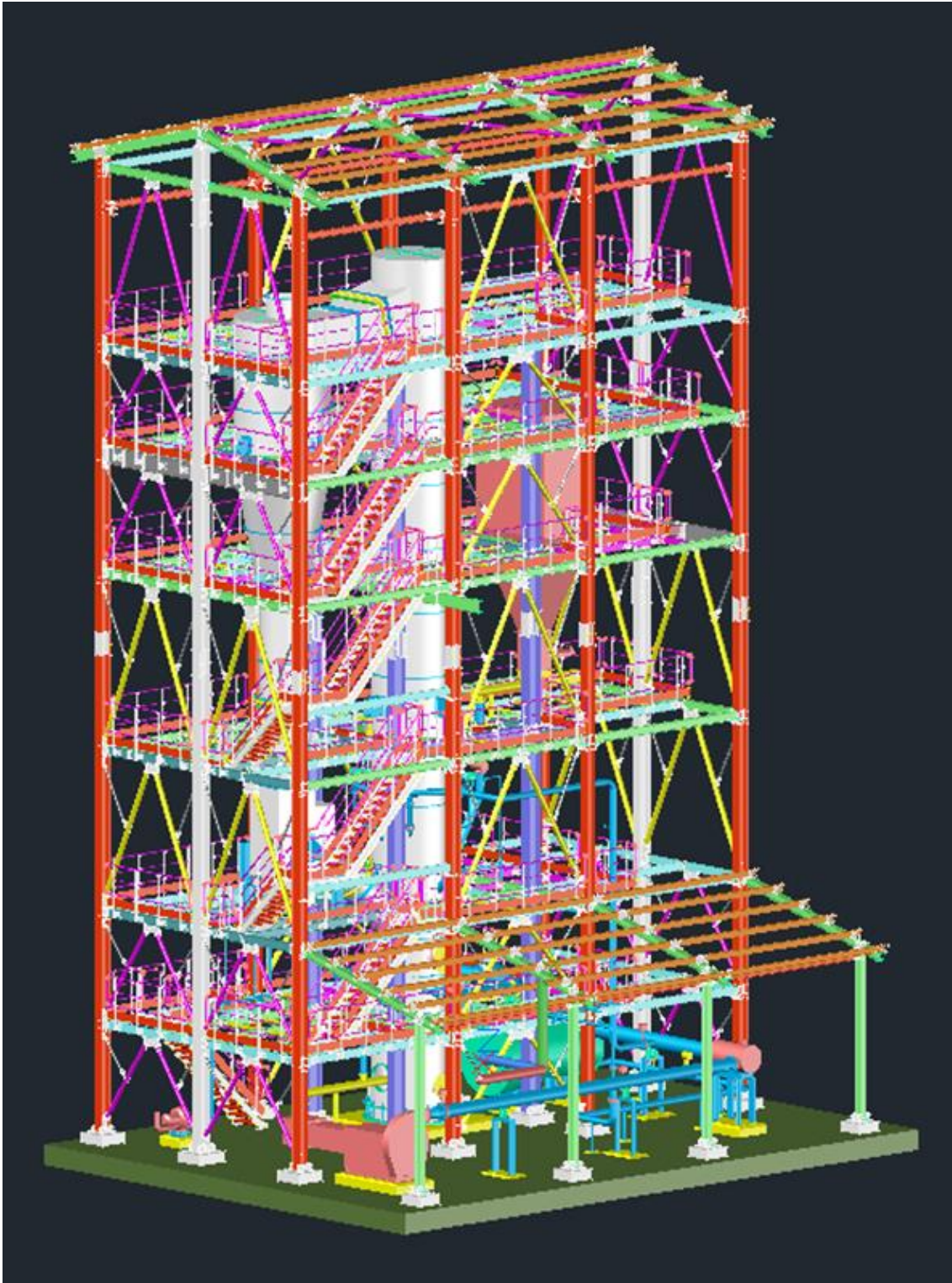


**Figure A.16** 3D layout of the whole CFBG system

**APPENDIX F - Transient Particle Accumulation Height Data in the Standpipe for All Cases Obtained from the Numerical Analysis of Non-Reactive Isothermal Hydrodynamics of CFBG**

