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Author(s) Hannula, Ilkka; Kurkela, Esa
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A semi-empirical model for pressurised air-blown fluidised-bed gasification of biomass

Ilkka Hannula* and Esa Kurkela

Technical Research Centre of Finland, P.O. Box 1000, FI-02044 VTT

Abstract

A process model for pressurised fluidised-bed gasification of biomass was developed using Aspen Plus simulation software. Eight main blocks were used to model the fluidised-bed gasifier, complemented with FORTRAN subroutines nested in the programme to simulate hydrocarbon and NH₃ formation as well as carbon conversion. The model was validated with experimental data derived from a PDU-scale test rig operated with various types of biomass. The model was shown to be suitable for simulating the gasification of pine sawdust, pine and eucalyptus chips as well as forest residues, but not for pine bark or wheat straw.

Keywords: gasification, biomass, fluidised-bed, model, Aspen Plus

1 INTRODUCTION

1.1 Biomass gasification

Global climate change together with increasing energy prices and depleting fossil resources have provoked major interest towards renewable forms of energy and resources. Gasification of biomass offers an efficient way to utilise renewable carbonaceous feedstocks and has significant commercial and environmental potential in the production of green chemicals, synthetic fuels and electricity.

* Corresponding author.

Address: Technical Research Centre of Finland, P.O. Box 1000, FI-02044 VTT

Tel. +358 20 722 6370

Fax +358 20 722 7048

E-mail address: ilkka.hannula@vtt.fi.

Gasification produces a gas mixture rich in carbon monoxide and hydrogen. Other major compounds include carbon dioxide, nitrogen, water, methane and a rich spectrum of hydrocarbons. A general objective of gasification is to try to maximise the yields of gaseous products and minimise the amounts of condensable hydrocarbons and unreacted char. Exact composition of product gas depends on the type of process feeds, their feed ratios, process parameters and the type of gasification reactor used.

In contrast to coal gasification, where char gasification reactions contribute most to the overall yield, in biomass gasification the devolatilisation stage and the secondary reactions of primary pyrolysis product play the major role (Kurkela 1996).

1.2 Modelling of biomass gasification

The objective of process modelling is to construct a mathematical description of a process that can be used to predict reactor temperature and outlet concentrations from inlet flows and operating conditions. A model that fits well to the experimental data can help to reveal major trends in a multivariable system and be a great comfort when an engineer is faced with scaling-up a reactor to produce the full-scale design (Rose 1982). A suitable model also permits more efficient control of the reactor and offers a safe way to simulate reactor behaviour in continuous and transient conditions (Buekens, Schoeters 1984).

Mathematical models of fluidised-bed gasifiers are usually based either on kinetic rates or thermodynamic equilibrium.

Models based on rates attempt to predict product gas concentrations by combining a hydrodynamic model of the fluidised-bed with appropriate kinetic schemes for the

heterogeneous and homogeneous processes occurring inside the gasifier (Gururajan et al. 1992). However, as a large number of dynamic parameters involved in fluidised-bed gasification are presently unknown and very difficult to measure, estimation of product gas composition through kinetic models often becomes exceedingly difficult (Kovacik et al. 1990).

A model can also be constructed by applying the principles of chemical equilibrium. In this approach, the complex kinetics can be disregarded by assuming that gasification reactions occur fast enough for them to reach equilibrium. However, it has been widely reported that for fluidised-bed gasifiers, product gas compositions are not in equilibrium, possibly due to the slow kinetics involved (Schuster et al. 2001). Kilpinen et al. (1991) has shown that for CO, CO₂, H₂, and H₂O the equilibrium seems to be established under certain assumptions, whereas the amounts of solid carbon, methane, HCN and NH₃ are underpredicted. In this work, experimental data is used to take account of these above mentioned conversions, which would otherwise be estimated wrong by equilibrium approach.

Despite their limitations, equilibrium models have been widely published in the literature. Gururajan et al. (1992) have critically examined several simulation models proposed for fluidised-bed gasification of coal. The work done in fluidised-bed gasification of biomass has been more limited and described in some detail by Schuster et al. (2001).

De Kam et al. (2009) have recently modelled gasification with an Aspen Plus RGibbs reactor by separately specifying a set of reactions with temperature approach to equilibrium, and by fixing the production of certain species based on the amount of fuel

being used. However, data about validation results was not reported.

Doherty et al. (2009) divided the gasifier into six separate blocks to cater for drying, pyrolysis, partial oxidation and gasification reactions. The outcomes of these processes were then fed to equilibrium reactor where final composition of the syngas was formed under restricted conditions. The final block was used to separate and recycle solids entrained in the gas, thus simulating a CFB cyclone. The validation of the model was performed for three test runs and the results were reported to be in good agreement with experimental data, with the exception of overpredicted methane. Heavier hydrocarbons were not considered in the model.

The approach of Nikoo and Mahinpey (2008) was to divide the gasifier to decomposition of the feed, volatile reactions, char gasification and gas-solid separation. In addition, the effect of hydrodynamic parameters and reaction kinetics of biomass gasification in fluidised-beds were simulated with FORTRAN codes. This slightly more complex approach did not seem to result in much improved predictions, probably due to the inaccurate methane estimations and the absence of higher hydrocarbons in the model.

2 Materials and methods

2.1 Experimental work

In this work, a model for pressurised air-blown fluidised-bed gasifier using biomass as a feedstock is developed using Aspen Plus simulation software.

The model is fitted with experimental data originally derived from fluidised-bed air gasification studies with pine sawdust in 1991 – 1992. The testing was conducted in a

VTT's PDU-scale test rig as a part of the National Combustion Programme LIEKKI, and was aimed to support the development of simplified integrated gasification combined-cycle process. The results of these sawdust gasification experiments are published and summarised by Kurkela et al. (1993). A brief description of the process and the gasification experiments used to fit the parameters of the model is given in the following paragraphs.

2.2 Description of the experimental equipment and arrangement

The heart of the pressurised fluidised-bed gasification test rig is a refractory-lined reactor with a bed diameter of 15cm and freeboard diameter of 25cm. The height from the air distributor to the gas outlet pipe is 4.2m. Typical gas phase residence times range from 5 to 8s depending on the fluidising velocity.

Primary air and a small amount of steam are introduced into the reactor through a multiorifice plate distributor. Two different distributor plates can be used depending on the required range of fluidising air flow rate. The first plate is a 10mm thick slightly conical plate with 21mm holes and an open area of 0.62% of the reactor cross-sectional area. The other air distributor is a 10mm thick horizontal plate with 2mm holes and a total open area of 1.7% of the reactor area. Bottom ash is removed through a 38mm(id) pipe located in the centre of the distributor plate.

