Aspen modelling and life cycle assessment of the gasification of DKR 350 plastic waste

Based on modelling data which is validated through experimental research

Thesis report

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Abstract

The Netherlands is frontrunner in the European Union in recycling plastic waste, employing mechanical recycling for 45% of the waste stream while incinerating the remaining 55% with energy recovery. The share which currently not recycled are mostly mixed and contaminated plastics. Mechanical recycling requires a mostly pure and uncontaminated input and is therefore not well-suited for handling the remaining plastic waste flow. Gasification, on the other hand, is a chemical recycling technique which is better suited for handling streams of contaminated plastics. Despite its potential, modelling of plastic waste gasification is still in its infancy. Furthermore, no life cycle assessment (LCA) research had been conducted evaluating the influence of reactor parameters on the global warming potential (GWP) associated with this process. This thesis aims to bridge the gap by further developing and validating an Aspen Plus model and using it to address the following research question: *"What is the global warming potential of gasifying DKR 350 plastic waste compared to incineration in the Netherlands and what are the key reactor parameters that influence this impact?"* Based on the LCA results, the study reveals that gasification of DKR 350 plastic waste in the Netherlands has approximately half the GWP impact of incineration. Furthermore, increasing the reactor parameters steam to feedstock ratio and equivalence ratio exacerbates the GWP impact, indicating the importance of optimizing these parameters. The reactor parameters 'air on the riser' and temperature show minimal influence on GWP. However, the conclusions drawn from the LCA are subject to inaccuracies stemming from the Aspen model. Recommendations are proposed to enhance the accuracy of both the Aspen model and the LCA, enabling verification and increased significance of the results. Future research should focus on obtaining additional experimental data to improve the accuracy of the Aspen model, while keeping in mind that the purpose of modelling is to supplement and reduce the required experimental research.

Preface & acknowledgements

Nomenclature

Abbreviation	Description	Unit
ΔT	Temperature difference	°K
A	Area	m^2
AGR	Acid gas removal	
BFB	Bubbling fluidized bed	
C	Carbon	
CGS	Cold gas scrubber	
CHP	Combined heat and power	
Cl	Chlorine	
daf	Dry ash free	
DKR	Deutsche Gesellschaft für Kunststoff-Recycling	
dnf	Dry nitrogen free	
ER	Equivalence ratio	
GHG	Greenhouse gas	
GWP	Global warming potential	
H	Hydrogen	
HTC	Heat transfer coefficient	$Wm^{-2}K^{-1}$
IHBFBSR	Indirectly heated bubbling fluidized bed steam reforming	
k	HTC of the insulating material	
KPI	Key performance indicator	
k_{tot}	HTC of gasifier	$Wm^{-2}K^{-1}$
LCA	Life cycle assessment	
LCI	Life cycle inventory	
LCIA	Life cycle impact assessment	
N	Nitrogen	
O	Oxygen	
Q_{Lost}	Heat lost	W
S	Sulphur	
SFR	Steam to feedstock ratio	
α_o	HTC of air boundary layer	$Wm^{-2}K^{-1}$

1. Introduction

The annual worldwide production of fossil-based plastics amounted to 390.7 million tons in 2021 [1]. In total, it is estimated that primary plastics production between 1950 and 2021 amounted to 10.7 billion tonnes [2], [3]. In 2019, of the globally produced plastic waste, only 9.3% was recycled, 19.0% was incinerated and the remaining 71.7% was either landfilled or discarded into the natural environment [4]. In the past years, plastic pollution has gained significant international attention as a major environmental challenge [5]. Both conventional forms of managing plastic waste, namely landfilling and incineration, have a significant adverse effect on the environment and human health [6]–[8]. Moreover, the processes of landfilling and incineration result in plastic leaving the economy and necessitating the production of new plastics. The current fossil-based production and waste management of plastics is inherently unsustainable, and a shift towards a circular form of managing plastics is required through the utilisation of recycling [9].

However, the process of recycling can result in significant environmental impacts as well. Previous research, focussing on quantifying the impact of plastic waste management techniques through a life cycle assessment (LCA), has often been conducted by comparing these techniques to one another [10]–[16]. Some general conclusions can be drawn from comparative studies focussing on environmental impact; firstly, in terms of environmental impact all recycling options perform better than incineration. Secondly, incineration still creates less environmental impact than landfilling. And thirdly, when possible, mechanical recycling should be favoured over chemical recycling. However, in general, mechanical recycling has more stringent requirements for the feedstock composition and is therefore hard to implement for contaminated plastic waste streams [17]. Ideally, these recycling options can be used to complement each other by applying chemical recycling for plastic waste which cannot be processed through mechanical recycling [18].

In 2022, out of a group of countries, consisting of the 27 European Member States, Norway, Switzerland and the United Kingdom, the Netherlands was recycling the highest fraction of post-consumer plastics. In the Netherlands 45% of plastic waste was mechanically recycled, 55% was incinerated with energy recovery and no share was landfilled [19]. This 55% plastic waste which is currently incinerated is usually too contaminated and mixed to be mechanically recycled. Chemical recycling is not yet implemented at an industrial scale. Its development does however receive significant funding and attention from the scientific community and industry. Incineration of plastic waste is associated with major greenhouse gas (GHG) emissions [20]. Through recycling the emission of the carbon contained in the plastics can be prevented. Therefore, replacing incineration with chemical recycling can potentially decrease GHG emissions and help reach the Dutch governments emission target for 2050 [21].

There are four chemical recycling techniques available for processing plastic waste; these are pyrolysis, gasification, hydrocracking and depolymerization [22]. Pyrolysis is by far the most researched chemical recycling method and is also modelled most frequently in LCA research [18]. Pyrolysis is frequently cited as the superior chemical recycling method. However, Davidson et al. [18] concludes that there may be an unintentional bias towards pyrolysis caused by higher quality data availability for this technique. Therefore, they stress the importance of further research and LCA modelling of other chemical recycling methods to increase data availability and allow for better comparisons.

Gasification is considered the most promising chemical recycling technique when it comes to handling very contaminated and unsorted plastic waste streams [23]. Gasification is therefore chosen to be focussed on in this research. The definition for gasification that is used in this research is: "The conversion of carbonaceous solids or liquids mainly into a combustible gas at temperatures around 600–1500 °C under the presence of a gasifying agent and an oxygen feed below oxidation stoichiometric values" [24]. Gasification can convert plastics, using high temperatures, oxygen and

steam, into syngas composed of mainly carbon monoxide, hydrogen, methane and carbon dioxide. It is a mature technology currently in use for gasifying coal and biomass. The operating parameters of a gasification reactor significantly impact the syngas composition, gas yield and tar yield. These parameters include but are not limited to temperature, steam to feedstock ratio, and equivalence ratio [25]. They therefore also potentially influence the GHG emissions associated with this process.

The conclusions from previous comparative LCA research, which compares gasification of plastic waste to other waste management options are indecisive and very much depend on assumptions and decision made in the research. The only general conclusion which can be drawn is that gasification is a potentially promising technology, but more research is required. Jeswani et al. [26] stress that the location and energy mix used in the LCA strongly affect the results. The results are also found to be sensitive to which application for the syngas is included in the LCA, such as energy recovery [12], [27], chemical feedstock [28], hydrogen production [29]–[31] and methanol production [31]. Furthermore, a plethora of technologies and reactor types are suitable for the implementation of gasification, this also has a strong effect on results. To acquire inventory data on the gasification process often considerable simplifications are made, such as, assuming and idealised reactor as done by Meys et al. [28]. Also, a range of feedstocks are used ranging from virgin pure plastics to mixed waste. No research has been identified which directly evaluates the impact of changing reactor parameters on the environmental impact of the gasification of mixed plastic waste.

Modelling has the potential of significantly reducing the amount of experimental work required to optimise reactor parameters. Modelling of plastic waste gasification, however, is still in its infancy. For using a model to this end it is important to first perform validation of the model with accurate experimental data [32]. In literature two equilibrium models were identified on the gasification of mixed plastic waste, published in the works by Rosha and Ibrahim [33] and Saebea et al. [34]. The only kinetic Two-phase model on the gasification of mixed plastic waste found in literature was published by Roman [35]. This type of modelling can provide a more realistic representation of the process and more accurate results than equilibrium modelling [36]. The model developed by Roman simulates the gasification of polyolefins in an indirectly heated bubbling fluidized bed steam reforming (IHBFBSR) reactor. This work was made in the context of the Dutch government programme "A circular economy in the Netherlands by 2050" and the project "Towards improved circularity of polyolefin-based packaging". The input for this model is a plastic waste mixture which meets the DKR 350 specifications [37]. At the time that this model was developed no experimental data was available on the gasification of DKR 350 plastic waste. Therefore, it was not possible to validate the results with experimental data. Data from research by Martinez-Lera et al [38] and by Zaccariello & Mastellone [39] was used as a substitute. These sources describe the gasification of a PE+PP mixture and the co-gasification of PE+PP with wood and coal respectively.

In the present, experimental data on the gasification of DKR 350 in an IHBFBSR is available through the unpublished report of Gilvari and De Jong [40]. This research validates and further develops Romans model with the newly available experimental data and in doing so contributes to scientific knowledge on the modelling of plastic waste gasification. The second contribution of this research is to use the model combined with an LCA to assess the global warming potential (GWP) of the gasification of DKR 350 plastic waste when compared to incineration with energy recovery. Therefore, at the heart of this research is the pursuit to find an answer to the following question: *"What is the global warming potential of gasifying DKR 350 plastic waste compared to incineration in the Netherlands and what are the key reactor parameters that influence this impact?".*

The following chapters in this thesis are structured in the following way: Chapter two describes the technological and gasification process background. Chapter three delineates the methodology that is used during this research. The chapter thereafter is dedicated to the results produced during this research. In chapter five, the limitations and implications of this research are discussed. Finally, in

chapter six, a conclusion is drawn to provide an answer to the research question. At the end, the list of references and appendices are added.

