Recycling of polystyrene-based external thermal insulation composite systems – Application of combined mechanical and chemical recycling

Niklas Netsch [a,*], Martin Simons [b,*], Alexander Feil [b], Hans Leibold [a], Frank Richter [a], Julia Slama [a], Savina Padumane Yogish [a], Kathrin Greiff [b], Dieter Stapf [a]

[a] *Karlsruhe Institute of Technology (KIT), Institute for Technical Chemistry, Hermann-von-Helmholtz-Platz 1, Eggenstein-Leopoldshafen 76344, Germany*
[b] *RWTH Aachen University, Department of Anthropogenic Material Cycles, Wuellnerstrasse 2, Aachen 52062, Germany*

A B S T R A C T

The material recycling of complex waste streams such as external thermal insulation composite systems (ETICS) is challenging, which is why their recycling in the sense of a circular economy is currently hardly established. Therefore, the combined mechanical and thermochemical recycling of ETICS based on expanded polystyrene (EPS) is investigated experimentally and by simulating full process chains in order to evaluate circular economy opportunities. Model ETICS as example for building and construction waste is pretreated mechanically, followed by either pyrolysis and / or gasification steps, and full mass and energy balances are derived. By the combined recycling, inorganic compounds can be separated to a large extent allowing a pre-concentrate generation. The plastic-rich pre-concentrate is converted into either pyrolysis oil with a high styrene monomer content of 51 wt% or to synthesis gas in the subsequent thermochemical conversions. The holistic approach enables a high carbon recycling rate between 53 and 68 wt%. In addition, the investigation reveals technology limitations and opportunities to be further developed and optimized.

1. Introduction

Plastics production in the European Union (EU) reached an amount of approximately 57.9 million tons in 2019 (PlasticsEurope 2020). To address both the reduction of green house gas emissions in combination with the issue of plastics in the environment, the resource efficiency has to be increased by avoiding landfill or incineration of plastic waste while implementing a circular economy for plastics. (Geyer et al. 2017).

Following the usage of plastics as packaging material, the application in buildings and construction is the most important area with a share of 20.4 wt% (PlasticsEurope 2020). Waste management statistics for Germany show that approx. 46 wt% of the plastic waste generated was recycled in 2019. The remaining 54 wt% were used for energy recovery (Lindner et al. 2020), which does not qualify as material recycling under the definition of a circular economy.

The accumulating plastics from demolition waste mainly consist of polyvinyl chloride, polyolefins, and polystyrene-based (PS) products

(Lindner et al. 2020). For specific polymers, e.g. PVC-containing construction products, functioning recycling systems are already established in Germany (Rewindo GmbH 2020), while other plastics including polystyrene in ETICS are rarely recycled.

In Europe, approx. 15 million tons of polystyrene were consumed in 2019 (PlasticsEurope 2020). The production share of foamed PS is approx. 43 wt%. (Lindner et al. 2020). This foam is mainly generated in form of EPS and installed as part of ETICS for facade insulation (Albrecht and Schwitalla 2015).

The high content of complex composite materials in combination with high quality requirements for secondary raw materials are the two major challenges preventing mechanical recycling from achieving a higher recycling percentage (Dogu et al., 2021). An ETICS is a multi-component composite that combines various materials, such as plastic, reinforcing fabric, mineral and plaster as shown in Fig. 1. The insulation material is applied to the building envelope, which usually consists of masonry or concrete, with the aid of adhesive mortar. This can

additionally be mechanically fastened with dowels or rail systems. A reinforcing mortar including reinforcing mesh is applied to the outside of the insulation material to absorb tensile and compressive stresses. Commonly consisting of a plaster layer or stone slab, the final layer of the ETICS serves to protect the inner layers and provides an aesthetic exterior effect (Heller and Flamme 2020). The combination of composites hinders the disassembly into individual components and thus recovery. Additionally, products that are contaminated with flame retardants e.g. hexabromocyclododecane (HBCD) are in circulation (Rani et al. 2014). Although HBCD is no longer produced or used since 2015, as the REACH classification prohibits it (European Union 2006), due to its long lifetime, it wiII be found in future waste (Jang et al. 2017).