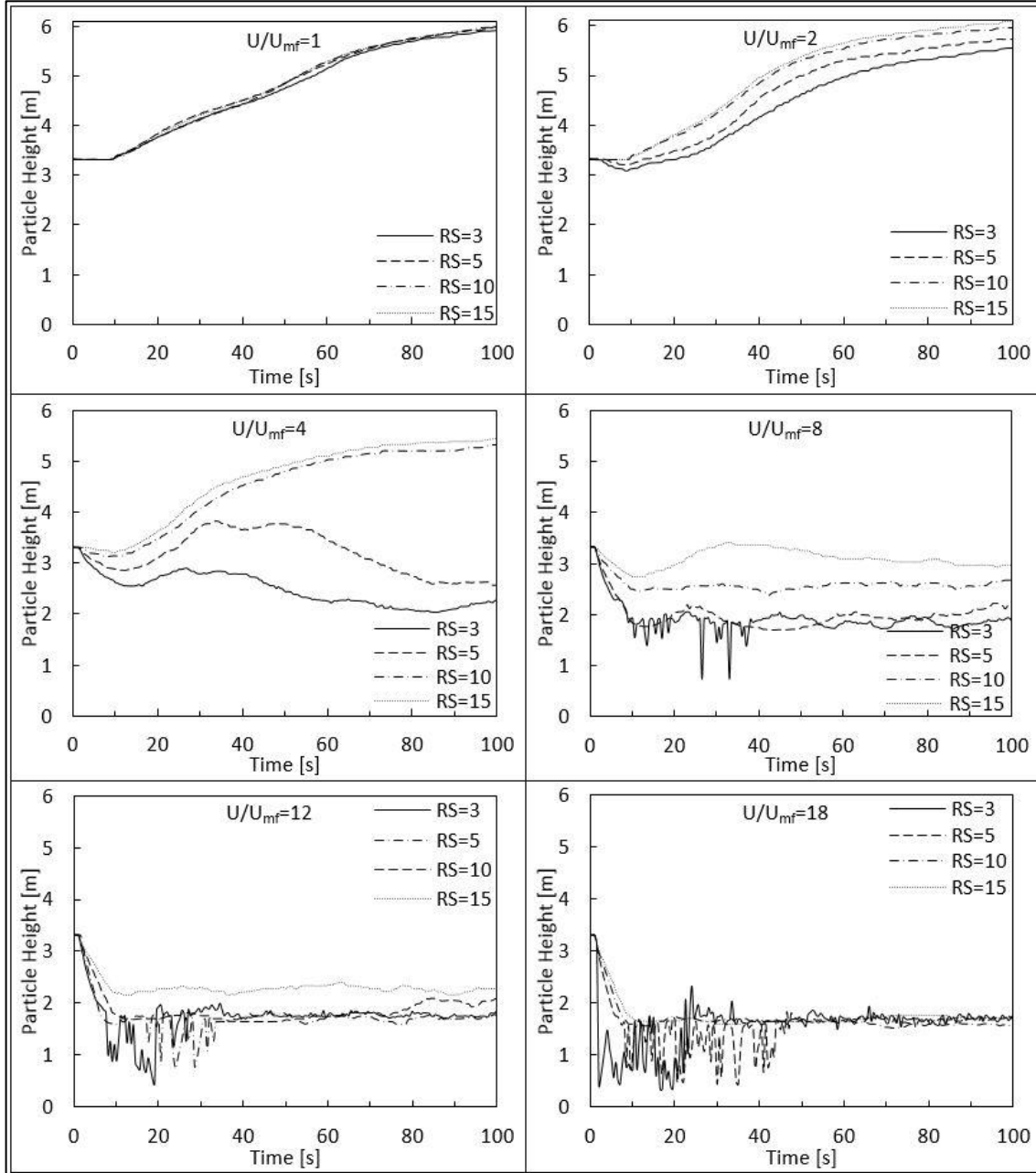


**Figure A.17** Time-dependent particle height for isothermal analysis of industrial scale CFBG

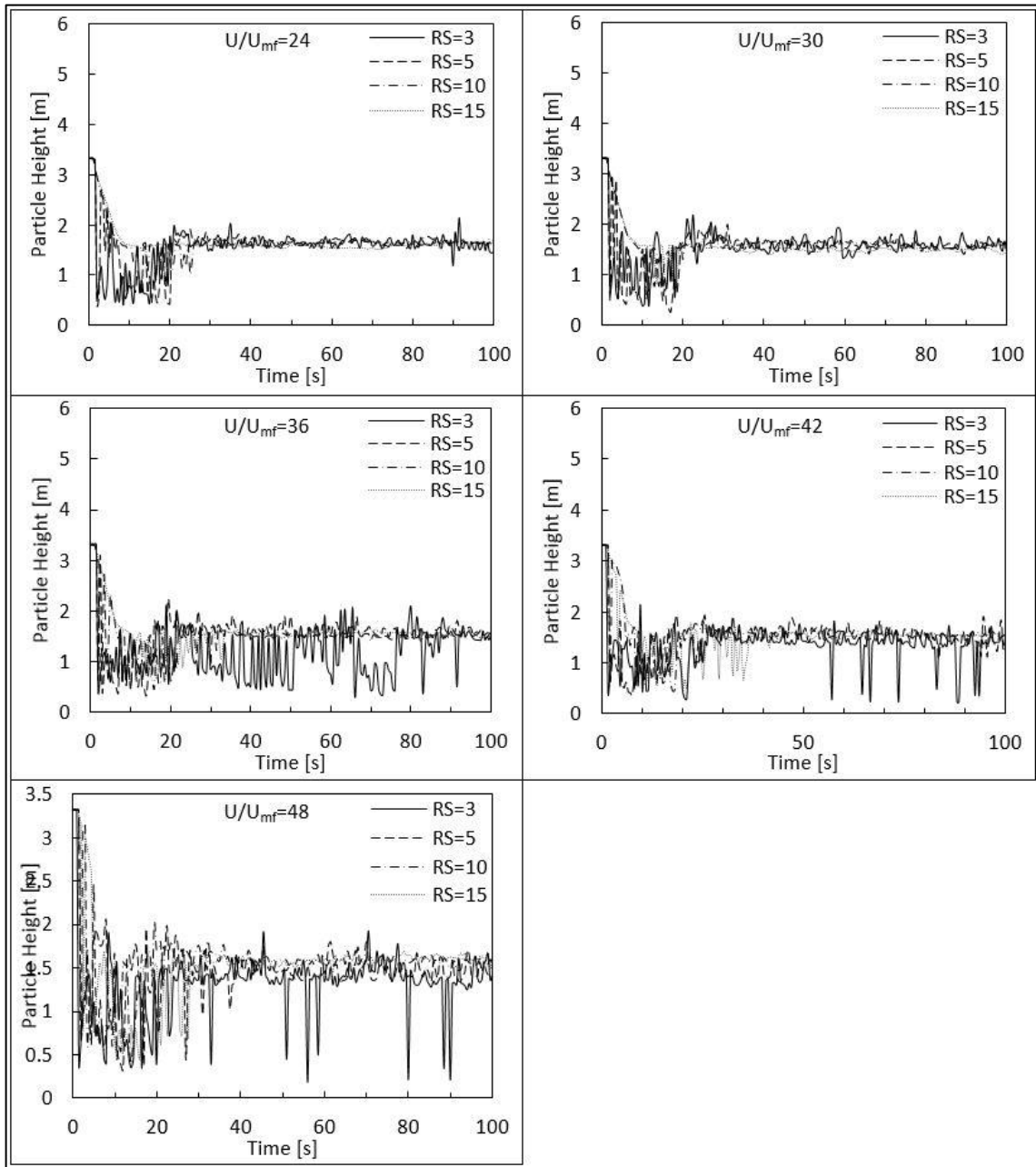


**Figure A.17(Cont.)** Time-dependent particle height for isothermal analysis of industrial scale CFBG

# ÖZGEÇMİŞ

## YALÇIN YÜKSELENTÜRK

### Eğitim Bilgileri

|           |                                                                        |
|-----------|------------------------------------------------------------------------|
| 2011-2022 | Doktora: Marmara Üniversitesi, Makine Mühendisliği                     |
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| 2001-2007 | Lisans: İstanbul Teknik Üniversitesi, Makine Mühendisliği              |
| 1998-2001 | Lise: Eskişehir Fen Lisesi                                             |

### İş Deneyimleri

|            |                                                                 |
|------------|-----------------------------------------------------------------|
| 2021-Halen | Uzman Mühendis, TUSAŞ Motor Sanayi A.Ş. (TEİ)                   |
| 2018-2021  | Ar-Ge Yöneticisi, Radim Radyatör LTD. ŞTİ.                      |
| 2013-2018  | Ar-Ge Mühendisi, Bersey Kazan LTD. ŞTİ:                         |