2. Technology and process background

This chapter contains an additional description of the technology and processes relevant to this research. It starts by describing the gasifier type which is modelled in this research. Thereafter, multiple approaches to the modelling of gasification are discussed. Lastly, the origin and composition of the DKR 350 plastic waste mixture are explained.

2.1 Indirectly heated bubbling fluidized bed steam reformer

Tsekos et al. [25] proposed a novel gasification reactor design. The novelty of this reactor results from the method of heat provision for the gasification process; two radiant tubes that burn natural gas are inserted in the bottom and top of the reactor chamber. These tubes provide the heat indirectly and prevent the dilution of the product gas by reactants of combustion (e.g. carbon dioxide). Besides the gasification agent, which enters from the bottom of the reactor, a second air inlet is situated directly above the bed area into the area called the freeboard. This raises the temperature in the freeboard and promotes tar cracking. This is desirable, since tar formation is considered the main challenge of gasification due to it causing operational difficulties such as clogging, which decrease efficiency [41], [42]. A visualisation of the design of this reactor is presented in Figure 1. This reactor type is referred to as an indirectly heated bubbling fluidized bed steam reformer (IHBFBSR). This specific reactor type is used in this study due to the availability of both experimental research on the gasification of a DKR 350 mix in an IHBFBSR [40] and the availability of a model of this same reactor [35]. This experimental data is used to validate and further develop the model.
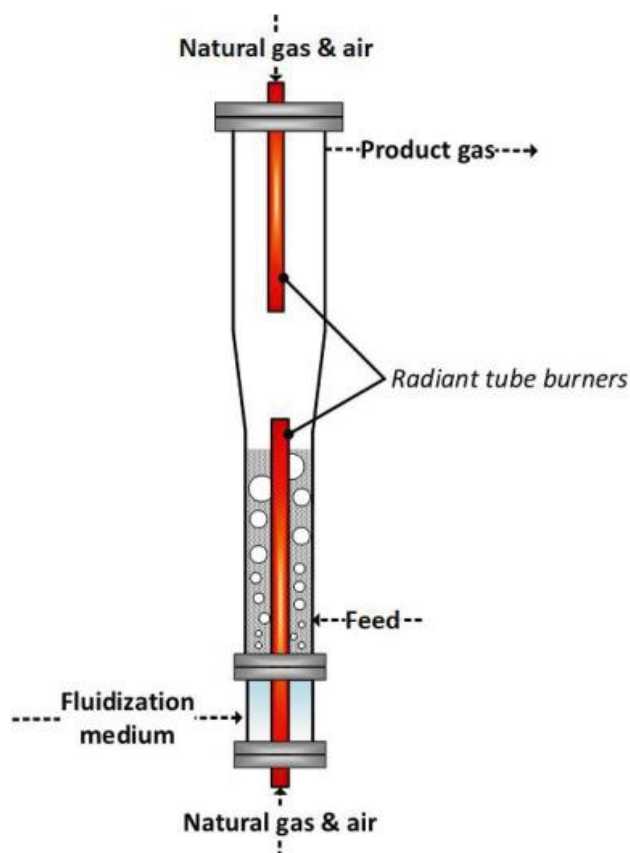

Figure 1: Conceptual design of the indirectly heated bubbling fluidized bed steam reformer (IHBFBSR) [25]

The performance of a gasifier can be evaluated with so called key performance indicators (KPI) [35]. The KPI's that were evaluated in the work by Gilvari and De Jong [40] are carbon conversion efficiency (CCE), CCE to gas, cold gas efficiency (CGE), overall efficiency (OE) and gas yield (GY). These parameters can be calculated with the equation 1-5 respectively which are presented below [40]. In these equations $\dot{m}$ denotes a mass flow, $\dot{v}$ denotes a volume flow.

$$CCE = 1 - \frac{\dot{m}_{C,solids}}{\dot{m}_{C,feedstock}}$$
(1)

$$CCE\ to\ gas = 1 - \frac{\dot{m}_{C,solids} + \dot{m}_{C,tar}}{\dot{m}_{C,feedstock}}$$
(2)

$$CGE = \frac{\dot{m}_{product\ gas} \cdot LHV_{product\ gas}}{\dot{m}_{feedstock} \cdot LHV_{feedstock}}$$
(3)

$$OE = \frac{\dot{m}_{product\ gas} \cdot LHV_{product\ gas}}{\dot{m}_{feedstock} \cdot LHV_{feedstock} + \dot{Q}_{Radiant\ tubes} + \dot{Q}_{Preheaters}}$$
(4)

$$GY = \frac{\dot{v}_{gas}}{\dot{m}_{feedstock}}$$
(5)

There are numerous reactor parameters that can be changed which potentially impact the KPI's and the environmental impact of the gasifier. Testing all parameters in all combinations is not within the scope of this research and therefore parameters are selected which are expected, based on experimental results [40], [43], to significantly influence the environmental impact of the waste to syngas process. The identified reactor parameters that are tested are: Steam to feedstock ratio (SFR), equivalence ratio (ER), the amount of air entered on the riser and the temperature in the gasifier.

2.2 Modelling of gasification

Aspen Plus (hereafter referred to as Aspen) has established itself as the most frequently used process simulation tool for both academic and industrial applications. Furthermore, a surge in the number of published papers on Aspen modelling of biomass gasification is observed in the past decade [44]. In Aspen multiple approaches can be implemented to model the gasification process. Approaches which can be followed include (but are not limited to); equilibrium modelling, quasi-equilibrium modelling, kinetic modelling and kinetic modelling with the inclusion of fluid hydrodynamics.

In equilibrium modelling it is assumed that the reactions within the model are able to reach equilibrium conditions. One of the reactor blocks which can be used in Aspen to implement this approach is the RGIBBS reactor block. This block calculates the output composition by minimising Gibbs free energy. Quasi-equilibrium modelling, also referred to restricted equilibrium modelling, takes a semi-empirical approach and makes a correction for reactions which in practise cannot reach equilibrium. This is implemented through specifying a degree of approach to equilibrium in Aspen for the system as a whole or per reaction. This degree of approach is an offset between the temperature which is used in the model calculations and the actual temperature of the reactor. This approach allows for fitting the model results to empirical data by iteratively changing the degree of approach [45]. Inaccuracies incurred by the assumption that reactions occur to equilibrium can't always be sufficiently compensated for through a degree of approach.

In kinetic modelling a more rigorous approach is followed by taking reaction rate and residence time into consideration. In Aspen the reaction rate is approached through the Arrhenius equation. This equation requires the pre-exponential factor and activation energy to be specified for each reaction

which is entered into the model. These kinetic parameters are determined through empirical research and are influenced by the conditions under which the reaction takes place. Therefore, finding the kinetic parameters in scientific literature or determining them in experimental research for all reactions which can occur in the gasifier is a tedious task. It is common practice to change the pre-exponential factor to some extent to fit the model results to empirical results. A robust kinetic model requires the hydrodynamics of the material in the bed zone and freeboard to be modelled. This is a complex task which requires solving equations for the momentum, energy, and mass of the gas and solid particles [35].

An easier approach to modelling the hydrodynamics is based in two-phase theory [46]. Gómez-Barea and Leckner [47] conclude that two-phase models are the most widely implemented approach to modelling biomass fed fluidised bed gasifiers. In this approach the bed zone is divided in two phases: an emulsion phase and a bubble phase. Typical assumptions are the emulsion phase is at minimal fluidization conditions and the bubble phase is free of solids [47]. This can be implemented in Aspen by splitting the flow to a sequence of RCSTR and RPFR pairs which represent the emulsion and bubble phases, respectively. After each pair the flow is recombined and separated again to the next pair. The freeboard can be represented by a single RPFR block.

2.3 DKR 350 plastic waste

In the Netherlands, the collected plastic waste is sorted into multiple different waste streams. A simplified flowsheet of this sorting process is shown in Figure 2. The higher purity flows, which results from the sorting process, are better suited for mechanical recycling. DKR 350 is the name for the remaining mixed flow, and is uneconomical or unfeasible to separate any further. This flow is a complex mixture of plastics that is representative for a post-consumer waste stream. It comprises mostly of packaging material but also contains biogenic and inorganic material and halogens [48]. This stream makes up 35% of the total plastic waste stream and is difficult to separate further or recycle mechanically and has the highest economical potential for chemical recycling [49]. The specifications for the allowed contents of DKR 350 are defined by the waste management agency Deutsche Gesellschaft für Kunststoff-Recycling (DKR) [37]. These specifications can be found in Appendix Figure 10.
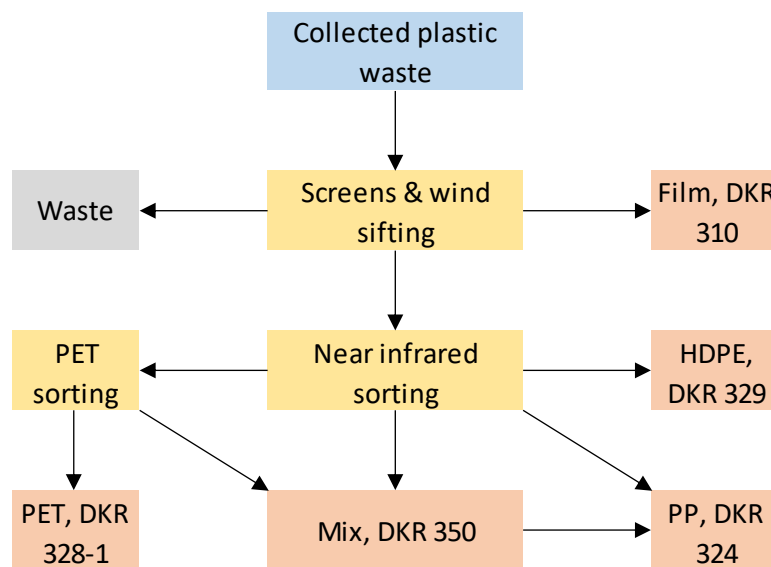

Figure 2: Simplified sorting flowsheet for post-consumer packaging plastics. Adapted from [50]

3. Method

This chapter describes the methods that were applied during this research. The first section contains a visualisation of the research flow. The second section describes the approaches which are used to improve and expand the Aspen model. The third section describes all changes which were made to the model. The fourth section discusses the validation case and model settings used for validation of the model. The last section describes the methods for the LCA.

