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TNO report

TNO 2020 R11466 | Final report

RECOVER (TBBE218003) Public Report

Date	30 September 2020
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Number of pages	27 (incl. appendices)
Number of appendices	0
Sponsor	The Dutch Ministry of Economic Affairs is gratefully acknowledged for the funding of this TKI project (TBBE 218003)
Project name	RECOVER
Project number	060.36035

Managementuittreksel

Titel :	RECOVER: Terugwinnen van producten uit kunststof afval
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Datum :	30 september 2020
Opdrachtnr. :	TBBE218003
Rapportnr. :	TNO 2020 R11466

Het project is uitgevoerd met subsidie van het Ministerie van Economische Zaken, Nationale regelingen EZ-subsidies, Topsector Energie uitgevoerd door Rijksdienst voor Ondernemend Nederland

Summary

‘RECOVER – Recovery of products from plastic waste’ (060.36035) is a TKI innovation project financed by RVO (reference TBBE 218003) in which technologies developed by the consortium partners were used to convert three plastic waste streams into valuable products. These products include benzene, toluene, xylene (BTX), ethylene and propylene.

The gasification and BTX scrubbing technologies developed at TNO were used to gasify the plastic waste streams provided by AEB (DKR310 Big foils, DKR310 Small foils, Mixed Plastics) and recover the value they contain, exploring the conversion and product recovery from the feedstock. For this particular task the knowledge provided by SRT was crucial for the test programme. The three waste streams were successfully converted into a gas containing valuable products in the Milena gasifier. Ethylene followed by methane are the main components produced regardless of the feedstock used. Also mass yields of at least 6% are obtained for benzene. A conversion of 66-71% was achieved into product gas on a carbon basis.

Experimental studies on the catalytic pyrolysis of selected plastic streams using small-scale set-ups to optimize the yields of BTX products were performed at RUG with relevant knowledge provided by BioBTX. The work using LDPE shows that the pyrolysis temperature has a major effect on the BTX yields and a higher yield is obtained when using a high pyrolysis temperature. The experiments using DKR310 Small foils and H-ZSM-5 as the catalyst show limited signs of catalyst deactivation after 10 plastic injections. The experimental studies for the catalytic pyrolysis of LDPE, show that of the two selected catalysts in this study (H-ZSM-5 and a standard FCC catalyst), the H-ZSM-5 catalyst performs better than the FCC.

In this project the focus lies on these first market applications where the technology can be used to chemically recycle waste streams and utilize existing infra structure to facilitate the implementation. AEB and SABIC are two good examples of these first locations where chemical recycling can be shown, without the need of a complete standalone unit developed. Moreover for AEB a process flow diagram was developed, highlighting how the technologies can be implemented in their process to achieve a higher recycling efficiency. For SABIC a similar result was derived, highlighting how the technologies can be implemented in a cracker setting, making use of the vast infrastructure available for further cleaning and upgrading towards products. The research and development work was complemented with a simplified Techno-Economical Evaluation (TEE). These implementation tasks were performed by SRT and Synova taking into account the experimental results as well as the input from all partners involved.

With regards to the technical and economic viability, two concepts were evaluated: (1) M6 scale with 1.1 tonne/h plastic waste conversion to BTX liquid and electricity with existing gas engines at AEB, and (2) M30 scale with 6 tonne/h plastic waste conversion to a olefin-rich gas ready to be injected into cold separation units downstream existing naphtha cracker plants of Sabic. From a simple economic analysis, it follows that the small-scale BTX plant can only be economically viable when all parameters are chosen in the extreme of the ranges considered. The BTX plant hence preferably should be larger to benefit from economy-of-scale. For the

Waste-to-Olefins case, the base case already shows (very) attractive economics. This is the result of the larger scale, higher yield of valuable products, and the use of existing downstream processing for the production of polymer-grade monomers.

This report is the public version of the main achievements of the project.

Project reference number	TBBE 218003
Project title	RECOVER: Terugwinnen van producten uit kunststof afval
Project partners	TNO (Coordinator), AEB, BioBTX, RUG, SABIC, Synova, SRT
Project duration	01-01-2019 / 30-06-2020

Contents

1 Introduction

The social pressure on brand owners (partly because of plastic waste in environment/ocean, partly CO_2 emissions) is rapidly increasing. Companies cannot afford to do business as usual. In EU, targets are defined for (plastic) packaging, and individual companies formulate their own targets. At the same time, this offers unique opportunities for the European chemical industry that generally has difficulties to compete against (larger) units operating on cheap feedstock.

The Netherlands has a limit on landfilling, which resulted in a high recycling efficiency for waste (> 30%), although still a large part of the Dutch waste ends up in a combustion/incineration facility. Packaging material has a recycling efficiency of over 45% through mechanical recycling, which puts the Netherlands in the third place in Europe[1]. Mechanical recycling can be improved for certain waste streams, but there will be limitation to this. The Dutch government has expressed the ambition to increase the recycling efficiencies. Chemical recycling will play an important role in reaching higher circularity targets and provide a solution for food packaging. However, some hurdles still have to be tackled and research is needed to make chemical recycling cost competitive. The Dutch chemical industry is not yet equipped to take in different feedstocks for the production of plastics and chemical recycling has to compete with incineration. In order to make chemical recycling feasible and economically viable the Dutch government is funding research projects such as the current one. An important question that this project will answer is:

- Can pyrolysis/gasification provide a good recycling alternative for materials that cannot be mechanically recycled, offering a final product efficiency of at least 70% on a hydrocarbon mass basis?

1.1 Project Overview

Project reference number	TBBE 218003
Project title	RECOVER: Terugwinnen van producten uit kunststof afval
Project partners	TNO (Coordinator), AEB, BioBTX, RUG, SABIC, Synova, SRT
Project duration	01-01-2019 / 30-06-2020

TNO, SRT, Synova, BioBTX and RUG have developed technologies that allow efficient conversion of various feedstocks, including waste streams, into valuable products. These products include benzene, toluene, xylene (BTX) and others. These technologies can also be used for chemical recycling. In this project the focus lies on these first market applications where the technology can be used to chemically recycle waste streams and utilize existing infra structure to facilitate the implementation. AEB and SABIC are two good examples of these first locations where chemical recycling can be shown, without the need of a complete standalone unit developed.

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[1] Plastics the facts 2017, PlasticsEurope

This project will explore how the technologies will operate with the feedstock chosen for the purpose and how the process can be optimized for the specific feeds.