Secondary air can be introduced above the fluidised-bed through two pipes which both have eight air nozzles. The heights from the air distributor to the secondary air injection ports are 1.5 and 1.9m. The locations of the measuring and sampling points are shown in Figure 1. The test facility is equipped with a wide variety of process

measurements, which are collected to a data logger and processed by a microcomputer. The product gas outlet temperature used in the model was measured with thermocouple t_{13} also visible in Figure 1.

2.3 Description of the data used in parameter fitting

The data used to fit the parameters of the gasification model is described briefly in this paragraph. Most of the data is from two one-week test runs, dedicated to sawdust gasification.

The measurements with pure saw dust were carried out in 10 different operating variable sets. The length of different set points varied from 2 to 9 hours. During the set point periods the feed rates of fuel, air and steam were kept as constant as possible. At some set points, however, changes in fuel quality had to be compensated for by small ($\pm 5\%$) changes in feed rate. All process data were recorded by a computer at 3 – 5 minutes intervals, and all discharged cyclone and filter fines as well as bottom ashes were collected, weighed and samples at the set points. The fuel was also weighed and sampled before charging it into the feeding system.

After the test run a material balance was calculated for each of the set points, based on the average values of the data. Hydrogen and nitrogen balances were used to calculate the water vapour content of gas and raw gas flow rate, which are difficult to measure with the same accuracy as the other measurements. The closure of carbon and oxygen balances (out/in) at qualified set points were within 5%, but the ash balance was worse, since the ash content of sawdust was very low and part of the fine filter dust was lost in the depressurisation of the dust removal hopper of the ceramic filter unit.

A summary of the key operational parameters at different set points is shown in Table 1. Set points 1 – 7 were run with sawdust A (SD A) while sawdust B (SD B) was used at set points 8 – 10. The ultimate and proximate analyses of the feedstock are presented in Table 2.

The main variable in the tests was gasification temperature, controlled by changes in air-to-fuel ratio.

2.4 The utilisation of experimental data in the model

The limitations of equilibrium approach, as summarised in paragraph 1.2, were dealt in this work by fitting a selection of parameters to experimental data. The intention was to construct a simple and generic model for gasification of biomass that could be fitted to match a specific gasification reactor using easily measurable empiric correlations. For this reason the incorporation of such parameters as feedstock particle size and reactor geometry were decided to be ignored, as their inclusion would lead to a need of a more complex model, still not necessarily able to generate more accurate predictions. The selection of the approach stemmed from the lack of complete understanding about the kinetic and hydrodynamic phenomena pertaining to fluidised-bed gasification of biomass as discussed in paragraph 1.2.

Although the experimental data used in this work has been published already a while ago, to our knowledge, it has not yet been used for validation of a gasification model based on thermodynamical equilibrium approach. However, a model for bubbling fluidised bed, incorporating bed and freeboard hydrodynamics, fuel drying, devolatilisation and chemical reaction kinetics has been published by Hamel (2001) and

validated with the same data from Kurkela (1993) with seemingly good, although narrowly reported results. The aim of this work was to develop a considerably simpler model, with the ability to yield equally good results.

2.5 Process scheme

Eight main blocks were used to model the fluidised-bed gasifier, complemented with FORTRAN subroutines nested in the programme to simulate carbon conversion, as well as NH_3 and hydrocarbon formation. All calculation blocks were thermally integrated in order to represent a single gasification reactor.

The core of the model is the equilibrium reactor block (RGibbs), where major part of the feed is converted to gasification products according to equilibrium approach. Almost all the other blocks included in the model are used to cater for the non-equilibrium behaviour perceived in real life gasifiers. These phenomena consist of incomplete carbon conversion as well as formation of hydrocarbons and nitrogen species. The division of the model to separate blocks could also have been executed differently, as the model examples of paragraph 1.2 imply. However, it was considered more rational to handle each non-equilibrium phenomenon in a separate block, rather than treating them in a one RGibbs block by restricting the equilibrium of the reactor.

The main structure of the model is illustrated schematically in Figure 2. As a first step of the simulation, biomass is decomposed to hydrogen, oxygen, nitrogen, carbon, sulphur and ash, based on the ultimate analysis of the feedstock. Then all of the ash is separated to the ash outlet, followed by the modelling of carbon conversion by extracting a certain amount of elemental carbon to an outlet stream. As the

hydrocarbons are largely formed from the volatile components of the biomass, the feed is first divided into two separate streams of volatiles and fixed carbon. The volatiles are then led to a simulation block where parts of the stream are converted into hydrocarbons according to experimental data. The stream of fixed carbon is mixed with air, steam and unreacted volatiles and converted to gasification products according to thermodynamic equilibrium. As the last step of the simulation, hydrocarbons and gasification products are mixed together to form the final product. More detailed description of the main unit operations of the simulation is given in the following paragraphs.

2.5.1 Biomass decomposition

A yield reactor (Ryield) was used to simulate the decomposition of the feed. In block number 1, biomass was converted to hydrogen, oxygen, nitrogen, carbon, sulphur and ash by specifying the yield composition based on ultimate analysis of the feedstock.

2.5.2 Ash removal

In the block number 2, ash removal was simulated with component separator by removing all the feedstock ash to the ash outlet.

2.5.3 Carbon conversion

According to equilibrium, all of the feedstock's carbon should exist in gas-phase under typical gasification conditions. However, a significant amount of carbon is usually found from the bottom and fly ash of an air-blown fluidised-bed reactor. To overcome this discrepancy, a FORTRAN equation was created to represent observed correlation between carbon conversion and gasification air ratio (See Figure 3a). The correlation

(See Table 3) was nested in block number 3 and used to calculate the amount of elemental carbon that has to be extracted from the feed to simulate the incomplete conversion. The air ratio (E) and carbon conversion (η_c) were defined, respectively, as:

$$E = \frac{m_O / m_B}{m_{St.O} / m_B}, \quad (1)$$

where m_O is the weight of used oxygen

m_B weight of biomass

$m_{St.O}$ weight of stoichiometric oxygen

$$\eta_c = \frac{C_{gas} + C_{tar}}{C_{fuel}}, \quad (2)$$

where C_{gas} is the carbon output in dry gas (g/s)

C_{tar} carbon output in tar (g/s)

C_{fuel} carbon input in fuel (g/s).

2.5.4 Separation I

In block number 4, the feed was separated to streams of fixed carbon and volatiles according to proximate feedstock analysis. In reality the yields of char and volatiles depend also from particle size, heating rate and other parameters. However, these factors were not considered in the model.