| 2011-2013  | CFD uzmanı, Marmara Üniversitesi Makine Mühendisliği Bölümü:    |
| 2008-2011  | Bursiyer Araştırmacı, Marmara Üniversitesi TUBİAK-1001 projesi: |

### Bilgisayar Deneyimi

Programlama dilleri: C, Fortran, Matlab

Çözüm ağı oluşturma: Gambit, ICEM CFD, Gridgen

CAE yazılımları: Star-CD, Star-CCM, ANSYS, Fluent, CFX, Es-ICE, CPFD Barracuda, Solidworks Flow Simulation, Solidworks Simulation

CAD: Solidworks, CATIA, AutoCAD

Diğer mühendislik araçları: ModFrontier (Optimizasyon), GT-Power (1B İY- Motor analizi), Ensight (Ardıl-İşleme)

### Akademik Çalışmalar ve Endüstriyel Projeler

2008-2011 **ARDEB 1001** “Determination of design parameters of a HCCI type diesel engine by using computational fluid dynamics methods”

2010-2011 “Dairesel ve Dairesel olmayan Jet Akım Alanlarının Deneysel ve Sayısal Olarak İncelemesi” (YL Tez Çalışması)

2012-2015 **TEYDEB 1511** “Design of a 10MWth capacity Circulating Fluidized Bed Gasifier for low rank coals”

2012-2015 **TEYDEB 1511** “Design of a low NOx industrial pulverized coal burner”

2019-2021 **TEYDEB-1501** “Alüminyum radyatörlerde homojen yüzey sıcaklığının sağlanması için alternatif su kanalı tasarımı”

## **Yayınlar**

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## **Yabancı Dil**

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