2 Pyrolysis and Catalytic Conversion for End-of-Life Plastics

This work package (WP2) involves experimental studies on the ex-situ catalytic pyrolysis of selected end-of-life plastics. This ex-situ approach involves the initial thermal pyrolysis of the plastic feed followed by a catalytic vapor upgrading using appropriate aromatization catalysts. The following objectives were set:

1. Determination of the optimum temperatures for the thermal pyrolysis and subsequent catalytic upgrading of the vapor
2. Insights in the stability of the catalysts and development of measures to reduce deactivation levels
3. Selection of preferred catalyst based on BTX yield and stability, to be used as input for WP4
4. Mass balances for the catalytic pyrolysis process.

Experimental studies to reach objectives 1, 2, and 3 were carried out in a tandem microreactor (TMR) coupled to a GC/MS. This device, with feed intakes in the order of mg's, allows rapid testing of conditions and catalysts and quantification of the BTX yields. Two commercially available aromatization catalysts (H-ZSM-5(23)) and a ZSM-5 based FCC catalyst were used, as this will simplify further scale-up activities in subsequent WP's. The effects of the temperature of the thermal pyrolysis, the temperature of the catalytic aromatization step, and the plastic – catalyst ratio were optimized for a model plastic (LDPE) to obtain the highest BTX yields, and the results were quantified using statistical modelling approaches. For objective 4, the TMR findings were scaled up to g scale in a dedicated unit, which also allows for mass balance investigations (liquid, char, and gas formation.

Experimental studies with model plastics (LDPE, HDPE, PP, PBT-PET) were carried out in TMR and g scale unit, whereas experiments with the different DKR310 samples were performed in the TMR unit.

2.1 Objective 1: Determination of the optimum temperatures for the thermal pyrolysis and subsequent catalytic upgrading of the vapor

A systematic experimental study involving 20 individual experiments using LDPE in the TMR (Figure 2.1) show that the pyrolysis temperature has a major effect on the BTX yields and a higher yield is obtained when using a high pyrolysis temperature. The effect of the catalytic aromatization reaction is less pronounced.

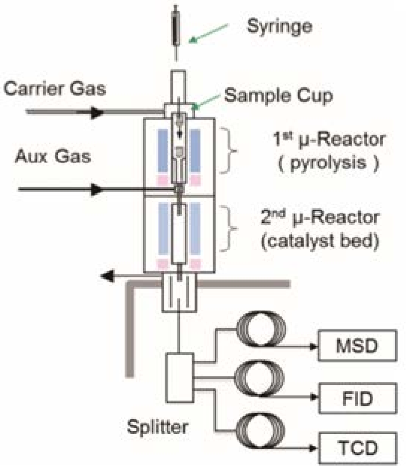

Figure 2.1: Schematic diagram of the micro-pyrolysis system

2.2 Objective 2: Insights in the stability of the catalysts and development of measures to reduce deactivation levels

Experiments performed in the TMR using DKR-310 (small foils) as the feed and H-ZSM-5 as the catalyst show limited signs of catalyst deactivation after 10 plastic injections. Despite some scatter in the data, it is evident that the BTX yield is approximately stable.

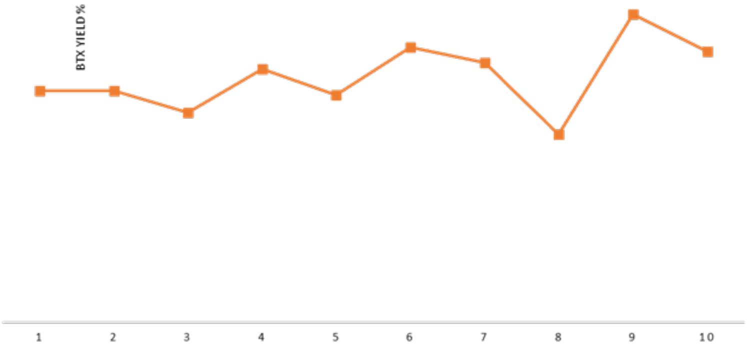

Figure 2.2: BTX yield versus the number of plastic injections (TMR, DKR-310, big foils)

2.3 Objective 3: Selection of preferred catalyst based on BTX yield and stability, to be used as input for WP 4

Based on the experimental studies for the catalytic pyrolysis of LDPE, it is concluded that of the two selected catalysts in this study (H-ZSM-5 and a standard FCC catalyst), the H-ZSM-5 catalyst performs better than the FCC one.

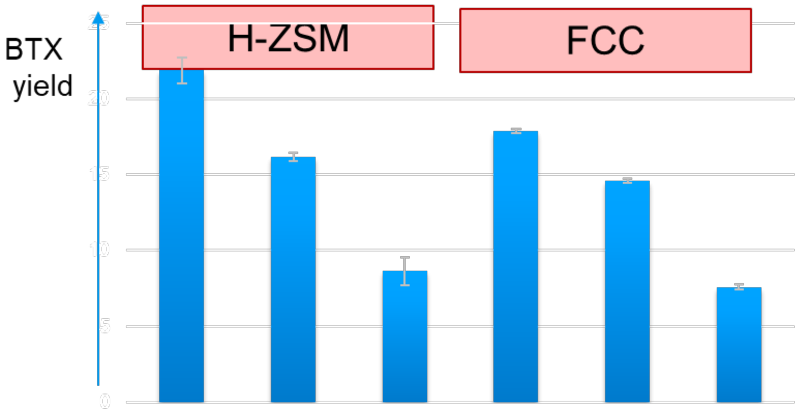

Figure 2.3: BTX yield for different catalysts

2.4 Objective 4: Mass balances for the catalytic pyrolysis process

Pyrolysis experiments with LDPE were performed in a gram-scale unit in batch mode to determine the mass balance closures (Figure 2.4). Mass balance closures were very satisfactorily and above 90%. Only minor amounts of (carbonaceous) residues were obtained, in line with the TGA data for LDPE.

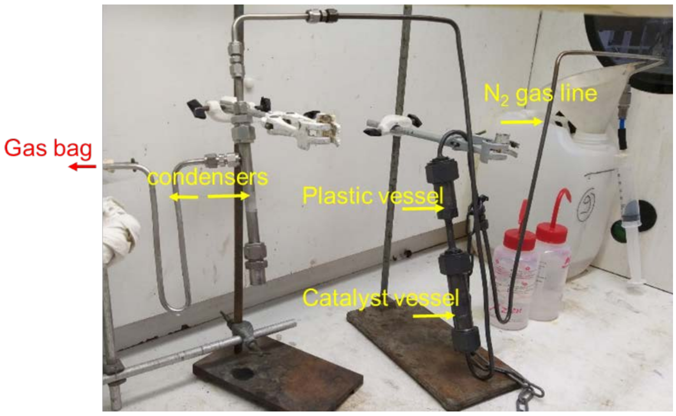

Figure 2.4: Picture of the gram-scale setup

3 Gasification and Tar Removal for End-of-Life Plastics

The RECOVER project aims the production of valuable products such as BTX from the gasification of end-of-life plastics. Three different plastic streams were provided by AEB which were obtained directly from their Post Separation facility:

- DKR310 Small Foils, 2D
- DKR310 Big Foils
- Mixed Plastics

3.1 Experimental Work

Three different end-of-life plastic streams were provided by AEB which were obtained directly from its Post Separation facility. The following figure shows these streams as received at TNO facilities.