2.5.5 Hydrocarbon formation

As already mentioned, the equilibrium approach also underpredicts the amounts of hydrocarbon and nitrogen components. To tackle this shortage in the approach, a FORTRAN subroutine was nested in block 5 (Rstoic) to calculate the conversions based

on experimental data illustrated in Figure 4a,b,c,d. CH₄, C₂H₂, C₂H₄, C₂H₆ and NH₃ were chosen to be included in the model as they represent the most voluminous hydrocarbon and nitrogen compounds in biomass-derived product gas. These experimentally observed correlations (See Table 3) were used in the model to calculate the fractional conversion of elemental carbon in the stream 'Volatiles' separately for each component.

All the curves were drawn as a function of air ratio, as Kurkela et al. (1993) have shown that it correlates well with the average freeboard temperature of a fluidised-bed gasifier (See Figure 3b) and as according to Bruinsma and Moulijn (1988) as well as Simell et al. (1992) the total concentration of tar from fluidised-bed gasifier depends mainly on gasification temperature and the type of feedstock.

2.5.6 Separation II

In the block number 6, the hydrocarbons were separated from the stream to a bypass using an Aspen Plus component separator and the remainder of the stream was directed into the gasification block. This was necessary to prevent the NH₃ and hydrocarbons from decomposing in block 7.

2.5.7 Gasification

In the block 7, a Gibbs reactor (Rgibbs) was used to mix the feed with streams of air, steam, purge nitrogen and remaining volatiles from block 5, and to convert them to equilibrium products. Nitrogen was added to simulate purge nitrogen, originally used in the test rig to seal off leakages and to keep the measurement equipment operational.

2.5.8 *Mixing*

In the block number 8, a stream mixer was used to connect the bypass stream with products from block 7.

2.5.9 *Heat integration*

All the heat streams related to endo- and exothermic reactions taking place in the process were connected together and summed up by a calculator block (not shown in Figure 2) representing the heat loss from the system to the surroundings.

2.6 Model description

SOLIDS and RK-SOAVE were chosen as base and property methods in Aspen Plus, based on the instructions of Aspen Plus User guide and VTT's in-house experiences about gasification modelling.

The following substances were considered as main components in the product gas: CO, H₂, CO₂, N₂, H₂O, CH₄, C₂H₂, C₂H₄, C₂H₆, NH₃ and O₂.

When gasifier temperature was fixed as an input, energy balance was used to calculate the heat loss and when heat loss was assumed, energy balance was used to predict the gasification temperature.

Inlet temperatures for steam, air and nitrogen were set to 200°C and for biomass to 30°C.

3 Results and discussion

3.1 Validation of the model

The data used for validation of the model is based on pressurised fluidised-bed air gasification of pine sawdust, pine bark, forest residues, wheat straw, and eucalyptus (see Table 4 for feedstock analyses). The reactor used in the studies was the PDU-scale gasification test rig presented in paragraph 2.2.

The results of these studies are published in detail by Kurkela et al. (1995). For validation purposes, six different operating variable sets, called set points, were chosen from five test campaigns conducted in 1993-94 as a part of the APAS Clean Coal Technology Programme.

First the values of biomass feed, air ratio, steam to fuel ratio, outlet temperature of the gasifier and process pressure were set in the model to correspond with the values of validation data as listed in Table 5. The model predictions for the product gas composition and carbon conversion were then compared with experimentally acquired values at the same set point conditions and are illustrated in Figure 5a,b,c,d.

The predicted and measured concentrations of the main components are summarised in Figures 5e,f with lines demonstrating +10% and -10% deviations between the values.

Judging from the results, a fairly good agreement between experimental data and model predictions has been achieved for the main gas components. The average relative error for components H₂, CO, CO₂, and CH₄ was 14%, while the magnitude of experimental error in the data is expected to be around 5%.

Figure 6 illustrates model estimations for carbon conversion along with the experimentally acquired results. It can be noticed, that although the model is able to produce relatively good product gas composition estimates for all set points, it manages to predict carbon conversion well for only 4 fuel types out of 6 used in the validation. Possible reasons for this outcome are discussed in the next paragraph.

3.2 The range of validity

A semi-empirical model can be considered valid only within the range of the data that was used to fit the model parameters. To assess this range of validity, the limiting values of the experimental data that was used in the parameter fitting, are listed in Table 6. A good prediction capability can be expected only within these values.

The gasifier type should also be considered when estimating the suitability of this model for process simulation purposes. It is emphasised that only gasifiers that share a similar type of geometry with the PDU-gasifier described in paragraph 2.2 should be simulated with this model.

The carbon conversion of a gasifier is known to be closely related with the gasification rate of the fuel, i.e. it's reactivity. The set point 4 was run with pine bark and set point 6 with wheat straw. The poor prediction capability of the model with these fuels seems to suggest that the reactivities of pine bark and wheat straw differ much from the reactivity of pine sawdust, used in the parameter fitting. For the same reason, the reactivities of pine sawdust, pine chips, forest residues and eucalyptus chips could be expected to be quite similar to each other.

There are some experimental results that seem to support this argument. For example Moilanen and Kurkela (1995) have studied carbon conversions for different types of fuels in a fluidised-bed gasifier and have found great differences in their gasification behaviour. Especially bark and straw were found difficult to be completely gasified.

Moilanen (2006) measured the instantaneous reaction rate of several fuels at 95% fuel conversion and with 1 bar steam and found the reaction rates to be 25%/min for pine sawdust and 30%/min for forest residue (pine), whereas the rates were 17%/min for wheat straw and 13%/min for pine bark. These numbers seem to imply that the differences in the accuracy of the model predictions can be explained, to some extent, by differences in reaction rates between the fuel types.

Thus, it can be concluded that the gasification model presented here, should not be used for fuels whose gasification reactivity differs greatly from the reactivity of pine sawdust.

3.3 Future work

The estimation of gasification behaviour requires detailed knowledge about the fuel structure and ash chemistry. Moilanen (1997) has speculated that the differences in the gasification behaviour of different fuels are due to the behaviour of the ash-forming substances in gasification. Therefore, in order to improve the prediction capability of the present model, information about the fuel characteristics should be incorporated in the carbon conversion predictor. This task will be one of the goals in the future work aiming to improve this model.

4 Conclusions

Experimental data from a PDU-scale reactor was used to fit and validate a semi-empirical model for the gasification of biomass. The model seems to be suitable for simulating gasification of pine sawdust, pine and eucalyptus wood chips as well as forest residues, but is not suitable for pine bark or wheat straw. The model is capable of predicting the concentrations of main product gas components with an average relative error of 14%. The greatest weakness of the model pertains to the prediction capability of carbon conversion when using fuels whose gasification reactivity differs greatly from the reactivity of pine sawdust.

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7 Figure captions

Figure 1. Schematic diagram of the pressurised fluidised-bed gasification test rig (Kurkela 1993).