Current mechanical recycling processes have only been used for offcuts from new and HBCD-free installations, but potential technologies for the recycling of PS-based insulation materials have been developed recently, where mechanical processes alone are insufficient. For example, in the EU project PolyStyreneLoop, PS foam is recycled in a solvent-based process, including HBCD-containing PS foam, (PolyStyreneLoop B.V. 2020). However, the effectiveness of this process is only given if a maximum impurity content of 7 wt% is present in the feedstock (PolyStyreneLoop B.V. 2019). Therefore, energy recovery by waste incineration is the common disposal method for heterogeneous construction waste comprising a mixture of various plastics (Lindner et al. 2020).

Consequently, the application of chemical recycling is to be considered to enable the carbon efficient recycling of complex construction waste (Ragaert et al., 2017; Rahimi and García, 2017). More specifically, robust thermochemical processes like pyrolysis or gasification appear promising solutions for the recycling of the plastics content (Dogu et al. 2021) since the generated products, synthesis gas and pyrolysis oil, may be used as petrochemical feedstock replacing fossil feedstocks. Achilias et al. show that, due to the chemical structure of polystyrene polymers, thermal pyrolysis is a promising recycling route (Achilias et al. 2007). This is explained by the preferential cleavage at weak links in the polymer chain which is induced by the aromatic side chains (Faravelli et al. 2001). Thus, PS belongs to the small group of polymer types that depolymerize during thermal pyrolysis, causing a beneficially high yield of monomer, such as poly(methyl methacrylate) (Moens et al. 2020) or polyamide 6 (Bockhorn et al. 1999).

To process ETICS with its complex heterogenious composition a mechanical preatreatment is needed so organic material can be separated from inorganic composite elements before further recycling of the plastics. For this purpose, various pathways for combined mechanical and chemical recycling of EPS-based ETICS are developed within this study. Mass and energy balances are determined via experimental investigations and simulations. The derived database should enable the comparison of the combined recycling processes in order to evaluate the recycling efficiency of the proposed approaches and also to identify optimization opportunities and technological challenges.

2. Material and methods

2.1. Feedstock

The composition of an ETICS depends on many influencing factors and varies from building to building (Riedel et al., 2010).

For the investigations described in this work, the insulation material EPS was selected because it accounts for a market share of 80 wt% of ETICS applications in Germany, and has been used extensively since the 1970s. EPS is one of the standard thermoplastics and belongs to the group of particle foams (Albrecht and Schwitalla 2015). The service life of ETICS is estimated between 40 and 80 years, which is why increasing waste masses are to be expected in the future (Geyer et al. 2017).

In this work, test samples were prepared by ANTS. EPS as insulation material and a plaster coating were used for this purpose. The reinforcement used is a fiberglass mesh with a mesh size of 6 mm. The density of the ETIC EPS panels was measured to be approx. 14 kg/m^3. The components used were provided by Sto SE & CO KGaA. In this way, it was possible to ensure that the materials used were approved by current building regulations and suitable for joint use.

For this purpose, consecutive small-scale comminution and sorting tests with defined parameterization were carried out at the test facilities of ANTS to identify the optimum process parameters. To ensure comparable and reproducible results, test samples of identical composition and geometry (with L × W × H dimensions of 250 × 100 × 90 mm) were generated and used for all test series of the mechanical preparation. The average sample mass was approx. 220 g and consists of approx. 13 wt% EPS, approx. 85 wt% mortar, plaster and adhesive, and approx. 2 wt% reinforcement. While mineral components such as adhesive or plaster account for the major mass content of an ETICS, the insulation layer has