Figure 3.1: The three plastic streams “as received” at TNO: A) DKR310 Small Foils (2D); B) DKR310 Big Foils; C) Mixed Plastics

These streams had to be pre-processed at TNO in order to be fed into the lab-scale Milena gasifier. This is a feature in the lab-scale reactor due to the small size of the orifice that goes into the riser. Hereby, all the received streams had to be shredded, pelletized (6 mm die) and then milled (8 mm sieve), as schematized in the following figure:

Figure 3.2: Pre-processing of the waste plastic streams prior to the gasification tests

At commercial scale only shredding is expected (no need for pelletizing and further milling). Nevertheless at lab-scale there was the need for the pre-processing. The following figure shows the milled streams prior to the gasification tests.

Figure 3.3: Milled plastic streams prior to the gasification tests. Left) DKR310; Right) Mixed Plastics

The experimental work was carried out in the lab-scale Milena (25 kW_{th}), depicted in Figure 3.4. Milena is an indirect, fluidized bed gasifier, meaning that the product gas is not diluted with N_2 from combustion. It consists of a riser, where the fast devolatilization/gasification of the solid feedstock takes place, and a BFB combustor, where the remaining char is burned. In the settling chamber, solids (char and bed material) are separated from the product gas and recirculated to the combustor via the downcomer. Heat is transferred between the combustor and the riser through the circulation of bed material.

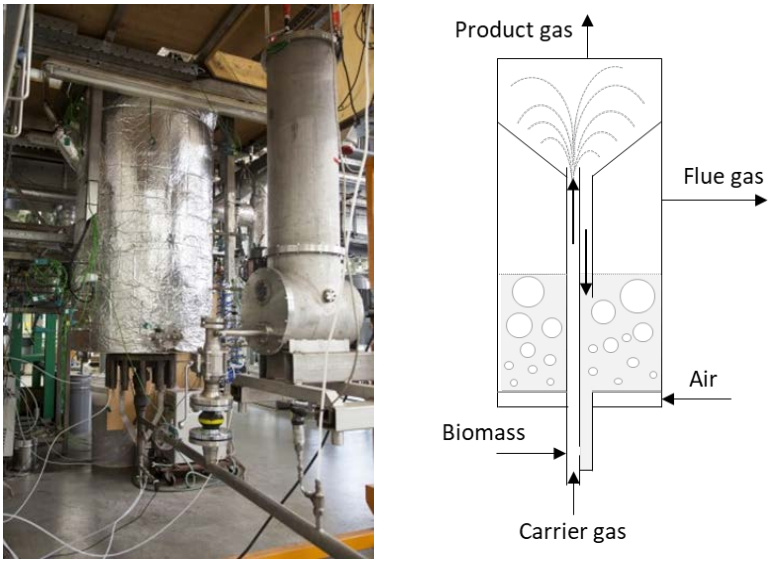

Figure 3.4: Scheme of the lab-scale Milena gasifier (25 kW_{th})

3.2 Results

The following figures will show the summary of the results obtained with the lab-scale Milena gasifier with the three different feedstocks. Figure 3.5 shows the ultimate analysis of the tested plastic waste streams.

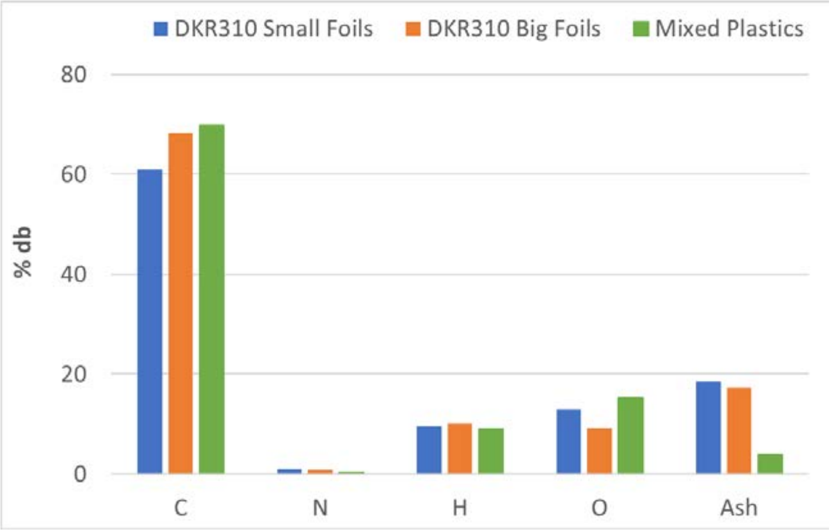

Figure 3.5: Feedstock analysis of the tested end-of-life plastic streams

The composition of the different streams is similar except for the ash content, which is lower for the Mixed plastics than for the DKR310 streams. There are also some minor differences among the DKR310 fractions. Big foils have a larger carbon content and less oxygen than Small foils.

Figure 3.6 shows the overall carbon mass balance from the gasification tests. Here it can be seen where the carbon present in the original feedstock will be present at the end of the gasification tests.

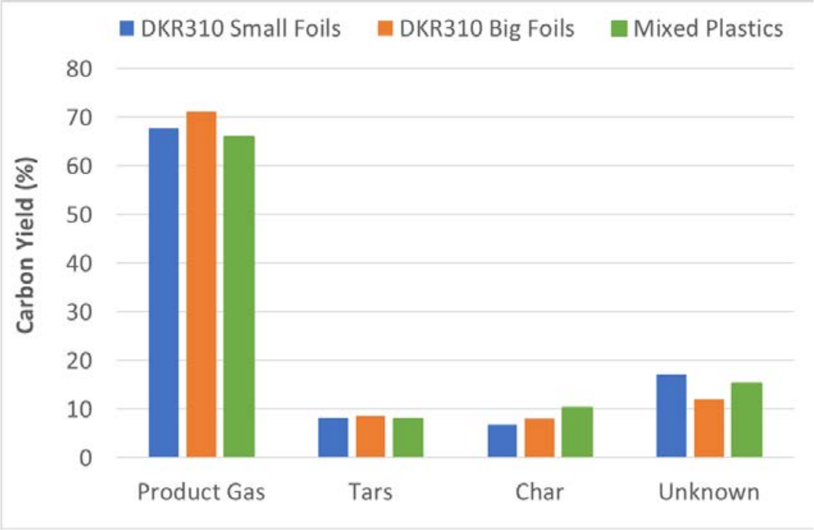

Figure 3.6: Carbon mass balance in the gasification of the plastic streams

The results are very similar among all tested materials, though more char seems to be generated with Mixed Plastics. Also, more gas seems to be produced with DKR310 Big foils. The total measured carbon results in values around 85% on average. Figure 3.7 shows the carbon yield of the four main components present in the product gas.