Figure 2. A schematic illustration of the gasification model.

Figure 3a,b. Perceived correlations of freeboard temperature and carbon conversion as a function of air ratio (Kurkela 1993).

Figure 4a,b,c,d. Hydrocarbon and NH_3 conversions as a function of the air ratio (Kurkela 1993).

Figure 5a,b,c,d,e,f. Comparison of measured values with values predicted by the model for CO , H_2 , CO_2 and CH_4 concentrations in wet gas.

Figure 6a,b,c,d. Comparison of measured values with values predicted by the model for carbon conversion.

Table 1. Key parameters related to the experimental data used in model fitting.

Table 2. Proximate and ultimate analyses of the feedstock used in model fitting (Kurkela, 1993).

Table 3. Empirical correlations used in the model to simulate carbon, hydrocarbon and NH_3 conversions.

Table 4. Proximate and ultimate analyses of the feedstock used in the model validation (Kurkela et al. 1995).

Table 5. Process parameters related to the set points in the validation data.

Table 6. Assessment of the model's validity range.

8 Tables and figures

Set point	1	2	3	4	5	6	7	8	9	10
Feedstock	SD A	SD A	SD A	SD A	SD A	SD A	SD A	SD B	SD B	SD B
Fuel moisture, wt%	11,3	5,8	10,4	4,0	4,0	4,0	4,0	6,7	15,3	6,9
Pressure, bar	4	4	4	4	4	4	4	5	5	5
Air ratio	0,28	0,39	0,43	0,32	0,30	0,34	0,31	0,31	0,39	0,39
Gasifier outlet temp, °C	882	856	955	901	881	941	893	868	919	864
Steam to fuel ratio, kg/kg-daf*	0,08	0,28	0,23	0,17	0,16	0,17	0,16	0,15	0,20	0,18
Nitrogen to steam ratio, kg/kg	0,60	0,45	1,05	0,95	0,90	0,80	0,70	0,85	1,25	0,85

daf = dry, ash free

Table 1

	Sawdust A	Sawdust B
Proximate analysis, wt% d.b.:		
Volatile matter	83.0	82.8-83.1
Fixed carbon	16.7-16.8	16.8-17.0
Ash	0.20-0.34	0.08-0.23
Ultimate analysis, wt% d.b.:		
C	50.2-50.4	51.0-51.4
H	6.00-6.10	5.99-6.20
N	0.08-0.11	0.08-0.09
S	n.d.	< 0.01
O	43.2-43.5	42.1-42.8
Ash	0.20-0.34	0.08-0.23
Moisture content, wt%	4.0-11.3	6.4-15.5
LHV (dry), MJ/kg	n.d.	19.03-19.07

Table 2

Carbon conversion	25.7*E+88.5
NH ₃ conversion	0.819-1.154*E
Hydrocarbon conversions	
CH ₄	0.5166-0.8621*E
C ₂ H ₂	0.0046
C ₂ H ₄	0.138-0.311*E
C ₂ H ₆	0.02-0.038*E

E = air ratio

Table 3

	Pine sawdust	Pine chips	Forest residues	Pine bark	Eucal. Chips	Wheat straw
Proximate analysis, wt% d.b.:						
Fixed carbon	16,8	18	21,2	26,7	18,8	18,2
Volatile matter	83,1	81,5	76,7	71,8	80,4	75,8
Ash	0,08	0,43	2,1	1,6	0,8	6,1
Ultimate analysis, wt% d.b.:						
C	51	50,5	52,3	53,9	51,2	46,1
H	6	6,1	6	5,8	6	5,6
N	0,08	0,17	0,56	0,35	0,17	0,52
S	0	0,03	0,04	0,03	0,02	0,08
O	42,8	42,8	39	38,4	41,8	41,6
Ash	0,08	0,43	2,1	1,6	0,8	6,1
Moisture content, wt%	6.1-16	6.3-6.7	9.2-12	5.6-6.7	4,3	6,1

Table 4

Set point	1	2	3	4	5	6
Feedstock	Pine sawdust	Pine chips	Forest residues	Pine bark	Eucal. Chips	Wheat straw
Pressure, bar	5	5	5	5	5	5
Air to fuel ratio, kg/kg-daf*	2,33	1,65	1,97	2,27	1,90	1,37
Gasifier outlet temp, °C	930	905	890	980	935	835
Steam to fuel ratio, kg/kg-daf	0,20	0,09	0,14	0,12	0,14	0,31

Table 5

Type of feedstock	Sawdust
Feedstock moisture content, wt%	4.0 – 15.3
Gasification temperature, °C	856 – 955
Steam-to-fuel ratio, kg/kg	0.08-0.28
Air ratio	0.28 – 0.39