1	Building envelope	5	Mortar
2	Adhesive mortar	6	Reinforcing mesh
3	Insulation material	7	Outer ETICS shell (plaster layer / stone slab)
4	Dowel		
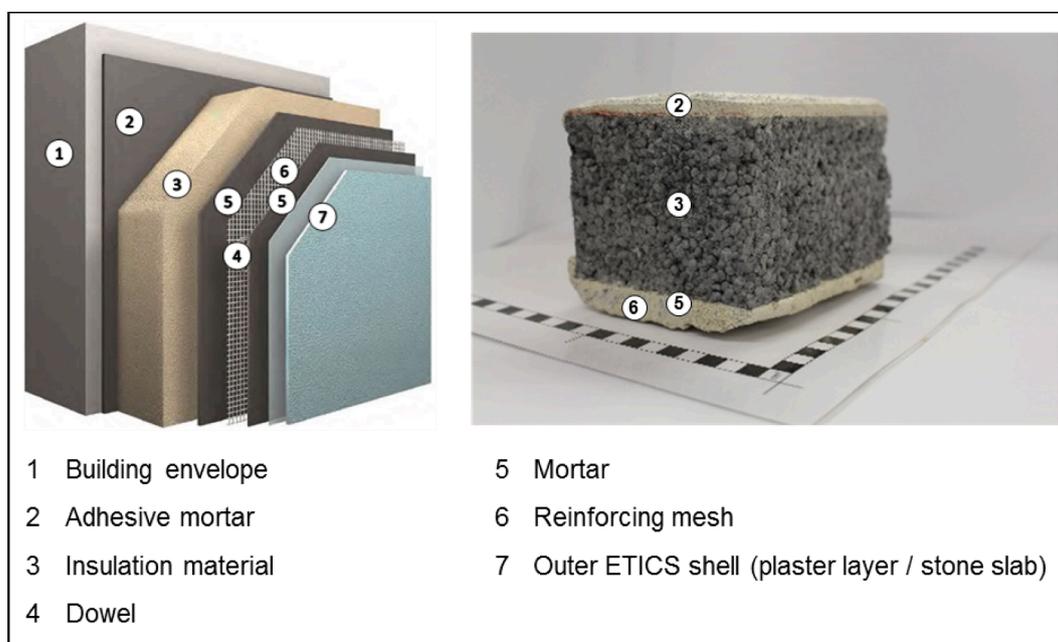
Fig. 1. Schematic structure of a state-of-the-art ETICS (left) and a sample of the material under investigation (right).

the largest volume.

2.2. Mechanical treatment

Mechanical processing aims to separate components of the composite ETICS and precondition them for subsequent treatment. The EPS is then fed into a thermochemical recycling process.

In the present case, the objective was to transfer a maximum EPS content in the pre-concentrate. In order to achieve maximum EPS content, it was necessary that the ETICS is released in the composite materials so that the successive separation of the crushed material, according to material properties, is possible.

Upon identifying the process chain parameter to gain a maximum EPS content in the pre-concentrate, an ETICS mass of approx. 40 kg was prepared and the generated pre-concentrate was provided for thermochemical conversion.

2.2.1. Methodology

A requirement for the separation of composite materials is the release of composite systems into material-specific individual fractions. Material systems that react differently to the type of stress resulting from comminution ("selective comminution") are advantageous in this respect allowing a maximum release of the composite material.

The objective of obtaining a maximum EPS content in the pre-concentrate requires specific process settings. The results of the different settings were balanced and evaluated. Both the quality achieved (content c_i), the mass yield Y_M and the recyclable material yield Y_R are included in the balance. The content c_i according to equation (1) is defined as the ratio of the recyclable mass to the total mass. The recyclable output according to equation (2) is the proportion of the recyclable material in the feed ($m_{R,Input}$) that is transferred to a separation fraction ($m_{R,sep,i}$). The mass yield according to equation (3) represents the ratio of the mass in the input, which is discharged into a separation ($m_{M,sep,i}$).

$$c_i \quad \frac{m_{R,i}}{m_{M,i}} \qquad (1)$$

$$Y_R \quad \frac{m_{R,sep;i}}{m_{R,Input}} \quad \frac{c_{i,sep}*m_{sep}}{c_{i,input}*m_{input}} \qquad (2)$$

$$Y_M \quad \frac{m_{M,sep,i}}{m_{M,Input}} \qquad (3)$$

For the investigations, a hammer mill was selected (rotor: Ø 470 mm, length: 540 mm; 30 kW drive power; self-made by ANTS). Comminution in hammer mills is primarily carried out by impact stress, which favors the comminution of brittle, i.e. mineral, raw materials. Different comminution intensities, in terms of peripheral speed (13, 27, and 40 m/s) and the discharge grate with different circular opening width (20 and 40 mm), were applied. In the series of tests, each of the possible parameter combinations was repeated five times, thus a total of 30 comminution tests were carried out with the ETICS. The test procedure in the hammer mill was discontinuous. Upon completion of the comminution of each sample, the comminution chamber was opened and the residues remaining in the discharge grate were removed, verified by material type, and weigthed.

The crushed material was screened following DIN EN 66165:2016-08. The material composition of the individual screen fractions was determined up to a particle size of 2 mm by manual sorting. The crushed material was sorted in four fractions (EPS, reinforcement, mortar-plaster-adhesive and composite). The screen fraction with a size < 2 mm was determined by loss on ignition in accordance with DIN EN 15169:2007-05.

In order to gain a maximum of EPS content, a specific enrichment is necessary, which was realized by air classification. Separation by air classification is performed according to the material properties density, particle size and particle shape.