3.1 Research flow

The research steps that are taken are visualised and presented in Figure 3. The first phase is concerned with validating and developing the model. The second phase is the LCA which in itself is made up out of 4 iterative phases. Both the model validation and development, and the LCA yield results.
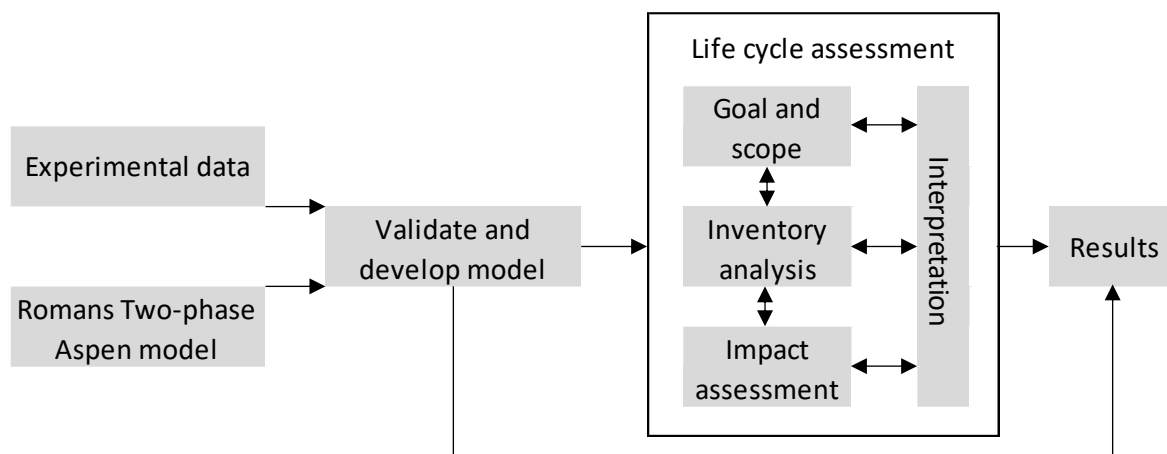

Figure 3: Schematic visualisation of the research flow. Partly adapted from [51]

3.2 Approaches to improving the model

Three approaches are employed to improve the model. The first approach is to validate the model with the experimental data from the work by Gilvari and De Jong [40]. By analysing the differences between the model and the experimental data potential avenues for improvement are identified. Validation is applied in an iterative way to continuously assess the impact of changes made to the model. The report of Gilvari and De Jong [40] describes four experimental cases. During case 1 the gasifier suffered from a blockage problem in the sampling line which caused no tars to be collected. Case 2 and 4 also suffered from this issue, causing the amount of collected tars to be below the minimum standard for the sampling method. Case 3 (hereafter referred to as TUD case 3), did not suffer from this issue and is therefore selected to be used for validation of the model in this research. The parameters retrieved from TUD case 3 and used during validation are reported in section 3.4.

The second approach used is to rigorously review the original Aspen model and the associated Fortran code for potential bugs and avenues of improvement. For a change to be considered an improvement, the change must either be both scientifically justifiable and improve the accuracy of the model or be considered a bug fix.

The third approach is to expand the model which makes the results suitable in the LCA and allow a fair comparison to the reference case of incineration. This includes adding heat recovery in the model, adding a cleaning stage for the product gas and fuelling the radiant burner with a fraction of the syngas product instead of using natural gas.

3.3 Changes made to the model

This section explains all changes made to the model based on the approaches described in the previous section. Figure 4 depicts the simplified overview of the Aspen model after all changes are

implemented. The blue boxes represent components which were already present in the original model, the grey boxes are the components that are added. Black arrows represent material flows and red arrows represent energy flows. The heat is directly transferred through heat exchangers and radiant tubes and thus has no physical energy carrier. The complex Aspen flowsheet is added to the Appendix. The original Aspen flowsheet can be found in appendix Figure 11 and the updated flowsheet is found in appendix Figure 12. All heat flows are removed from these Aspen flowsheets to increase readability.
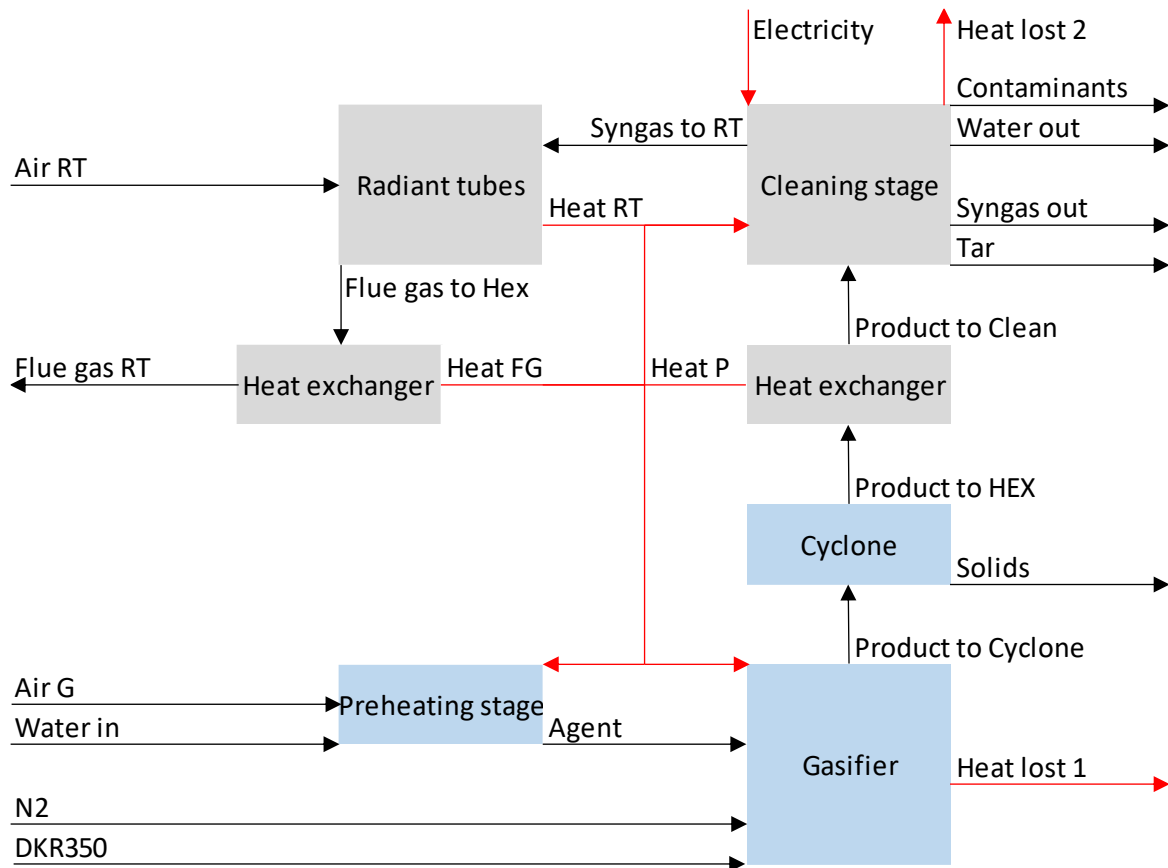
Figure 4: Simplified Aspen model flowsheet

The changes made to the model are categorised in to two types. Modifications to the original model and additions to the model. The first type is discussed below, the second type is described thereafter.

3.3.1 Model modifications

A summary of all modification made to the original model and the motivation for implementing these changes can be found in Table 1. A more detailed and technical explanation of these changes can be found in Appendix Table 9.

Table 1: Summary of modifications made to original model

Change number	Motivation for change	Implemented change
1	The volume which is calculated for the CSTR blocks of the CSTR-PFR pairs was incorrectly placed in the sequence of calculation steps, resulting in the model using a default volume value instead.	The model now correctly uses the calculated volumes instead of the default values.
2	For the CSTR-PFR pairs 2-4 the input flow was split 50/50, 50% to the CSTR and 50% to PFR block.	The split is now made dependent on the relative size

	However, this split should depend on the relative volume of these blocks as they are not necessarily the same size. Furthermore, the presence of the volume of the catalyst was not taken into account.	of the catalyst free volume of the CSTR and PFR blocks.
3	The automatic setting of Aspen to determine when calculator blocks should be run was used. However, a bug in Aspen prevents the calculation sequence of being correctly determined when running a sensitivity loop, resulting in some calculator blocks not being rerun for every iteration.	Manually indicated when each calculator block should be run in the sequence.
4	The rate equation of one of the partial oxidation reactions of naphthalene was modelled incompletely in Aspen. The temperature dependency was forgotten.	Temperature dependency is now activated.
5	The carbon monoxide combustion reaction was inactive in the freeboard reactor block of the gasifier. However, this reaction should take place here [35].	Reaction is now activated in the freeboard area.
6	Mass convergence errors occur in some of the CSTR blocks of the bed zone in some of the model runs.	This is a known bug in Aspen, however the solutions recommend in the help page did not suffice. So instead convergence was simplified by removing a tear variable, combined with a workaround of setting the model to stop at the iteration with the smallest root mean square error in the mass balance.
7	To fit the expected amounts of H2 and CO in the output gas the rate constants of combustion are altered.	To fit the model results to the experimental data the rate constants of the combustion reaction of H2 and CO are edited from respectively 2.2×10^9 and 2.32×10^{12} to 2.2×10^{11} and 2.32×10^{11}.
8	The feedstock is fed to the gasifier by a screw feeder which is pressurised with pure nitrogen to prevent reactions from occurring in the feeder. The nitrogen flowrate through the screw feeder was set to 50% of the weight of the feedstock flow rate. This significantly dilutes the product gas flow. However, no source was reported for this nitrogen flow rate nor is this value reported by either of the works by Gilvari and De Jong [40] or TNO report.	The nitrogen flow rate through the screw feeder is now set to 20% by weight. This value is based upon the flow rates reported by De Jong [52]. This research describes the gasification of biomass and coal in a BFB gasifier which is also fed with a screw feeder.