Table 6

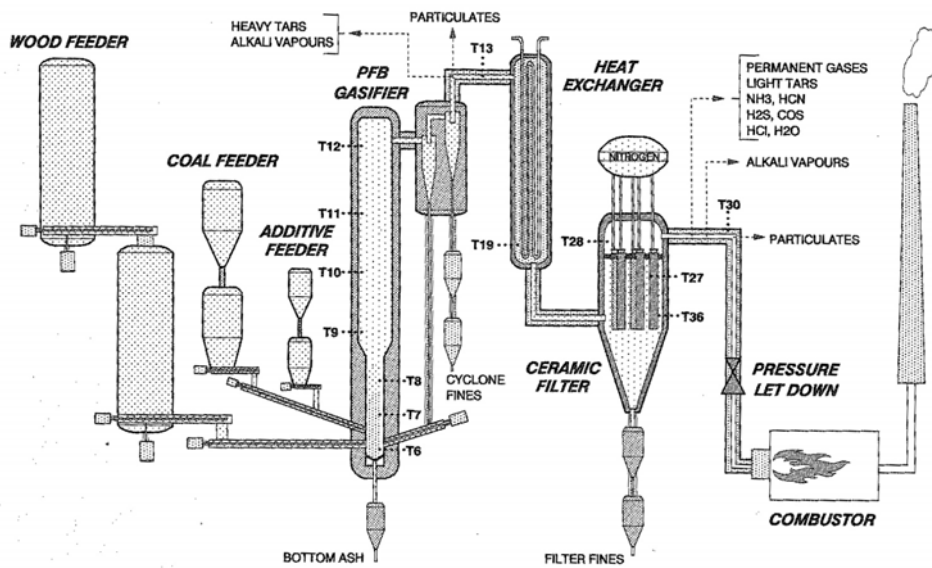


Figure 1

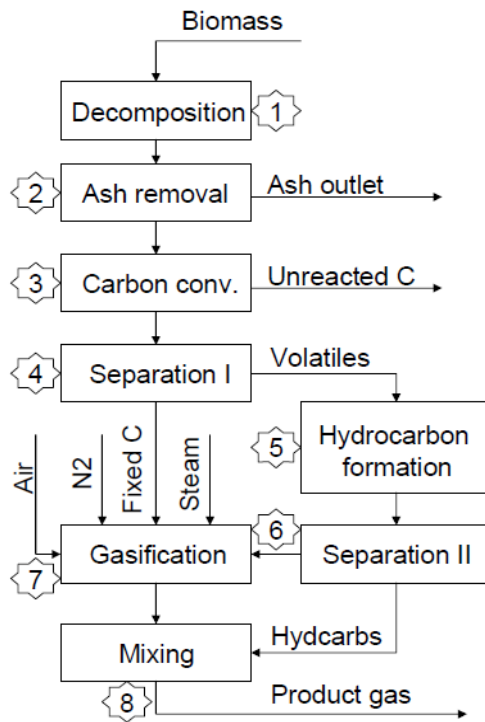


Figure 2

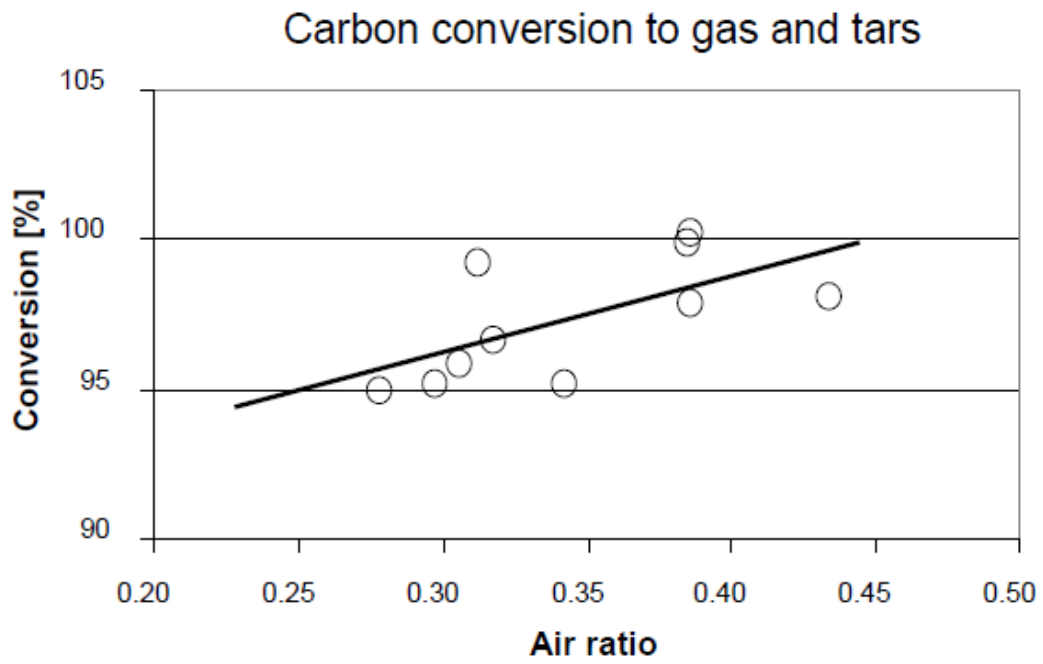


Figure 3a

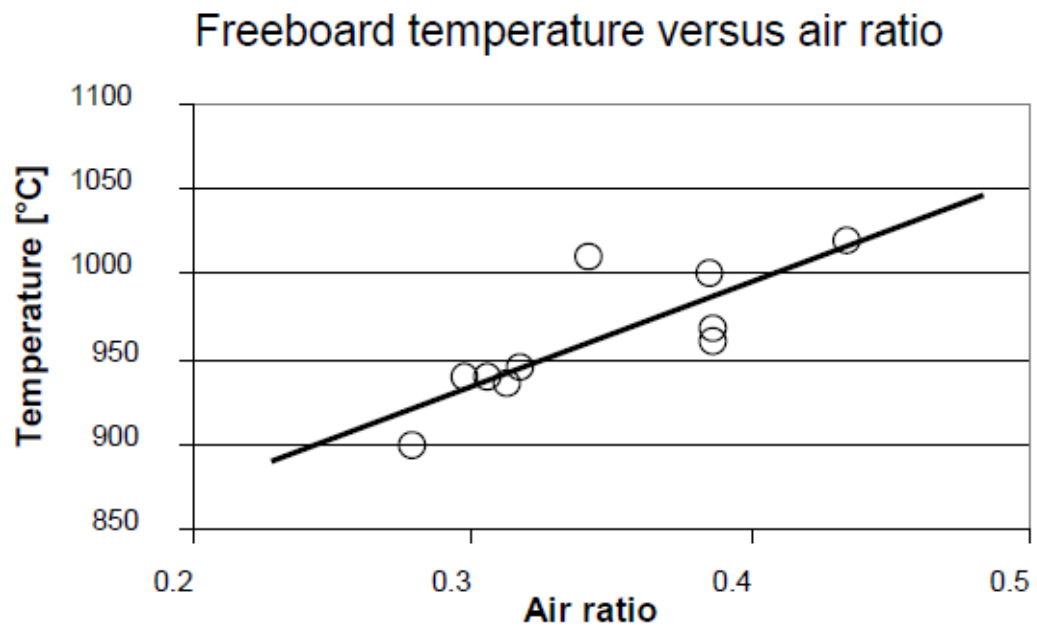