Given the large density difference between the components in the present mixture (EPS 15 kg/m^3, mortar, plaster and adhesive and reinforcement 1400 to 2600 kg/m^3) (Sto SE & Co. KGaA 2017), the separation by density of the comminuted products appears to be a promising option. For this purpose, a zigzag wind sifter type 125–516 provided by Graf Anlagenbau, was used. The process-technical control variable for the separation efficiency is the air velocity. The light fraction is guided upward with the incoming airflow, while heavier or larger particles are discharged downward by gravity. In order to classify only by density, it is necessary to categorize the input material by particle size, preferably with a particle size ratio d_U/d_L (ratio of upper to lower particle sizes of 3/1) (Pretz et al. 2017). Classification according to particle size can avoid possible uniformity classes and improve the result.

2.3. Thermochemical conversion

2.3.1. Identification of thermochemical conversion processes

Thermochemical conversion is defined as a chemical reaction that takes place in the presence of high temperatures. Applied to plastic waste, pyrolysis means thermal degradation in an inert atmosphere resulting in the products pyrolysis gas, pyrolysis liquids, and a solid pyrolysis residue. Gasification means partial oxidation of the waste resulting in synthesis gas consisting of carbon monoxide and hydrogen as the main products. The process selection is based on various criteria considering the feedstock, reactor system, auxiliaries and products. A detailed comparison of various pyrolysis and gasification reactors for thermochemical conversion of plastics and plastic wastes can be found in Chen et al. and Ciuffi et al. (Chen et al. 2014; Ciuffi et al. 2020). In this study, fluidized bed gasification, entrained-flow gasification, and intermediate pyrolysis (lower heating rate and pyrolysis temperature compared to fast pyrolysis) are selected as technologies due to their feedstock flexibility, scalability and potential technical availabilty (Arena 2012). Furthermore, the products of pyrolysis and both gasification processes have properties that enable the integration into existing petrochemical value chains, if feedstock impurities like HBCD or mineral content can be separated during thermochemical processing. The characteristic process requirements are listed as shown in Table 1 (Stapf et al. 2019).

2.3.2. Pre-concentrate characterization

Preliminary tests were conducted to ensure that the feedstock requirements for processing in the selected reactor systems are met. The ash melting temperature was determined by heating microscopy in a Hesse Instruments microscope (EM301). The decomposition behavior of the insulation material was investigated in an inert atmosphere and a heating rate of 10 K/min via thermogravimetric analysis, using a Netzsch TG209 thermo-microbalance. Breakdown of the porous structure of the EPS feedstock is necessary to increase the density and remove possible gaseous inclusions. For this purpose, the compacting was investigated through thermal treatment in a convection oven at 120 °C for 6 h and by mechanical treatment with a hydraulic press of a maximum compressive load of 150 bar.

2.3.3. Intermediate pyrolysis experiments

Pyrolysis is a process that is controlled by chemical kinetics, heat and mass transfer. Product yields strongly depend on both, the feedstock processed as well as the process technology. The mass and energy balance for the pyrolytic conversion of the EPS pre-concentrate is determined with an electrical heated auger-type reactor that has been described elsewhere (Tomasi Morgano et al. 2015; Zeller et al. 2021). The pilot plant provides intermediate pyrolysis for disperse organic feedstock materials in a sand-supported process. Inert quartz sand is used as a moderator to enable the transport of the molten polymer, as well as to improve heat transfer. An integrated filter system allows the generation of particle-free pyrolysis vapors. The vapors are then

separated into permanent gas and liquid condensates in three condensation stages, twice at 60 °C and once at 5 °C, before they are fed into electrostatic precipitators. The outlet of the reactor is connected to a flare, where the permanent gases are burned after passing through the online gas analyses, which include density, volume flow and calorific value measurements. The experiments were conducted at a reactor temperature of 450 °C, a solid residence time of 30 min, and at ambient pressure. The mass flow rate of the reactive feedstock was 1 kg/h. The quartz sand was added at a 4/1 sand-feedstock mass ratio which is equivalent to a volume ratio of approximately 1/1. For data collection, the EPS-based pre-concentrate was pyrolized after thermal pre-compacting.