3.3.2 Model development

Further development of the model is required to provide results which can be used to assess the environmental impact through an LCA. The needed additions are the following: 1) In practise the product flow exiting the gasifier will need to be cleaned to be usable in further applications. 2) The original version of the model assumes perfect insulation and thus no heat loss from the gasification setup. This is unrealistic and a calculation for the lost heat needs to be implemented. 3) The radiant burners in the experimental research by TUD burn natural gas. The process can be made self-sufficient

by burning part of the syngas product instead. 4) Heat available in the product flow exiting the gasifier and the flue gas leaving the radiant burners can be partly recovered and reused in the gasification process. Wasting this heat would be uneconomical when implementing gasification on industrial scale. The text hereafter describes the methods that are used to implement these changes in the model.

3.3.2.1 Syngas cleaning

The main purpose of cleaning the product flow is to create syngas which is usable in further applications. The tolerances to the composition and allowed impurities vary depending on the application of the syngas [53]. Also, a range of technologies exit that can be used to implement the cleaning. Optimizing the cleaning process for a certain application is a tedious task that is outside of the scope of this research. The same simplified approach to cleaning as was used by Piroddi [54] is adopted in this research. This approach is aimed at being able to assess the global warming potential of the cleaning process with decent accuracy. If other environmental impact categories are going to be assessed then the cleaning process will need to be modelled more thoroughly.

In this approach a cold gas scrubber (CGS) is employed to achieve the separation of the tars from the syngas. For simplicity this CGS is assumed to be 100% effective at removing tars and the flow of water and tars exit separately from the CGS. The electricity which is required to operate the CGS is assumed to be 9.8 kJ/kg of scrubbed syngas [55]. In practise the separated tars can be reinjected in to the gasifier to undergo further reactions. However this would severely complicate the convergence of the model results in Aspen and is therefore not implemented. An acid gas removal (AGR) unit is utilised to separate out the contaminants: NH_3, H_2S and HCl. Again for simplicity, this process is assumed to be 100% effective at removing these contaminants. Electricity and heat is required to operate the AGR, the amount of required electricity and heat are assumed to be 25.2 MJ/kg of separated contaminants and 63 MJ/kg of separated contaminants respectively [56].

3.3.2.2 Heat loss

The magnitude of heat losses from the gasification setup is not reported in either of the works by Gilvari and De Jong [40] or Grootjes [43]. Therefore, a calculation is required to estimate these losses. Modelling all convective and radiative losses from all components including the losses induced by the pre-heating stage, water cooled jacket of the screw feeder, cyclone and pipes is not within the scope of this research. An order of magnitude estimation is made by calculating the heat transfer losses from the surface of the gasifier. The retried value for the heat loss is multiplied by a factor of 3 to provide a rough estimate for all heat losses from the other components.

The heat transfer coefficient (HTC) of the gasifier (k_{tot}) is estimated with equation 6. Here k is the HTC of the insulating material in the gasifier wall. The insulation is made with a 20 cm thick matrass [40]. This matrass is assumed to be made of PU foam with an HTC of 0.125 $Wm^{-2}K^{-1}$ [57]. And α_o is the HTC of the boundary layer of air on the outside of the gasifier which is assumed to be 7.7 $Wm^{-2}K^{-1}$ [58].

$$k_{tot} = \frac{1}{\frac{1}{k} + \frac{1}{\alpha_o}}$$

(6)

This equation assumes that the radiative heat transfer is negligible. This assumption is justified because the radiative heat losses are usually a small fraction of the total heat loss [59]. Furthermore, the surface of the gasifier is assumed to be covered in a material with low emissivity. This equation also assumes the impact of the HTC of the gas boundary layer on the inside of the gasifier to be insignificant on the total HTC. This can be assumed because the gas flow on the inside is characterised as a strongly forced and turbulent flow. Furthermore, this calculation neglects the insulating effect of the gasifier wall. This is assumed because it is made of 4.78 mm thin steel [40] which is a material with

a high HTC relative to the matrass. Also, this equation assumes a one-dimensional character for heat transfer through the gasifier wall. This assumption is made to make a simple estimation possible.

The total flow of heat lost through the gasifier wall (Q_{Lost}) is calculated with equation 7. Here ΔT is the temperature difference between the inside of the gasifier and the ambient temperature. 25°C is used for the ambient temperature, and A is the area of the gasifier. This equation is applied separately for the bed zone and freeboard areas as they have a different internal temperature.

$$Q_{Lost} = \Delta T \times k_{tot} \times A \qquad\qquad (7)$$

For this calculation the area of the gasifier is calculated by taking the log mean square of the area on the inside of the insultation and on the outside of the insulation. Heat losses through the corner areas of the top and bottom cylinders cannot be calculated with this equation because the assumption that heat transfer has a one-dimensional character is not valid in this region. The heat losses through this area are therefore neglected. The log mean square area of the bed zone and freeboard are calculated to be 1.84 m^2 and 2.86 m^2 respectively. The calculations for the lost heat are implemented in the model through a Fortran subroutine.

3.3.2.3 Syngas combustion in radiant burners

In the original model the radiant burners were not modelled. Only the required flow of heat from the radiant burners was calculated. The radiant burners are now added and a fraction of the cleaned syngas is combusted in these burners to provide heat for maintaining the temperature in the gasifier. Mewani [60] investigated optimal operating conditions for the type of radiant burners which were used in the experimental setup described in the work by Gilvari and De Jong [40]. That work found that the thermal efficiency is highest at a stoichiometric ratio of 1.2. At this stoichiometric ratio 77% of the HHV energy contained in the syngas is radiated through the burners [60]. The remaining heat of combustion is contained in the flue gas which exits the burners.

3.3.2.4 Heat recovery

Two sources of heat are available, in the gasification setup, which can be recovered through counter current heat exchangers. These are the product flow which exits the cyclone and the flue gas which exits the radiant burners. Furthermore, heat is provided by the radiant tubes to the gasification reactor. The product flow can be cooled to a minimum of 350 °C. This temperature is chosen as it is above the tar dew point [61] which prevent clogging of the system through condensed tars. Heat recovery from the flue gas exiting the radiant burners is assumed to be implemented with a condensing heat exchanger. Gang et al [62] determined that heat recovery from flue gas is economical to a minimum temperature of 98 °C. The primary heat sink is the preheater for the gasifying agents, air and steam, which heats the agent from 25 °C to 650 °C. Also, the heat required by the AGR is assumed to be provided through heat recovery. Here heat is required to recover the solvent used by the AGR at a temperature of 70°C [56].

Designing the most efficient heat exchanger network would yield a different result for each scenario; this goes beyond the scope of this research. Since the minimum temperatures of the heat sources are significantly higher than the desired temperature of the heat sinks, it is assumed that heat recovery will be possible as long as the following two constraints are satisfied; 1) the total heat available from the heat exchangers and radiant tubes must be enough to satisfy the heat requirement of the gasifier, preheaters and AGR and 2) the total heat delivered by the radiant tubes must be enough to satisfy the heat required by the gasifier. In Aspen a design spec block is utilised to find which fraction of syngas product needs to be burned in the radiant burners to satisfy both constraints.

3.4 Validation case

Validating and improving the model is an interleaved and iterative process and is used to continuously evaluate the impact of changes made to the model. The model settings which are used to validate the model with TUD case 3 are shown in Table 2. Not all required values are reported in the work by Gilvari and De Jong or the values reported need to be adapted to be used in the Aspen model. Decisions made in these instances are listed below.

The proximate analysis values for fixed carbon and volatile matter are not reported by Gilvari and De Jong [40]. Therefore, these values are taken from the proximate analysis of the DKR 350 mixture used in the work by Roman [35]. The proximate analysis values are then corrected to make the values add up to 1. The experimental work by Gilvari and De Jong [40] reports the ultimate analysis on an ash free basis. Aspen, however, requires these values to be entered including the ashes. The ultimate analysis data is therefore transposed and rounding is edited to make the values add up to 1. The freeboard area is modelled to be constant at the specified temperature instead of being modelled adiabatically. This is done to make sure that the freeboard temperature is maintained at the same level as was reported in the experimental work by Gilvari and De Jong [40].

Table 2: Model settings for validation taken from TUD case 3 [40]

Category	Setting	Value
Flow rates	Feed	10 kg/h
	N2/feedstock ratio	0.2
	Equivalence ratio	0.12
	Steam/feedstock ratio	2.04
	Secondary air flow	4 kg/h
Proximate analysis feedstock	Moisture	0.66%
	Fixed carbon	8.96%
	Volatile matter	84.43%
	Ash	5.95%
	HHV	41.87 MJ/kg
Ultimate analysis feedstock	C	71.6%
	H	11.2%
	N	0.535%
	Cl	0.273%
	S	0.122%
	O	10.4%
Catalyst	Loading	75 kg
	Density	3940 kg/m3
	Particle size	490 μm
Temperature	Bed area	757 °C
	Freeboard area	861 °C

3.5 Life cycle assessment

The research methodology for conducting an LCA has been standardised by the International Organization for Standardization in the standards ISO 14040 and ISO 14044 [51], [63]. These standards are intended to make LCA research comparable and reliable and will therefore be followed in this research. An LCA consists of four phases: 1) Goal and scope definition, 2) Life cycle inventory (LCI) analysis, 3) Life cycle impact assessment (LCIA) and 4) Interpretation. These phases are explained in

more detail below. An LCA is an iterative process and the choices made in each phase are refined during the research.