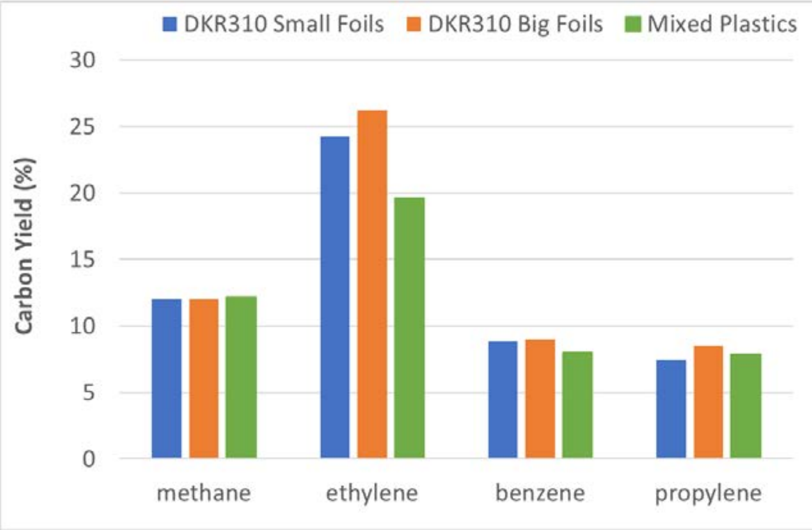

Figure 3.7: Carbon yield of some components present in the product gas from gasification of different plastics waste streams

The carbon yields are very similar with the difference of ethylene which the DKR310 streams (Big foils and Small foils) generate more. This is a probable result of the larger amount of PE present in the DKR310 streams (about 70%) as compared to Mixed Plastics (about 30%). Figure 3.8 shows the carbon yield of the other less prominent components.

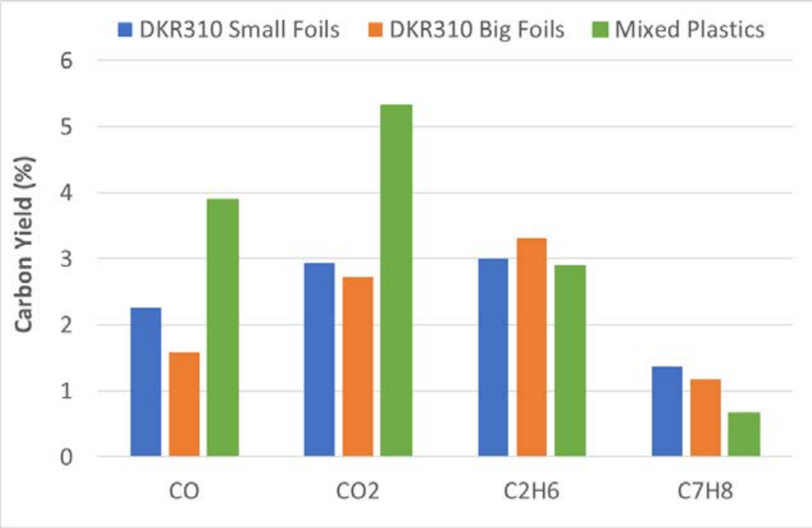

Figure 3.8: Carbon yield of some components present in the product gas from gasification of plastic waste streams

More CO and CO_2 is generated with Mixed Plastics than with the DKR310 streams, almost two times more. This is a result of the larger amount of oxygenates (biogenic components like paper and food, but also PET and polycarbonates) present in the Mixed Plastics as seen the feedstock analysis shown in Figure 3.5. The amount of ethane is similar to all streams and less toluene is generated with Mixed Plastics.

It can be concluded that ethylene is the component present in the highest amount regardless of the feedstock, though its presence is higher in case of DKR310 samples. About 50% of the carbon in the feedstock ends as ethylene, methane, propylene and benzene. Mixed plastics generate almost two times more CO and CO_2 though still with values below 10% in terms of carbon yield.

3.3 Impact indicators

The recycling efficiency of each feedstock is translated with the use of the Impact indicators provided by CE Delft[2].

- Indicator 1: Climate Advantage
- Indicator 2a: Resource score
- Indicator 2b: Feedstock value indicator

These indicators rely on a simplified Life Cycle analysis (LCA) based on several assumptions.

3.3.1 Indicator 1: Climate Advantage

The Climate Advantage indicator is calculated according to the following formula:

Formula 1
Climate Advantage = Climate Impact AEC – Climate Impact CR

Both terms in the formula above are calculate according to formula 2:

Formula 2
Climate Impact = Direct Emissions + Energy & Materials – Avoided Products

Table 3.1 shows the final results. The chemical recycling brings a positive impact in the climate advantage to all tested feedstocks. It can also be seen that DKR310 Big foils and Mixed plastics show a larger climate advantage than DKR310 Small foils.

Table 3.1: Climate advantage results, ton $CO_{2\text{-eq.}}$ per ton processed waste

	DKR310 Big Foils		*DKR310 Small Foils*		*Mixed Plastics*	
	AEC	CR	AEC	CR	AEC	CR
Direct Emissions	1.91	0.25	1.74	0.23	2.07	0.32
Energy & Materials	0.03	0.05	0.03	0.05	0.03	0.05
Avoided Products	0.54	0.81	0.50	0.69	0.56	0.74
Climate Impact	1.41	-0.51	1.27	-0.41	1.54	-0.37
Climate Advantage	1.91		1.68		1.91	

3.3.2 Indicator 2a: Resource Score

This indicator corresponds to the ratio of how much new products are generated from the plastic waste. It is calculated according to Formula 3

Formula 3
Resource Score = Products (kg) / Plastic Waste input (kg)

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[2] “Handleiding screening LCA: Vergelijken tussen chemische recycling en AEC-verbranding van afgedankt kunstof” CE Delft, 2018

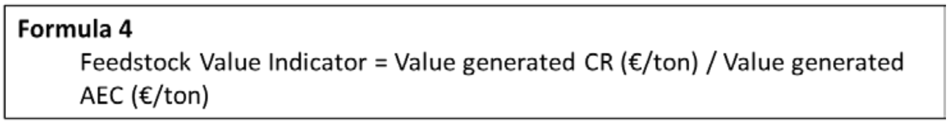
The results are presented in Table 3.2. According to this indicator the DKR Big foils show the highest resource score while Mixed plastics show the lowest value.