Figure 3b

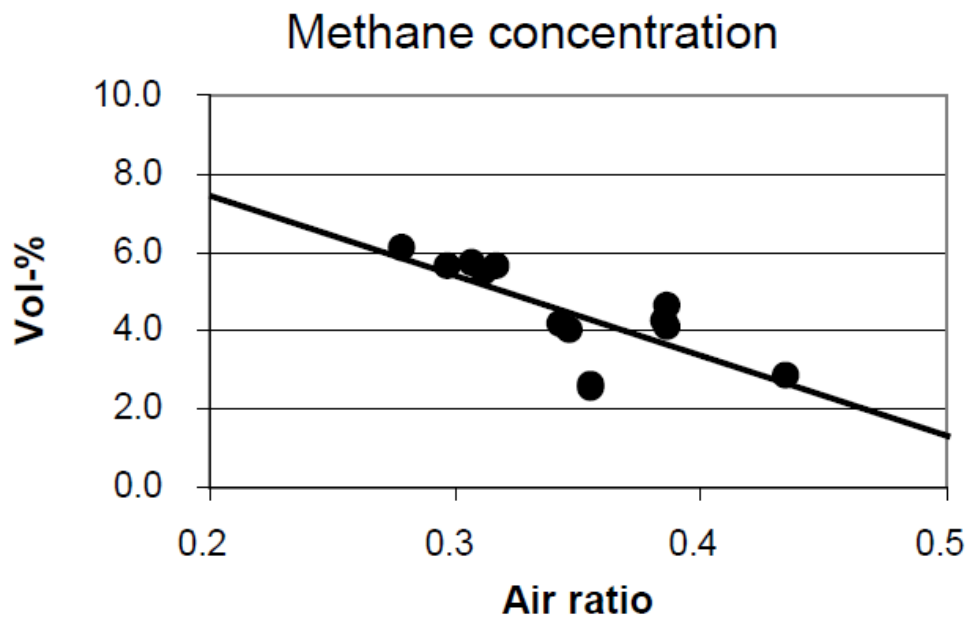


Figure 4a

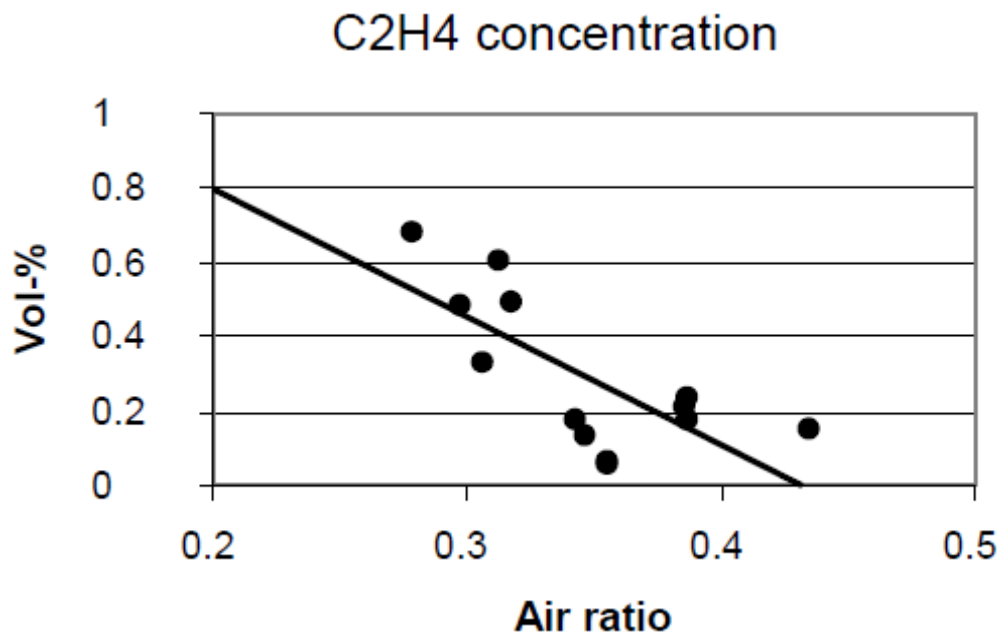


Figure 4b

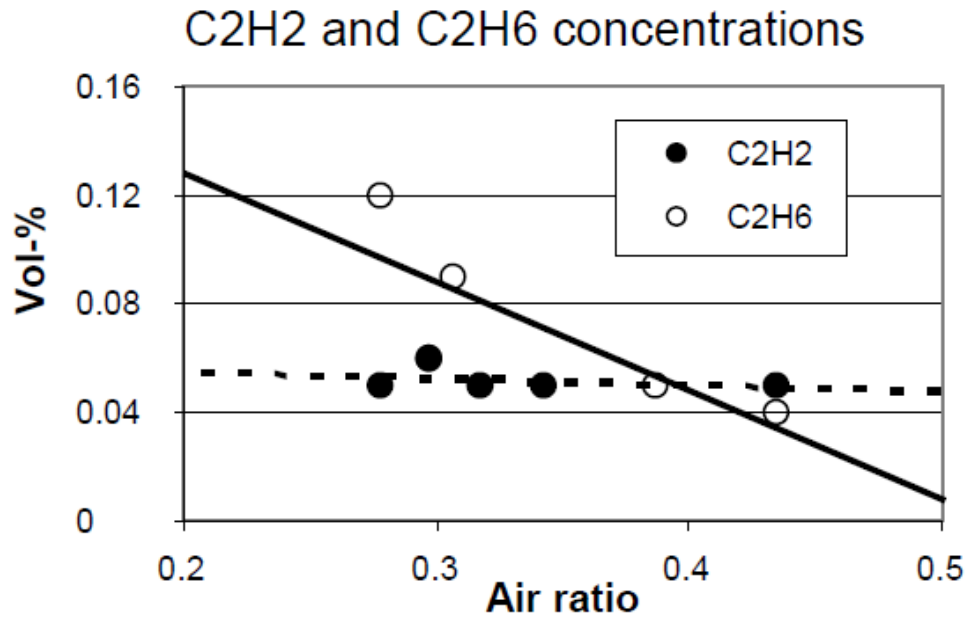


Figure 4c

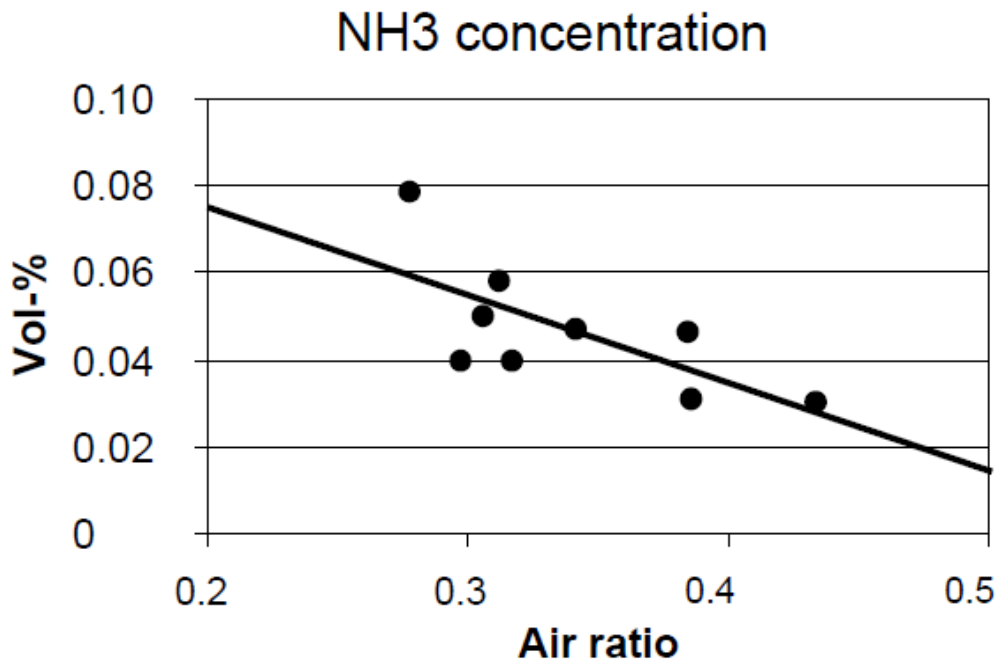