3.5.1 Goal and scope

The goal of this LCA is to gain further understanding of the environmental impact of the gasification of DKR 350 when compared to the current method for handling DKR 350 in the Netherlands, namely incineration [17]. The LCA is focussed on identifying key operating parameters which can help reduce the environmental impact of gasification of this waste stream.

The gasification of DKR 350 to syngas can be seen as a system with a dual function; processing waste and producing syngas. Reducing the environmental impact of plastic waste is the main goal of this LCA and therefore the main function of the system. Syngas is treated as a by-product, for which credits are applied. The functional unit in the LCA is defined as: the waste treatment of one tonne of DKR 350 produced in the Netherlands.

The system boundaries of plastic waste treatment are defined according to a cradle-to-gate approach. In this consequential LCA the reference system and the system understudy only differ in the treatment of the DKR 350 which exits from the sorting unit. Therefore, the cradle stage can be taken to be the DKR 350 mix leaving the sorting unit and a cut-off approach to the plastics initial life is justified. Furthermore, the economic value of DKR 350 is negative [64] and thus the impacts incurred through the production, usage during the initial life and sorting process of the plastics can be completely allocated to its initial life.

The syngas and tar produced by the gasifier are complex flows with a plethora of use cases which depend on the composition of these flows. Identifying the application or set of applications with the lowest environmental impact based on the composition of these flows is a study in itself and therefore outside the scope of this research. To still be able to attribute value to these flows, it is assumed that the syngas is combusted in a CHP plant and the tar is combusted in the same incinerator where the DKR 350 mix is incinerated in the reference case. This way energy can be recovered in the form of electricity and heat. The efficiency of the CHP plant and incinerator are taken from respective processes in the ecoinvent database [65] and can be found in Table 3.

Table 3: Heat recovery efficiency

Ecoinvent process	Simulates	Efficiency to		Total
		Electricity	Heat	
Treatment of waste plastic, mixture, municipal incineration with fly ash extract (CH)	Incinerator	11.5%	22.5%	34.0%
Heat and power co-generation, biogas, gas engine NL	CHP	32%	55%	87.0%

It should be kept in mind that combustion of the gasification products is not the desired pathway when the aim is to achieve a circular carbon economy. Even so, combustion of the syngas is potentially beneficial when compared to direct incineration of the plastic waste. This is because energy can be recovered more efficiently from a gas than from a solid. The system boundaries of the system under study are displayed in Figure 5. The reference system is displayed in Figure 6. In both figures the black arrows, again, represent material flows and the red arrows represent energy flows.
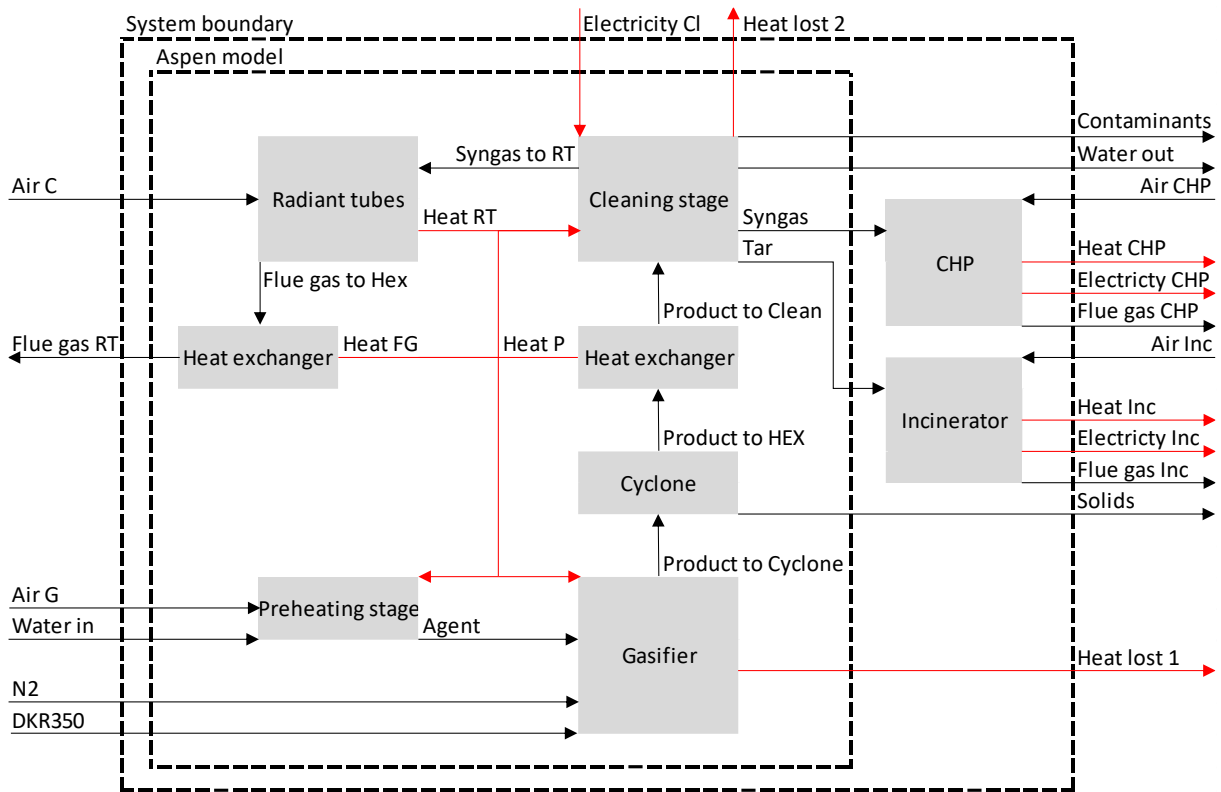
Figure 5: System boundaries of system under study.
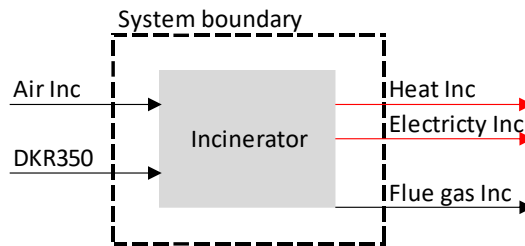
Figure 6: System boundaries of reference system

3.5.2 Life cycle inventory analysis

The primary inventory data for the LCA is provided by the Aspen model of the gasifier, this is supplemented with ecoinvent data for the CHP and incinerator. Five scenarios are simulated and evaluated in this research. A base scenario, for which the reactor parameters are taken from TUD case 3, is complemented by four scenarios in which the four previously identified reactor parameter, that potentially influence the environmental impact of the process, are varied. The scenario matrix showing the reactor parameters for these five scenarios is presented in Table 4.

Table 4: Scenario matrix

Scenario	Steam to feedstock ratio	Equivalence ratio	Air entering in freeboard	Temperature (bed zone)
1	2.04	0.12	4 kg/h	757 °C
2	3	0.12	4 kg/h	757 °C
3	2.04	0.2	4 kg/h	757 °C
4	2.04	0.12	6 kg/h	757 °C
5	2.04	0.12	4 kg/h	850 °C

The calculated magnitude of all mass and energy flows are presented in Table 5. The quantity of these flows is expressed relative to the functional unit i.e. 1 ton of DKR-350 plastic waste. The inventory data for the reference scenario to which all five scenarios are compared is also contained in this table. In the reference scenario it is assumed that all carbon present in the feedstock is emitted to the atmosphere in the form of CO_2. The magnitude of the flows of air to the CHP and incinerator are not calculated since air is freely available and has no impact on GWP. Also, the magnitude of these flows is not required for calculating the other flows. The same reasoning is also applicable for the non-CO_2 part of the flue gas from the CHP and incinerator.

Table 5: Inventory table of all mass and energy flows in system under study. The red font indicates energy flows.

Inputs (unit per tonne DKR 350)	Unit	S1	S2	S3	S4	S5	Reference
Air G	tonne	1.49	1.49	2.49	1.49	1.49	
Air RT	tonne	0.50	1.70	0.00	0.52	0.54	
Air CHP (not calculated)							
Air Inc (not calculated)							
Water in	tonne	2.03	2.98	2.03	2.03	2.03	
N2	tonne	0.20	0.20	0.20	0.20	0.20	
DKR 350 (by definition)	tonne	1	1	1	1	1	1
Electricity cl	GJ	0.31	0.32	0.32	0.31	0.31	
Flows	**Unit**	**S1**	**S2**	**S3**	**S4**	**S5**	**Reference**
Agent	tonne	3.32	4.27	4.31	3.12	3.32	
Product to Cyclone	tonne	4.72	5.67	5.71	4.72	4.72	
Product to HEX	tonne	4.66	5.61	5.65	4.66	4.66	
Product to clean	tonne	4.66	5.61	5.65	4.66	4.66	
Syngas to RT	tonne	0.16	0.52	0.00	0.16	0.17	
Flue gas to HEX	tonne	0.66	2.23	0.00	0.68	0.71	
Syngas	tonne	1.98	1.61	2.96	1.99	2.01	
Tar	tonne	0.33	0.33	0.33	0.33	0.33	
Outputs	**Unit**	**S1**	**S2**	**S3**	**S4**	**S5**	**Reference**
Solids	tonne	0.059	0.059	0.059	0.059	0.059	
Contaminants	tonne	0.011	0.011	0.011	0.011	0.011	
Water out	tonne	2.19	3.15	2.36	2.17	2.15	
Heat CHP	GJ	9.44	7.67	8.44	9.42	9.39	
Electricity CHP	GJ	5.49	4.46	4.91	5.48	5.46	
Flue gas CHP (not calculated)							
↳ CO2 in flue gas CHP	tonne	1.38	1.12	1.49	1.38	1.37	