Table 3.2: Resource score results

	DKR310 Big Foils	*DKR310 Small Foils*	*Mixed Plastics*
Products (kg/ton waste)	478	410	450
Plastic Waste (kg/ ton waste)	661	652	769
Resource Score	0.72	0.63	0.59

3.3.3 Indicator 2b: Feedstock Value Indicator

This ratio indicates how much value is generated by chemical recycling and is calculated according to Formula 4. Table 3.3 shows the results. Following the previous conclusions also according to this indicator the DKR310 Big foils presents the highest value.



Formula 4

Feedstock Value Indicator = Value generated CR (€/ton) / Value generated AEC (€/ton)

Table 3.3: Feedstock value indicator

	DKR310 Big Foils	*DKR310 Small Foils*	*Mixed Plastics*
Value generated AEC (€/ton waste)	263	245	278
Value generated CR (€/ton waste)	302	248	256
Feedstock Value Indicator	1.15	1.01	0.92

3.4 Conclusion

All the three feedstocks were converted into a gas containing valuable products successfully. Ethylene followed by methane are the main components produced regardless of the feedstock used. Also, mass yields of at least 6% are obtained for benzene. A conversion of 66-71% was achieved into product gas on a carbon basis. The composition of the gas generated is very similar among the three feedstocks with the exception of ethylene and oxygen compounds such as CO and CO_2. The DKR310 fractions generates more C_2H_4 and less CO and CO_2 as compared with the Mixed plastics fraction. Among the three feedstocks, the DKR310 Big foils lead to better results with corresponding highest impact factors (climate advantage, resource score and feedstock value indicator of 1.91, 0.72 and 1.15 respectively).

4 Industrial Design of Gasifier and Tar Removal for End-of-Life Plastics

4.1 MILENA Gasifier

4.1.1 Conventional MILENA design

MILENA is an indirect gasifier operating similarly to a fluid catalytic reactor (FCC) concepts: feedstock is heated and gasified in a circulating flow of hot sand and the less reactive remaining solid char is directed to the combustor where the circulating sand is heated.

The MILENA design is however different to most of the FCC reactors as it has a more integrated design. The gasification or cracking of the feedstock takes place in a central riser, whereas a surrounding bubbling fluidised bed serves as combustor. The two reactors are integrated as schematically shown in Figure 5.1. One similar integrated design can be found in FCC applications as well in the Orthoflow^{TM} designs by Kellogg Brown & Root (KBR).

Solid feedstock is fed into a bubbling fluidised bed at the bottom of the riser (1). This section is normally fluidised with air as only a small amount of gas is needed for this. As the hot bed material is cracking the feedstock, additional gas is being generated causing an increase of gas flow (2) and creating a spouting fluidised bed in the top part of the riser (3).

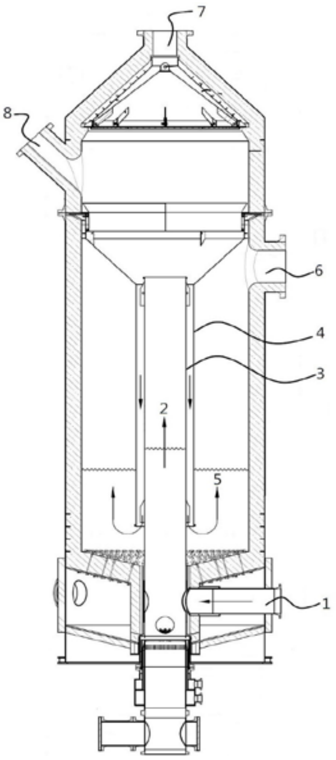


Figure 5.1 MILENA reactor

Above the riser, the velocity in the riser drops causing the unconverted char to fall via the down-comer (4) into the combustion zone. This combustion zone is operated as a bubbling fluidized bed with limited gas velocity (5) operated on air. As of the two reaction zones, the MILENA has two gas outlets being flue gas from combustion (6) and product gas (7) from which high value chemicals are being extracted.

The start-up in the MILENA is done in combustion mode, which means more fluidization air and less feedstock in order to generate the same amount of gas needed to establish the internal circulation. During start-up, the gas (flue gas) is flared. In order to accommodate different flows of air during operation (fluidisation air) and start-up (start-up air), the riser of the MILENA is equipped with two air inlets.

4.1.2 MILENA design with end-of-life plastics

When utilizing plastic waste like DKR310 as feedstock for chemical recycling, no significant changes are required to the design of the MILENA reactor itself. Only a combined start-up air / steam connection would be needed with a steam trap on that connection. The higher thermal load of the MILENA does require some additional cooling capacity on the flue gas and product gas coolers as well as the oil cooling capacity of the OLGA tar removal technology.

4.2 OLGA Tar Removal

4.2.1 Conventional OLGA design

The tar removal system of OLGA as shown in Figure 4.2 is based on a multistage scrubber in which gas is cleaned by a special scrubbing oil. The use of multistage scrubbing technology makes the OLGA tar removal a more robust technology compared to alternative processes and creates freedom to optimize the MILENA, e.g. optimizing towards higher content of High Value Chemicals (HVC) components as these, unlike tar components, are not scrubbed from the gas by the conventional OLGA.

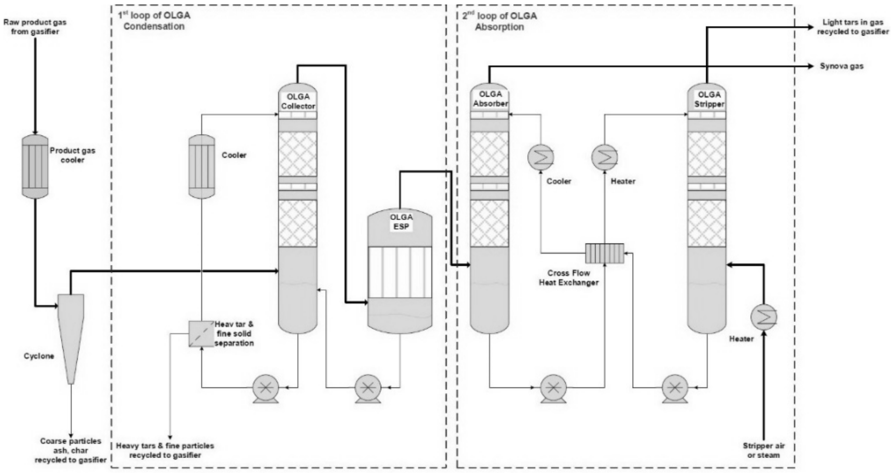


Figure 4.2: Scheme of the OLGA tar removal system

In the 1st loop of OLGA, the gas is quenched by scrubbing oil. Heavy tar particles condense and are collected as liquid, ready to be recirculated to the gasifier. This quenching of gas is also done in a similar way in large-scale coke oven gas systems in order to remove the heavier tars by condensation.