Figure 4d

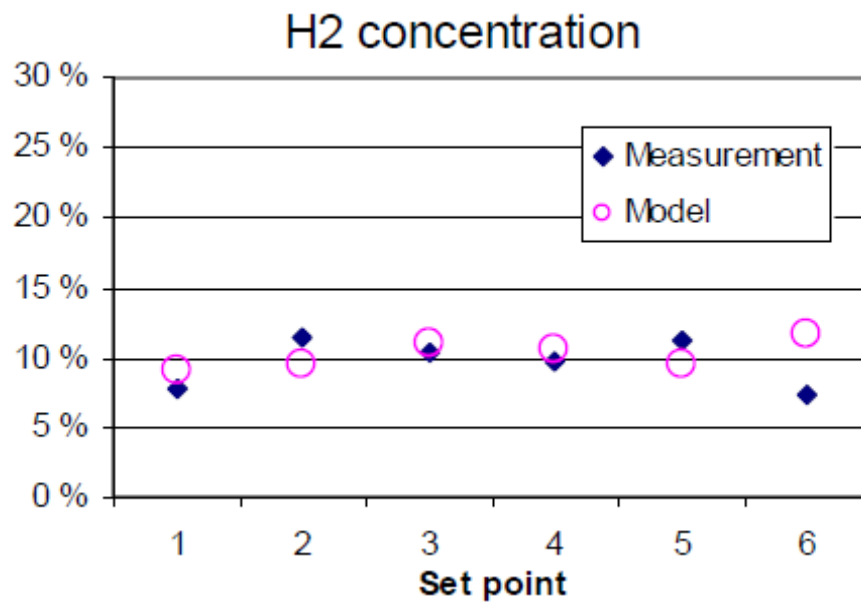


Figure 5a

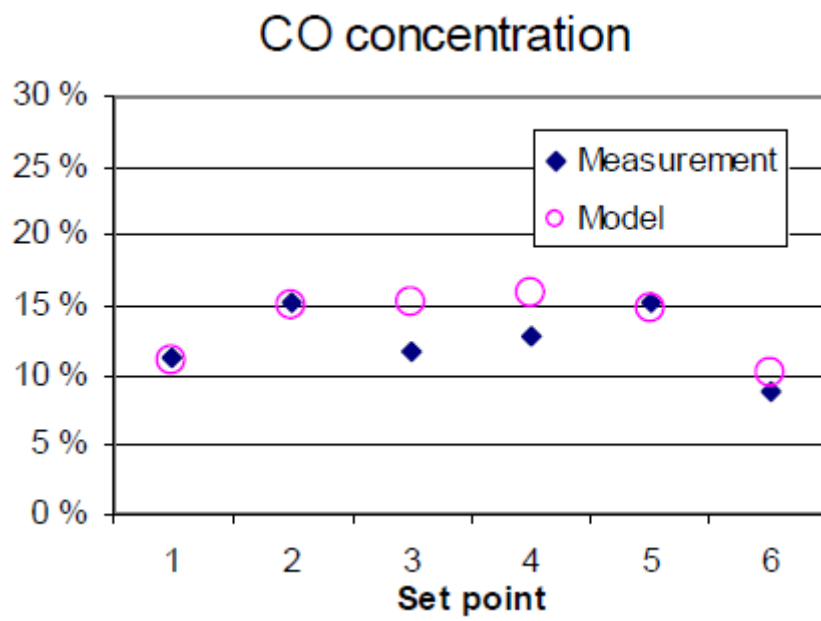


Figure 5b

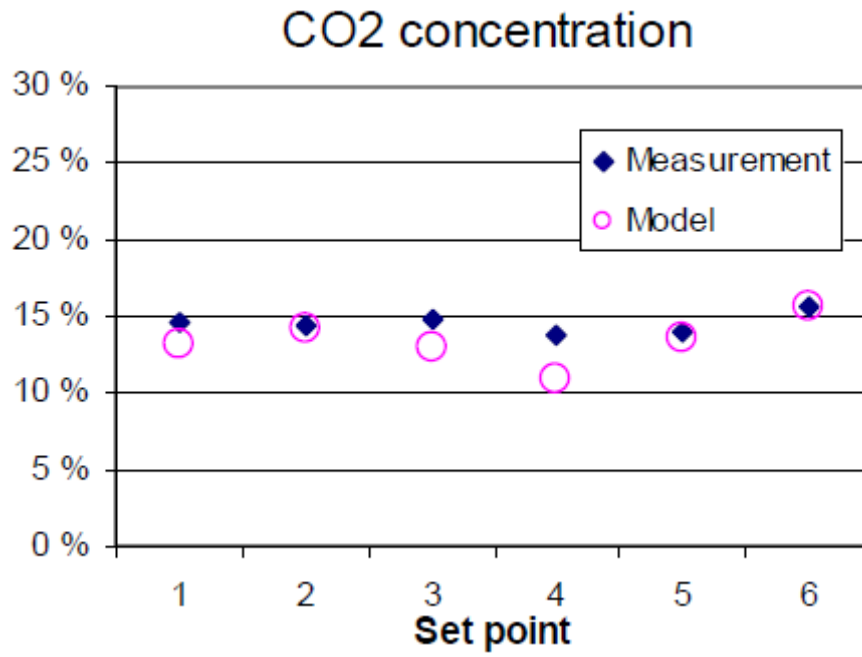


Figure 5c