Heat Inc	GJ	3.10	3.10	3.10	3.10	3.10	9.42
Electricity Inc	GJ	1.59	1.59	1.59	1.59	1.59	4.83
Flue gas Inc (not calculated)							
↳ CO2 in flue gas Inc	tonne	1.12	1.12	1.12	1.12	1.12	2.62
Flue gas RT	tonne	0.66	2.23	0.00	0.68	0.71	
↳ CO2 in flue gas RT	tonne	0.108	0.366	0.000	0.111	0.116	
Heat lost 1 (MJ/h)	GJ	0.50	0.49	0.50	0.52	0.56	
Heat lost 2 (MJ/h)	GJ	8.11	11.22	8.84	8.07	8.01	

3.5.3   Life cycle impact assessment

The impact of the gasification process is evaluated at midpoint since an evaluation at endpoint requires more assumptions and leads to a greater uncertainty in the results. The impact is only evaluated for the process itself and the production of the gasifier and infrastructure is not taken into account. This decision is made because the impact of these categories is expected to be minor in comparison. Furthermore, not incorporating these categories is common practise in LCA literature on gasification. Due to data availability and time constraints the only environmental impact category (EIC) which is evaluated in this research is global warming potential (GWP). Both gasification and incineration of plastic waste are carbon based energy intensive processes for which GWP is a relevant EIC to evaluate. Furthermore, all LCA literature on gasification referred to in this research evaluated this EIC, stressing the importance of this indicator. The unit of this EIC is kg CO_2-eq. The impact assessment method used is Environmental Footprint 3.0.

The ecoinvent consequential database of version 3.7.1 [65] found in SimaPro is used to acquire impact data for this LCA. The Netherlands is used as the reference location. If data is not available on the Netherlands, then representative data from a close by country or region is used. The processes which are selected for the input and output flows of the system are shown in Table 6.

Table 6: Global warming potential associated with the flows which cross the system boundaries

	GWP 100 (kg CO2 eq/unit)	Unit	Process
N2	4.11E-03	kg	Nitrogen, liquid {RER}| market for | Consequential
Water in	-9.92E-05	kg	Water, deionised {CH}| market for water, deionised | Consequential
Electricity	2.33E-02	MJ	Electricity, high voltage {NL}| market for | Consequential
Heat	1.60E-01	MJ	Heat, district or industrial, natural gas {RER}| market group for | Consequential
CO2	1	kg	By definition
Ash	3.19E-02	kg	Hard coal ash {CH}| treatment of, municipal incineration with fly ash extraction | Consequential

No GWP impact is attributed to the water output flow. This decision is made because in practise the waterflow which exits the CGS can be reused as the steam gasifying agent. Therefore, no waste treatment process is applicable for this flow. Also, in practice, further treatment of the contaminants (NH3, HCl and H2S) flow is required as it cannot be emitted in its current form. It is assumed that the contaminants flow can be recovered and reused, however, further research on these possibilities is needed. A cutoff approach taken as a second life is assumed, hence no impact is attributed to the contaminants flow.

4. Results

This chapter presents the results from this research. The first section is concerned with the model validation and the second section with the performed LCA.

4.1 Model validation

The composition of the main gasses in the product flow exiting the gasifier are presented in Figure 7. The key performance indicators (KPI's) of the gasifier can be found in Table 7. The model is able to decently simulate the carbon monoxide and carbon dioxide content of the syngas. However, the hydrogen content is significantly overestimated while the methane content is underestimated. Most tar compounds do not have hydrocracking and reforming reactions in the model. These reactions would both consume hydrogen and produce methane. This is a potential explanation for the difference between TUD case 3 and the model.

The KPI's show a similar trend where some KPI's are modelled with decent accuracy, while others are significantly different. In the model no carbon remains in the solids separated by the cyclone and a 100% CCE is thus achieved. This is close to the 99% efficiency reported by in TUD case 3. Both the lower CCE to gas and lower GY in the model results highlight that too much carbon is present in the tar output. Expanding the list of tar hydrocracking and reforming reactions in the model could potentially increase tar destruction and thus increase the accuracy of these KPI's. The CGE is accurately simulated by the model. The OE is 63.0% too high in the model results. This difference can at least partially be explained by the implementation of heat recovery for preheating the inputs instead in the model. While in the TUD research electric preheaters and a steam generator was used. Also, in the TUD research natural gas was burned in the radiant tubes instead of syngas.
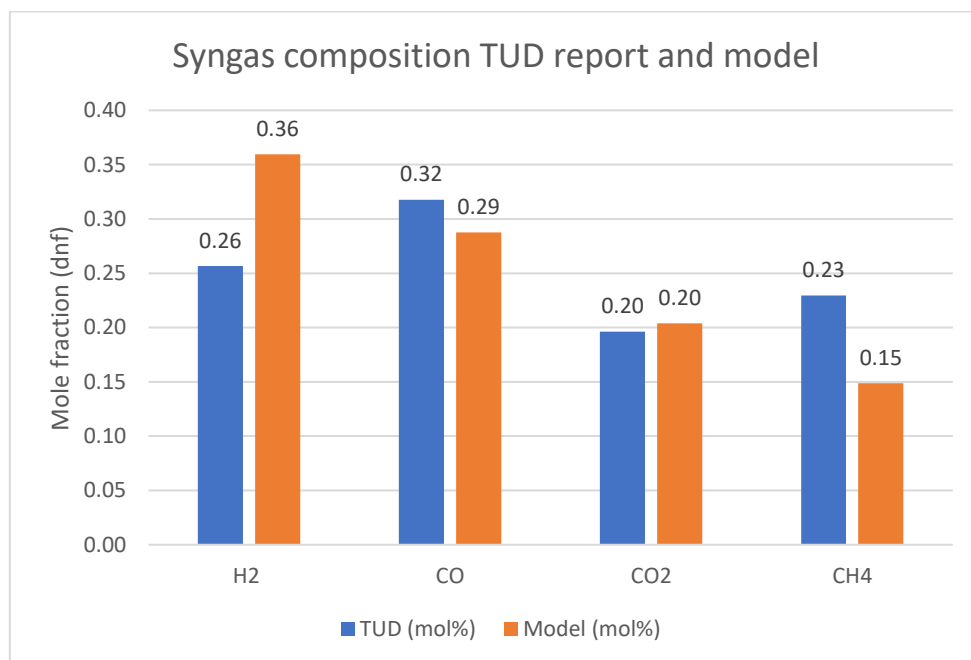

Figure 7: Comparison of the main gas composition in the syngas from the model and TUD case 3

Table 7: Comparison of KPI's from model results and TUD case 3 from the work by

KPI	TUD case 3	Model	Difference
CCE	0.99	1.00	1.0%
CCE gas	0.50	0.31	-38.7%
CGE	0.81	0.80	-1.1%
OE	0.47	0.77	63.0%
GY (Nm^3(dnf) kg^{-1}(daf))	0.96	0.68	-29.2%

4.2 Life Cycle Assessment

The results of the LCA are presented in this section. The impact of DKR 350 plastic waste is evaluated in terms of the GWP and the results for each scenario and the reference case are presented in Figure 8 and Table 8. These results show that the impact of the treatment of DKR 350 is reduced in all gasification scenarios compared to the reference scenario. This reduction in impact is primarily caused by the higher energy recovery efficiency for syngas in the CHP than from direct incineration of the plastic waste. The GWP reduction is most prevalent in scenario's 1, 4 and 5. A relatively smaller reduction is observed in scenario's 2 and 3. In these scenarios less syngas is produced and therefore fewer credits are received for energy recovery from the syngas. The impact of DKR 350 plastic waste is dominated by the emission of CO_2 which originates from the carbon which was present in the feedstock. It is observed that the impact, before subtracting the credits for energy recovery, is roughly equal in all scenarios, including the reference scenario. This is the case as the emission of CO_2 dominates the results and all carbon in the DKR 350 eventually leaves the model in the form of CO_2 in all scenarios. It is also observed that on average the impact of the gasifier itself is relatively small and even almost zero in scenario 3. This is because the CO_2 which is created by combustion reactions in the gasifier remains in the syngas and is only emitted after combustion in the CHP plant and thus the impact of this CO_2 is attributed to the CHP plant. The cleaning process does not significantly influence the overall impact. The heat required by the AGR is however supplied through heat recovery and therefore no impact for this heat is attributed to the AGR. If heat cannot be supplied by heat recovery then the impact of the cleaning process is expected to be larger.
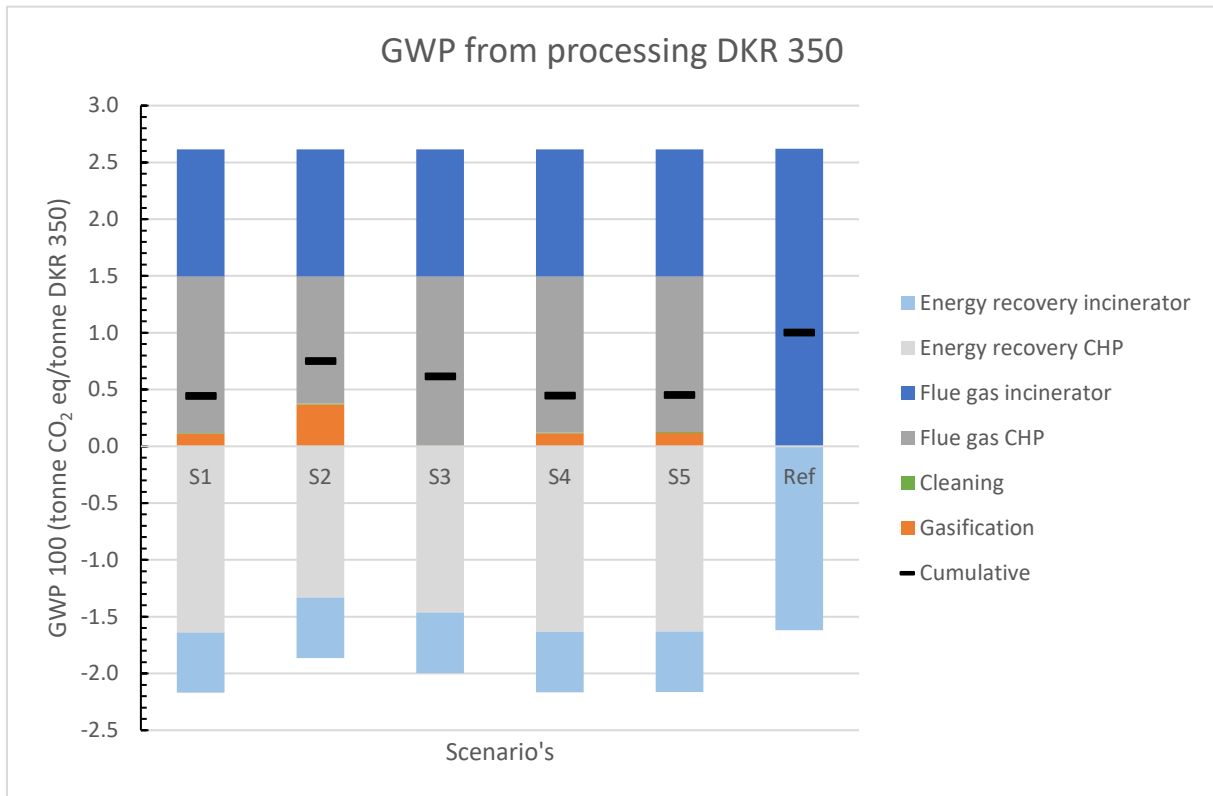
Figure 8: Cradle-to-grave GWP per unit process from gasification and incineration of DKR 350 waste