In the 2nd loop, lighter tars are absorbed by scrubbing oil at a temperature above the water dewpoint of the gas, this to avoid the mixing of water and tars. In the absorber column the scrubbing oil is saturated by these light tars. This saturated oil is regenerated in a stripper. Absorption/desorption processes are common in industry, in general like amine scrubbers but also specifically for hydrocarbons again in the coke oven gas industry. Hot air or steam is used to strip the tar out of the scrubbing oil.

All heavy and light tars can be recycled to the gasifier where they are converted and contribute to the energy efficiency. Plus, a tar waste stream is avoided. Water can be condensed from the gas in water scrubbers downstream the OLGA. As tars have been removed by absorption to a tar dew point way below the water dewpoint, this condensation of water is not associated with condensation of tars.

4.2.2 OLGA design with end-of-life plastics

Despite the MILENA being operated on DKR310 with steam being added to the riser, the moisture content of the gas and hence the water dewpoint of the gas is similar to that of gasifying biomass or RDF as the DKR310 itself has hardly any moisture. As such, operating conditions of the OLGA tar removal system don’t have to be modified. The increased thermal capacity of the M6 MILENA does require more cooling of oil in the OLGA as well as the product gas coolers upstream the OLGA. The flue gas cooler of the MILENA can remain within existing design.

5 Industrial Design of Catalytic Conversion and BTX Recovery

5.1 MILENA as Fluidized Bed Aromatization Reactor

Fluidized bed aromatization might be of interest as even small amounts of coking on the catalyst lead to the need for frequent regeneration which might suggest operating in a dual fluid bed mode. This leads to an analogy with a conventional fluid catalytic cracking (FCC) unit as well as with MILENA. Compared to the conventional design of the MILENA there are however several parameters that differ from the gasification of solid feedstock.

First, there is very little heat input expected to support the endothermic aromatization. If anything, the regeneration is exothermic. The combination of light coking and the assigned heats of formation might lead to a very modest heat withdrawal from the circulating catalyst. As of the expected increase in the ratio product gas to combustion air, it is expected that the reactor needs to be sized on the diameter of the settling chamber. The combustion zone however can no longer maintain the same diameter and should become smaller creating a mushroom shaped reactor as shown on the right. The expected longer residence time of several seconds needed for the aromatization will, despite the advantage of a lower minimum fluidization velocity, result in an increased length of the riser. The height of the settling chamber in addition is expected to increase as well.

5.2 OLGA as BTX Recovery Unit

The BTX recovery unit tested by TNO is comparable with the OLGA system, though operated at lower temperature and with more steam being used for stripping the BTX from the absorption oil. As of the condensation of water from the gas upstream the BTX scrubber, the volume flow through the absorber of the BTX recovery is approximately 60% of that through the OLGA absorber. For the stripper of the BTX recovery this is approximately 20% of that through the OLGA stripper. This is partially also related to the fact that with air more gas is needed to assure that the stripper column is operated safely below the LEL of tars. When stripping with steam this is not an issue.

In order to have similar hydrodynamics as in the OLGA, the absorber column of the BTX recovery would despite the lower gas flow need to be approximately 7.5% bigger in diameter as of the high oil flow and the risk of flooding. For the stripper as of the significant lower gas flow, the diameter can become approximately 20% smaller. Neglecting the required height of the packing, which is assumed to be rather similar for the BTX recovery unit compared to the OLGA, the columns of the BTX recovery unit would become significantly higher as of the additional sump required for the higher oil flows. This makes it safe to assume that the absorber-stripper section of the BTX recovery unit will be more expensive than for OLGA, not yet considering the need for a steam boiler and a BTX condensation and separation unit to recover the BTX from the steam.

6 Fitting Chemical Recycling into Existing Infrastructure

6.1 Waste-to-Energy Plant

A process flow diagram of the integration of a single M6 MILENA-OLGA plant with the waste-to-energy plant is evaluated and depicted in Figure 6.1. The feedstock considered is DKR310 films. The cleaned gas is compressed and sent to a conventional olefin aromatization reactor with the produced BTX and paraffins being collected by means of condensation. This liquid product finds its offtake in existing separation units of Sabic as additive to the existing pygas stream.

The flue gas from the MILENA normally is cooled down and cleaned by means of a bag house filter. A further integration with the AEB could be possible by injecting this flue gas directly into the existing flue gas treatment line of the AEB. The capacity of the existing AEB flue gas treatment is large enough to handle this additional flow of flue gas from the MILENA.