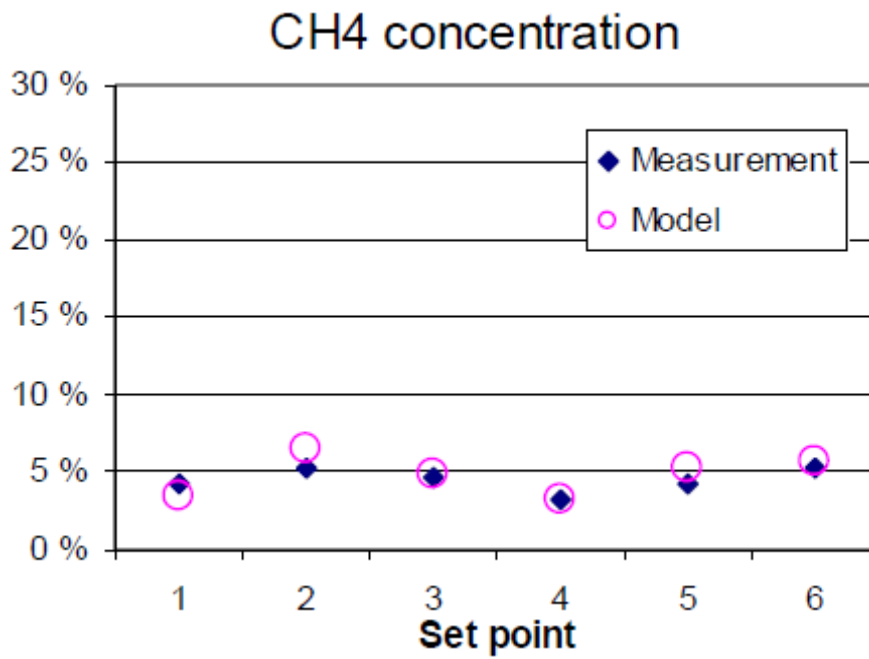


Figure 5d

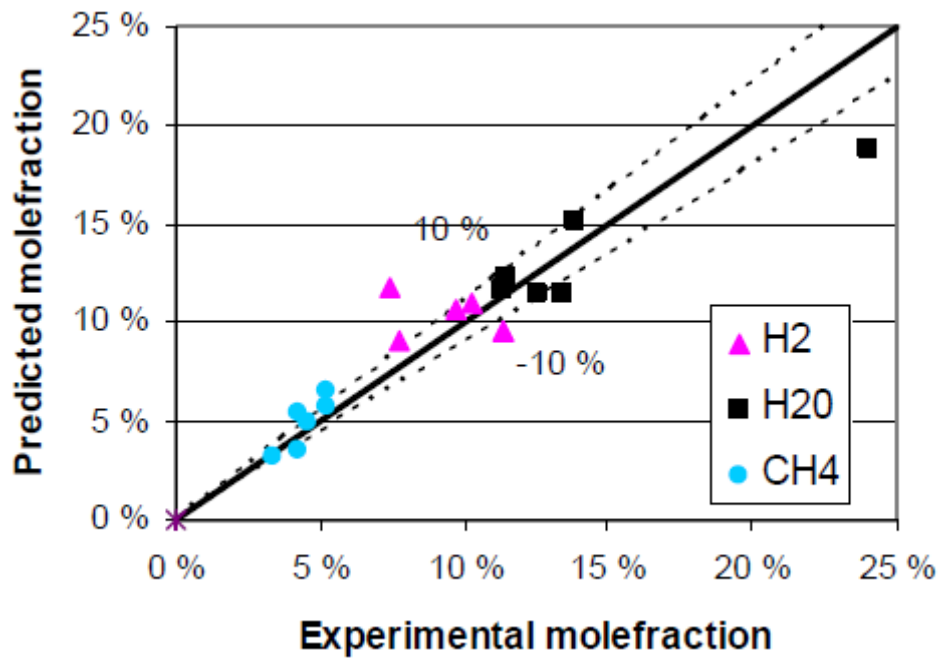


Figure 5e

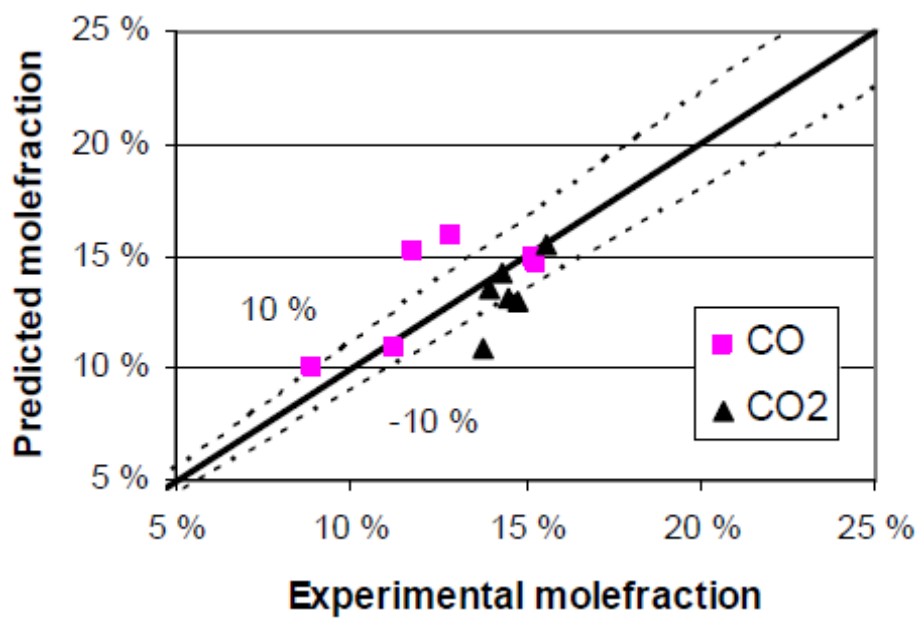


Figure 5f

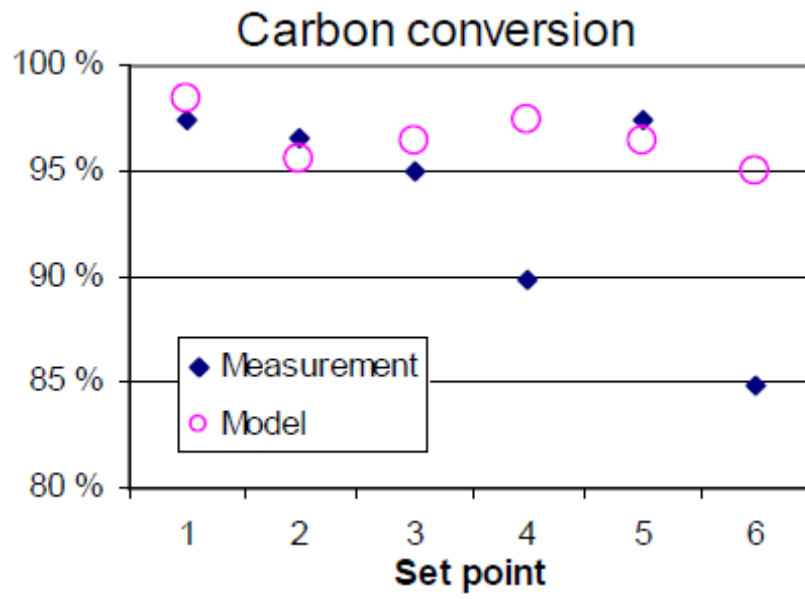


Figure 6.