Table 8: Cradle-to-grave GWP per unit process from gasification and incineration of DKR 350 waste

Tonne CO2/tonne DKR 350	S1	S2	S3	S4	S5	Ref
Gasification	0.110	0.37	0.003	0.114	0.119	0.0
Cleaning	0.007	0.007	0.007	0.007	0.007	0.0
Flue gas CHP	1.4	1.1	1.5	1.4	1.4	0.0
Flue gas incinerator	1.1	1.1	1.1	1.1	1.1	2.6
Energy recovery CHP	-1.6	-1.3	-1.5	-1.6	-1.6	0.0
Energy recovery incinerator	-0.53	-0.53	-0.53	-0.53	-0.53	-1.6
Cumulative	**0.44**	**0.75**	**0.62**	**0.45**	**0.45**	**1.0**

5. Discussion

This chapter contains a discussion of this research. It starts with a sensitivity analysis on what the influence on the results will be if there is no demand for the produced heat. Thereafter, limitations and implications of the results are discussed.

5.1 Sensitivity analysis

It is assumed that heat can be sold however this might not always possible, especially in summer when demand for heat is relatively low. The heat will then be lost and no credits can be applied for it in the LCA. A sensitivity analysis is conducted on this uncertainty and the results are presented in Figure 9. Most of the applied credits in the original LCA are for the exported heat, therefore it is found that the results are very sensitive to whether there is a demand for heat. This sensitivity analysis shows that the GWP of gasification is very similar to incineration in all scenarios. Therefore, if the products of gasification are going to be incinerated and there is no demand for heat then gasification of plastic waste is not a good option to decrease GWP.

Decarbonisation of the energy system and especially heat production can result in a similar decrease in credits received for the exported heat and electricity. The government of the Netherlands has set a goal to reach climate neutrality in 2050 [21]. Therefore, it must be stressed that in the future, gasification of plastic waste should only considered if the products of gasification can be reused in an industry that prevents the emission of the contained carbon in the form of CO_2.
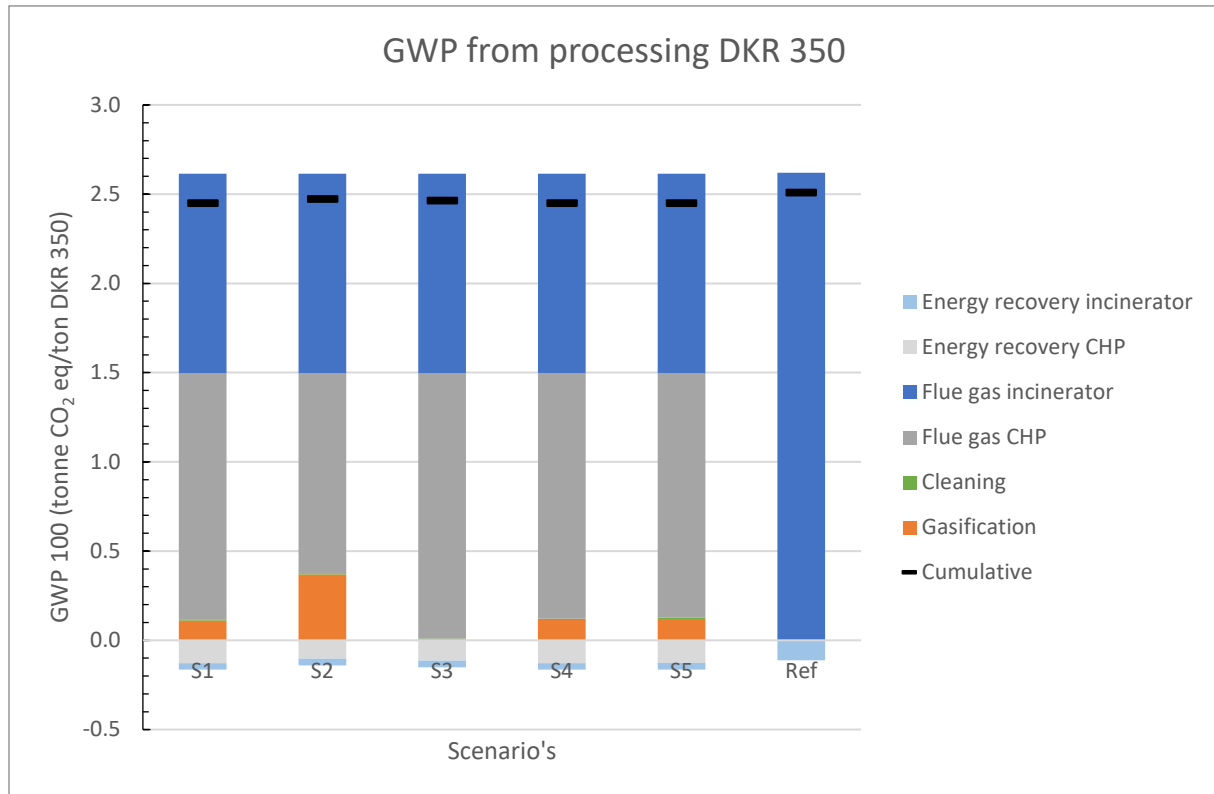

Figure 9: Cradle-to-grave GWP per unit process from gasification and incineration of DKR 350 waste when there is no demand for the produced heat

5.2 Limitations and implications

The LCA results show that gasification can potentially decrease the GWP potential from the treatment of DKR 350 plastic waste when compared to incineration. However, the LCA in this research is conducted with the results produced by the Aspen model. Therefore, inaccuracies and uncertainties in the Aspen model transfer over to the LCA results.

Validation of the Aspen model results was only carried out with one experimental case. This approach does not allow for validating whether the model results respond as expected to changes in reactor parameters. It is recommended that future research compares the model results to multiple experimental cases in order to validate whether the model responds correspondingly to changes in parameters. Based on literature it would be expected that all reactor parameters that were varied would impact the fraction of tars in the gasifier product stream. However, a similar tar flow is observed in all scenarios. A potential explanation here is that the lack of hydrocracking and reforming reactions of tar in the model cause the tar flow to not respond to changing reactor parameters. The lack of these types of reactions is also identified as a possible explanation for the differences observed between the Aspen model and TUD case 3. Therefore, it is recommended that further experimental research should focus on identifying the kinetic parameters of hydrocracking and reforming reactions of tar under similar conditions as found in the gasifier. This recommendation was also stressed in the work by Roman [35].

To increase the accuracy of the modelling of gasification and also LCA research on plastic waste gasification, future experimental research on plastic waste gasification is recommended to record the following data in addition to the data recorded in the work by Gilvari and De Jong [40]: The heating value of the syngas which is produced, the flow of natural gas or syngas which is burned to provide heat to the gasifier, the amount of electricity consumed by the preheating stage, the flow of nitrogen through the screw feeder and pyroprobe data on the pyrolysis of the feedstock as this can be used to update the pyrolysis correlations in the model. This last recommendation was also stressed in the work by Roman [35].

Furthermore, future LCA research is recommend to choose an application for the syngas in which the carbon in this gas is not emitted as CO_2. As such an application can be used in the transition to a circular economy. This approach comes with the additional benefit that constraints on the required composition of the syngas are known and thus the cleaning process can be modelled more thoroughly. If the produced syngas can be used to substitute some other feedstock or process this could potentially lead to considerable GWP savings as the gasifying process is shown to add little extra impact. However, this should be further investigated together with the applicability of the produced syngas as feedstock. Also, it is recommended that future LCA research evaluates other EIC's besides GWP.