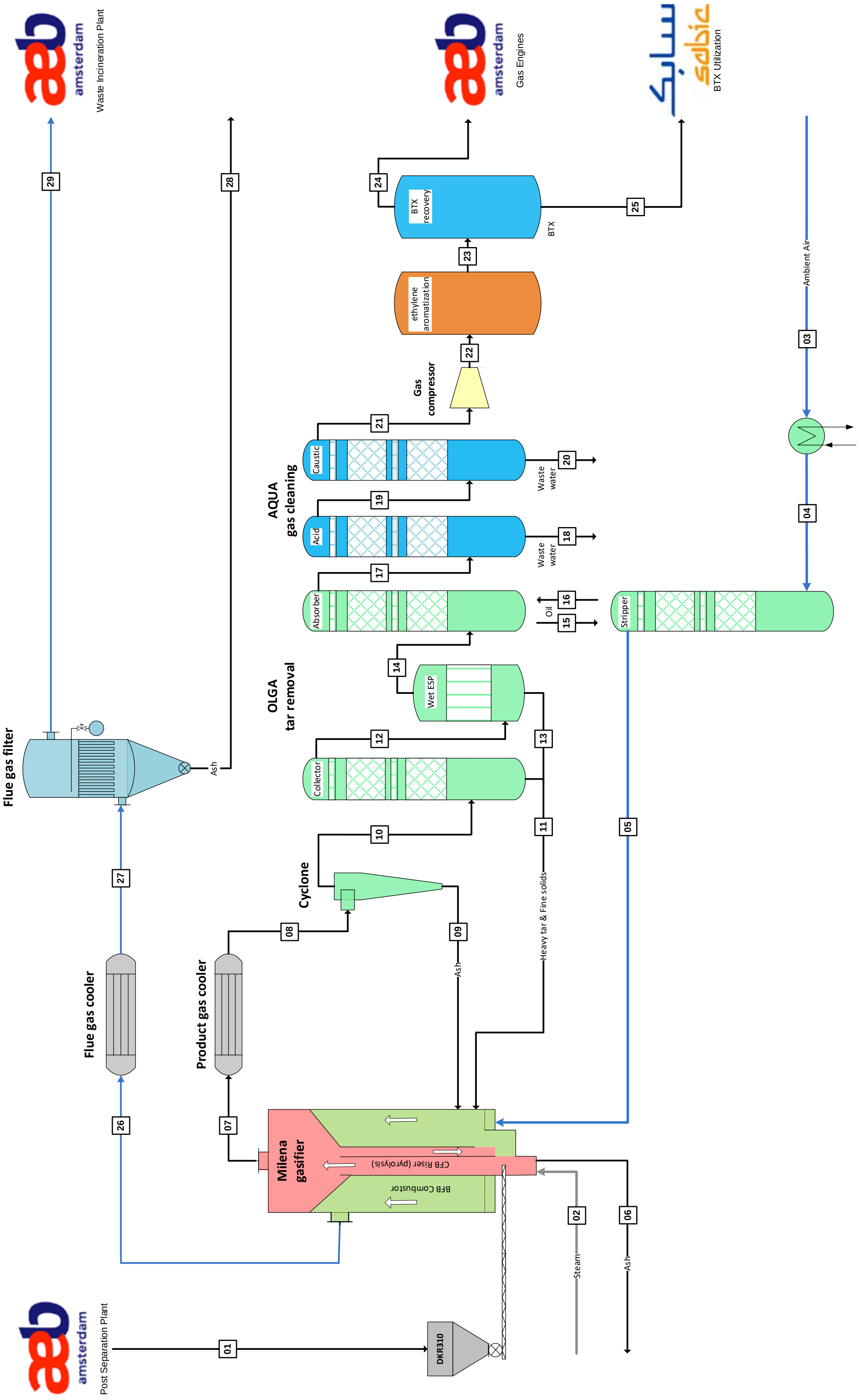


Figure 6.1: Process flow diagram of chemical recycling fitted into existing waste-to-energy plant

6.2 Refinery Setting

When integrating chemical recycling directly into a petrochemical setting, (plastic) waste can be imported from different locations. Within the process flow diagram shown in Figure 6.2, the traditional linear model of the refinery is shown in the top.

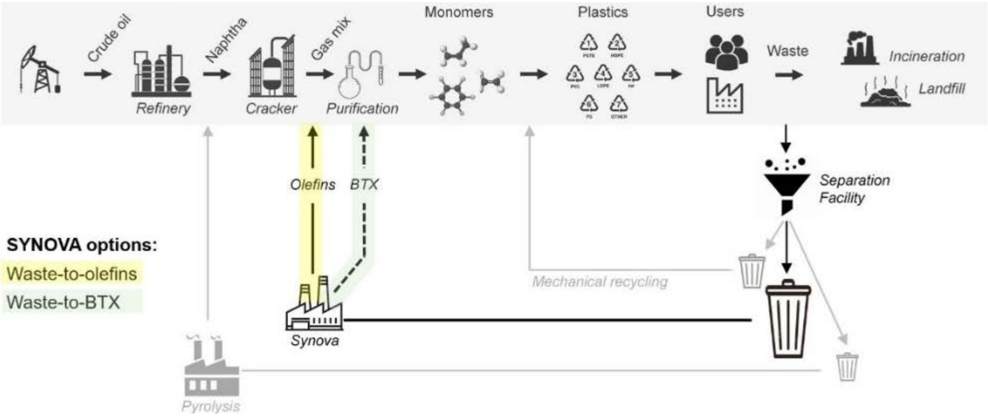


Figure 6.2: Process schemes of chemical recycling fitted into refinery setting

This figure shows three ways of recycling plastics, the first being that part of the waste can be separated mechanically to recycle plastics directly as plastics. The recycled material is mixed with virgin material to provide products with a certain recycled content.

The second is pyrolysis, a process where plastic waste is converted into a liquid, which (after hydrotreatment) can be blended with the naphtha to form the feedstock of the cracker. This is a form of chemical recycling, where the recycling takes place via the original building blocks like ethylene and propylene.

The third is cracking (gasification) of waste plastics in a way similar to what a steam cracker does with naphtha. The resulting gas can (after purification) be added to the gas produced in a naphtha cracker. Also, this is a way of chemical recycling. Alternatively, the unit converts the gases into liquid BTX.

The chemical recycling by means of gasification integrates with the refinery setting downstream the cracker either via the production of liquid BTX being blend into the existing pygas stream or via the production of an olefin rich gas being blend into the existing recovery of ethylene and other high value chemicals. A process flow diagram of a M30 is provided in Figure 6.3.