6. Conclusion

The aim of this research is to provide an answer to the research question: *What is the global warming potential of gasifying DKR 350 plastic waste compared to incineration in the Netherlands and what are the key reactor parameters that influence this impact?* Based on the LCA the following conclusions can be drawn: Firstly, the impact on GWP of the gasification of DKR 350 plastic waste in the Netherland is approximately halve of the impact that incineration of this same plastic waste. Due to heat exchangers and the utilisation of the produced syngas as fuel, the process is mostly self-sufficient in its heat demand, creating no extra impacts. Furthermore, the cleaning process was shown to have a comparatively low impact. These two factors lead to only a minor amount of added GWP when plastic waste is fed through a gasification process. However, in gasifying the feedstock is converted from a solid into a gas. In general a gas can be incinerated with energy recovery at a higher efficiency then solids. So, the gasification process leads to little added impacts, but creates a product that has a higher energy conversion efficiency. Hence, gasification is observed to create less GWP impact compared to straight up incineration of DKR 350 plastic waste. Creating incentive for the deployment of gasification as treatment for contaminated plastic waste. Furthermore, when the created syngas can be used as feedstock for other processes next to incineration with energy recovery, then this creates an opportunity for closing the carbon loop. Secondly, increasing the reactor parameters SFR and ER are shown to increase the GWP impact. The reactor parameters air on the riser and temperature are shown to have little influence on the GWP of gasification. This points towards an optimum for the SFR ratio, as previous research has shown that a too low SFR leads to clogging issues. The SFR should thus be chosen at such a level that clogging issues are prevented and not be increased any higher than necessary. However, the above conclusions are subject to inaccuracies resulting from the Aspen model. Recommendation are made in the discussion section to increase the accuracy of both the Aspen model and the LCA. Implementing these recommendation will allow for verifying and increasing the significance of these results.

In addition to the LCA this research contributes through the validation and further development of the two-phase Aspen model on the gasification of DKR 350 plastic waste with experimental data. This process has shown that more experimental data is required to increase the accuracy of the Aspen model. However, future research should keep in mind that the initial goal of modelling is to provide a tool which can supplement and decrease the experimental research required. Therefore, if more

experimental research is required to develop an accurate model, than can be offset by the model, than the benefit of the modelling approach is decreased.

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8. Appendix

8.1 DKR 350 specifications

Product Specification 04/2009
Fraction-No. 350

Der Grüne Punkt –
Duales System Deutschland GmbH

DKR
Deutsche Gesellschaft
für Kreislaufwirtschaft
und Rohstoffe mbH

Sorting fraction:	M I X E D P L A S T I C S

A   Specification/Description

Used, completely emptied, system-compatible articles made of plastics that are typical for packaging (PE, PP, PS, PET) incl. packaging parts such as caps, lids, labels etc.

The supplementary sheet is part of this specification!

B   Purity

At least 90 mass % in accordance with the Specification/Description.

C   Impurities

Max. total amount of impurities	10 mass %

Metallic and mineral impurities with an item weight of > 100 g are not permitted!

Paper, cardboard	< 5 mass %
Other metal articles	< 2 mass %
PET bottles, transparent	< 4 mass %
PVC articles other than packaging	< 0.5 mass %
Other residual materials	< 3 mass %

Examples of impurities:
- Glass
- Composite paper/cardboard materials (e.g. beverage cartons)
- Other materials (e.g. rubber, stones, wood, textiles, nappies)
- Compostable waste (e.g. food, garden waste)

D   Delivery form

- Transportable bales

- Dimension and density of the bales must be chosen so as to ensure that a tarpaulin truck (loading area 12.60 m x 2.40 m; lateral loading height min. 2.60 m) can be loaded with a minimum loading of 21 t

- Dry-stored

- Produced with conventional bale presses

- Identified with DSD bale label stating the sorting plant No., fraction No. and production date

Figure 10: DKR 350 specifications

8.2 Aspen flowsheet original model

Figure 11: Aspen flowsheet original model (heat flows removed)
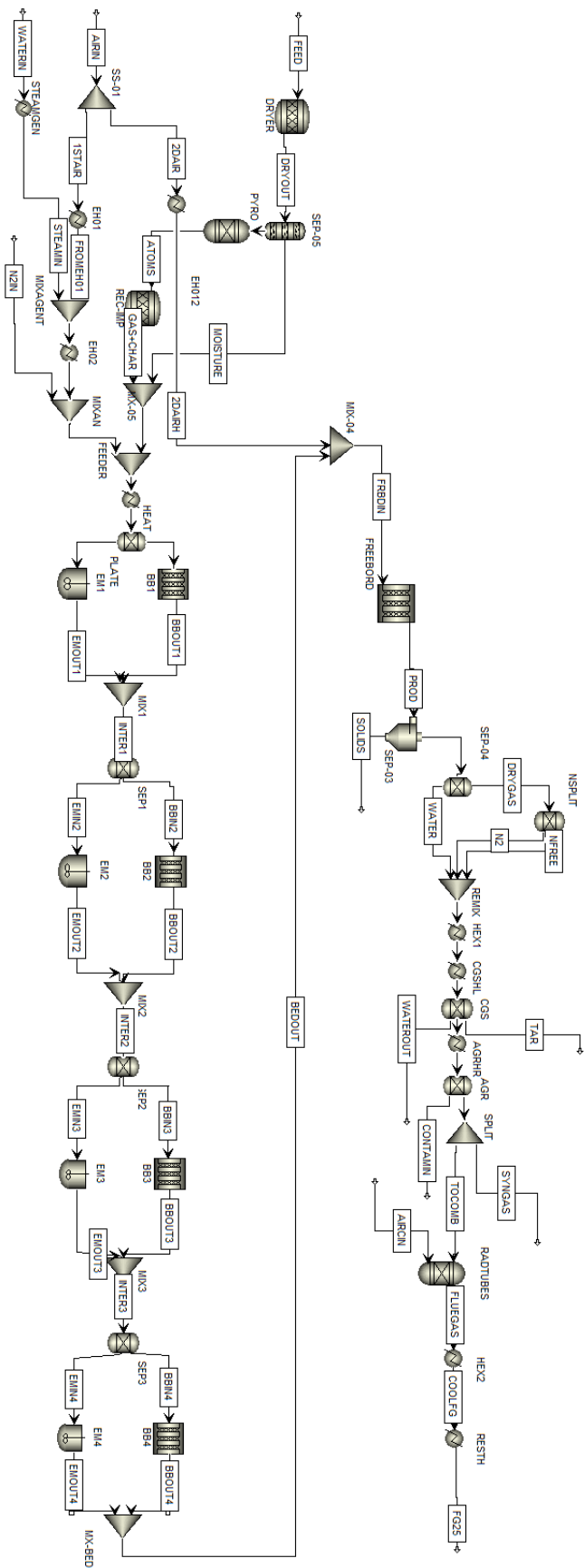
8.3 Aspen flowsheet updated model

Figure 12: Aspen flowsheet updated model (heat flows removed)

Table 9: Technical explanation of modifications made to original model

Change number	Detailed explanation
1	The calculator blocks labelled 'Pair 1' to 'Pair 4' play a crucial role in various calculations, including the determination of the volume for the CSTR blocks within the CSTR-PFR pairs. Initially, the calculated volume was written to a results variable. This causes the default value for the volume, which is entered in the CSTR blocks, to be utilized in the model calculation and the intended calculated volume to be written directly to the results. This is changed and the calculated volume is now used in the model calculations.
2	The input flow to the gasifier was split over the first CSTR-PFR pair based on the total relative volume of this pair. For the other 3 pairs a 50-50 split was used for the CSTR-PFR pair regardless of the relative volume. Now the flow is split over all pairs based on the relative catalyst free volume of these pairs.
3	The timing within a model run at which a calculator block needs to be executed can be specified in multiple ways in Aspen. By default this is set to "Use import/export variables", this setting was left at this default setting for most of the calculator blocks in the original model. This setting allows Aspen to automatically determine the correct moment for execution based on the availability of the import variables and demand for the export variables. Also, in the original model a sensitivity block was employed to vary certain model parameters and access the impact on the results. However, there is a bug in the Aspen software that causes the software to not always rerun calculator blocks in each loop of the sensitivity block even when this is required. This bug only occurs when the automatic default "Use import/export variables" setting is used SOURCE. This bug is found to occur in the original model. This is now prevented by manually specifying the timing at which each calculator block should be executed.
4	The rate equation of the following partial oxidation reaction of naphthalene: $C_{10}H_8 + 7O_2 \rightarrow 10CO + 4H2O$ was described to be temperature dependent in Romans thesis SOURCE. However, in the model the temperature dependency of this equation was left out. This is now changed.
5	The carbon monoxide combustion reaction was deactivated in the freeboard in the original model. This reaction was intended to be set to active as this was mentioned in Conrado's thesis. Therefore, this is now changed.
6	The model is found to suffer from mass convergence issues which causes the amount of mass which enters and exits the CSTR blocks of the bed zone to not always be the same. There is a page on this issue with the Aspen Plus Help "RCSTR Mass Balance Convergence Failures". All options mentioned on the page are tried but this did not solve the issue. To decrease the complexity of convergence of the results the following change was made to the model. The pre exponential factor for the char combustion reaction was calculated using the char content before and after the bed zone. However, the char combustion reaction occurs in the bed zone. Using a value in the model which is not yet available at the moment of usage results in a tear variable. Aspen then runs multiple loops within the model in which the value for the tear variable is taken from the previous run of this loop. This process significantly complicates convergence. It is found that the model results are not significantly changed when the char content after the bed zone is not used in the calculation of the pre exponential factor for the char combustion reaction and

| | | instead replaced by a static value. Thus this change is made to simply the convergence process. |
| | | • Still a workaround is required to continue with the convergence issue in the model. Whenever the results fail to converge Aspen continues the run with the results of the last iteration. However, the mass balance error varies erratically between iterations. In some iterations the error is negligibly small and in some large enough to significantly impact the results of the model. The following approach was used as a workaround: The maximum number of iterations for each of the CSTR blocks in the bed zone is set to 100. The model is then run and the iteration with the smallest root mean square error is chosen and the maximum number of iterations is changed to this number of iterations. The model is then rerun. This workaround needs to be done in sequential order for each block which runs into this convergence issue. |