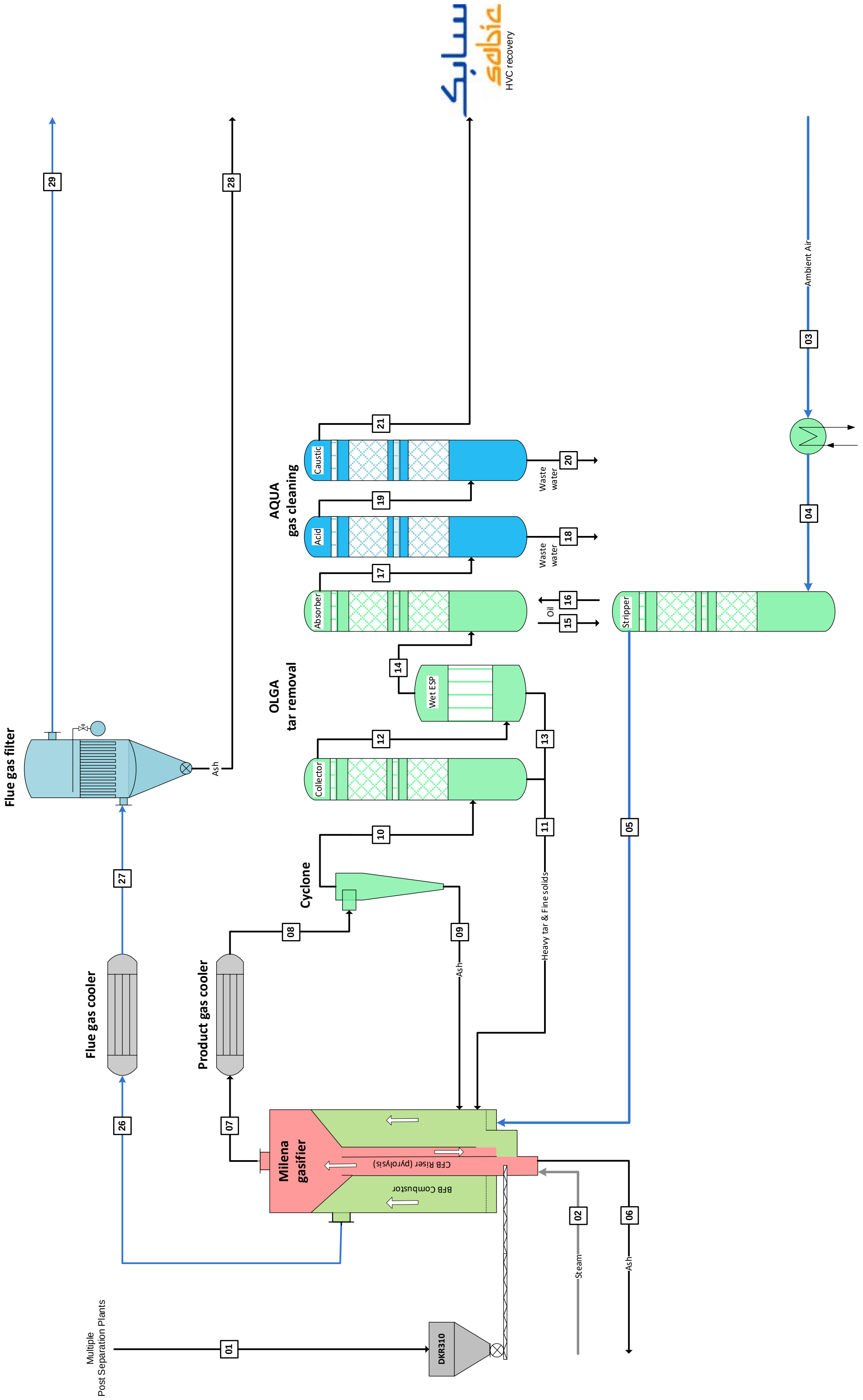


Figure 6.3: Process flow diagram of chemical recycling fitted into refinery setting

6.3 Economic analysis

With regards to the technical and economic viability, two concepts have been evaluated: (1) 1.1 tonne/h plastic waste conversion to BTX liquid and electricity with existing gas engines at AEB, and (2) 6 tonne/h plastic waste conversion to a olefin-rich gas ready to be injected into cold separation units downstream existing naphtha cracker plants of Sabic. The small-scale BTX plant produces almost 300 kg/h BTX liquid and just over 1 MWe net electricity. The larger scale olefins plant produces approx. 1.6 tonne/hour olefins and 370 kg/h BTX as part of a gas mixture, which is similar to the gas mixture produced by a naphtha steam cracker. Both options produce chemicals that are used to obtain recycled content in new plastics. Both options therefore can be called Plastic-to-Plastic concepts.

The value of the BTX and the olefins is calculated based on a 60 $/barrel crude oil assumption (range: 40-80 $/barrel) and a 15% premium is added for circularity. Furthermore, CO_2 prices are assumed 50 $/tonne CO2 (range 20-150 $/tonne CO2). Feedstock price is assumed to be zero (range -100 to +50 $/tonne). From a simple economic analysis, it follows that the small-scale BTX plant can only be economically viable when all parameters are chosen in the extreme of the ranges considered. The BTX plant hence preferably should be larger to benefit from economy-of-scale. For the Waste-to-Olefins case, the base case already shows (very) attractive economics. This is the result of the larger scale, higher yield of valuable products, and the use of existing downstream processing to produce polymer-grade monomers.

7 Conclusions

The experimental study using LDPE in the TMR shows that the pyrolysis temperature has a major effect on the BTX yields and a higher yield is obtained when using a high pyrolysis temperature. The experiments using DKR310 Small foils and H-ZSM-5 as the catalyst show limited signs of catalyst deactivation after 10 plastic injections. The experimental studies for the catalytic pyrolysis of LDPE, show that of the two selected catalysts in this study (H-ZSM-5 and a standard FCC catalyst), the H-ZSM-5 catalyst performs better than the FCC. Pyrolysis experiments with LDPE were performed in a gram-scale unit in batch mode to determine the mass balance closures. Mass balance closures were very satisfactorily and above 90%. Only minor amounts of (carbonaceous) residues were obtained, in line with the TGA data for LDPE.

Three plastic waste streams (DKR310 Big foils, DKR310 Small foils, Mixed plastics) were successfully converted into a gas containing valuable products in the Milena gasifier. Ethylene followed by methane are the main components produced regardless of the feedstock used. Also mass yields of at least 6% are obtained for benzene. A conversion of 66-71% was achieved into product gas on a carbon basis. The composition of the gas generated is very similar among the three feedstocks with the exception of ethylene and oxygen compounds such as CO and CO_2. The DKR310 fractions generate more C_2H_4 and less CO and CO_2 as compared with the Mixed Plastics fraction. Among the three feedstocks, the DKR310 Big foils lead to better results with corresponding highest impact factors (Climate advantage, Resource score and Feedstock value indicator of 1.91, 0.72 and 1.15 respectively).

When utilizing plastic waste like DKR310 as feedstock for chemical recycling, no significant changes are required to the design of the MILENA reactor itself. Only a combined start-up air / steam connection would be needed with a steam trap on that connection. The higher thermal load of the MILENA does require some additional cooling capacity on the flue gas and product gas coolers as well as the oil cooling capacity of the OLGA tar removal technology.

With regards to the technical and economic viability, two concepts have been evaluated: (1) 1.1 tonne/h plastic waste conversion to BTX liquid and electricity with existing gas engines at AEB, and (2) 6 tonne/h plastic waste conversion to a olefin-rich gas ready to be injected into cold separation units downstream existing naphtha cracker plants of Sabic. From a simple economic analysis, it follows that the small-scale BTX plant can only be economically viable when all parameters are chosen in the extreme of the ranges considered. The BTX plant hence preferably should be larger to benefit from economy-of-scale. For the Waste-to-Olefins case, the base case already shows (very) attractive economics. This is the result of the larger scale, higher yield of valuable products, and the use of existing downstream processing to produce polymer-grade monomers.

Finally, this project aimed at finding the answer to the question “Can pyrolysis/ gasification provide a good recycling alternative for materials that cannot be mechanically recycled, offering a final product efficiency of at least 70% on a hydrocarbon mass basis? The answer is yes!

8 Dissemination

The activities developed in this project were performed under confidential agreements since they required sensitive IP from the different partners. As such only a specific contribution was given to the workshop “Waste Gasification” organized by IEA Bioenergy Task 33

C.F. Mourao Vilela; “Gasification of end-of-life plastics”; IEA Bioenergy Task 33 Meeting, Workshop “Waste Gasification”26th November 2019, Birmingham (UK)
http://www.ieatask33.org/content/home/minutes_and_presentations/2019_Nov_WS/