During steady-state operation, two permanent gas samples of approximately 10 l volume were withdrawn from the gas stream for a GC-FID and GC-TCD analysis in an *HP 5890 II* gas chromatograph. Light hydrocarbons as well as CO, CO_2, N_2, and O_2 were detected. The condensate was tested with an *Agilent GC-FID 7890B* to determine the proportion of characteristic hydrocarbon species, i.e. aromatics, alkanes, and olefins. Further upgrading of the condensates as well as removal of heteroatom containing species (here bromine) is considered as post-processing step. The mass balances were calculated gravimetrically by weighting the solid and liquid products. Solid pyrolysis products were distinguished from sand by subtracting the mass of the dosed sand. Considering the nitrogen flushing gas, the mass of permanent gases was determined from the continuously monitored gas flow and gas density. The elemental analysis of solids and condensate enabled the evaluation of the carbon content. The char content in the solids was verified via combustion. The reactor energy demand was permanently monitored, which enables the calculation of the energy needed for the endothermic pyrolysis process. This was done by comparing the pyrolysis experiment with a reference test run where only sand was fed into the reactor. This energy demand includes the heating and phase transition of the feedstock, as well as the energy required for the endothermic pyrolysis including the evaporation of emerging products.

2.3.4. Gasification simulation

Gasification is considered as being thermodynamically controlled. The mass and energy balances of the two gasification processes are determined by simulations using the flowsheet simulation tool EBSILON Professional (Version 15) by STEAG Energy Services GmbH. The simulations are based on the ultimate analysis of the pre-concentrate. For calculating the equilibrium composition of the raw synthesis gas, the gasifier outlet temperature had to be specified. Steam and oxygen were used as gasification agents. The ratio of oxygen supplied by gasification agents to oxygen that is required for stoichiometric combustion was set to 0.4 in both processes.

Fluidized bed gasification was calculated as thermodynamic equilibrium at 950 °C and ambient pressure. The formation of tar and coke in the fluidized bed reactor, which could not be represented via equilibrium simulations, was derived from literature data (Han and Kim 2008; Sancho et al. 2008; Arena et al. 2009). Additional tar formation of 10 g/nm^3 related to the raw synthesis gas was implemented to reflect realistic gasification scenarios. The coke formation was based on a carbon gasification rate of 95 wt%. The carbon gasification rate is defined as the amount of gasified carbon related to the carbon introduced into the conversion by the feedstock.

Entrained-flow gasification operates at higher temperature and higher pressure than fluidized bed gasification. This is achieved by adding more oxygen and less steam to the feedstock. The synthesis gas quality is higher than for fluidized bed gasification, but the product yield is lower and more CO_2 and less hydrogen are produced. The gasifier's operating temperature and pressure were set to 1200 °C and 40 bars, respectively. Tar formation was neglected due to the high gasification temperature. The carbon gasification rate of this gasifier type was set at 98 wt%.

In order to make both gasification processes comparable, the process simulation includes a water–gas-shift reactor to adjust the H_2/CO ratio in the raw synthesis gas at the gasifier outlet. In both gasification variants, the synthesis gas composition was calculated for different H_2/CO ratios.

2.3.5. Process chains investigated

Three different combined recycling pathways shown in Fig. 2 are investigated. All are based on a recycling concept containing a decentralized mechanical generation and compacting of pre-concentrate combined with a centralized thermochemical conversion process. Processes were regarded at an industrial scale described by a thermal capacity of 100 MW of the first central thermochemical process step.

The process chains consist of mechanical treatment, followed by thermochemical conversion and subsequent product purification, which is considered to deliver product qualities as required by usage as chemical feedstock. Either synthesis gas or organic pyrolysis condensate represent the final products of the recycling concepts. Fluidized bed gasification can be fed directly by the solid pre-concentrate from mechanical pretreatment. In the case of the entrained flow gasification, the pyrolysis is implemented as a preparatory step to enable the processing of the feedstock, given that the solid pre-concentrate cannot be introduced to this gasifier type in the provided form. In contrary to the direct use of pyrolysis products as petrochemical feedstock e.g. for steam cracking, in this case no pyrolysis product purification is needed ahead of the gasification step. For both gasification alternatives, H_2/CO ratios of 2.1/1 and 1/1 are examined. Due to the varying complexity of the synthesis gas treatment, the modules of the gas conditioning are adjusted as per type of the gasification process. Mineral gasification by-products are disposed. Incineration of pyrolysis residues incuding flue gas cleaning is considered to convert them into disposable mineral by-products and to cover the energy demand of the endothermic conversion.

An important parameter of the material flow analysis is the mass fraction of the flows related to the mass of ETICS. Likewise, an integral estimation of the energy demand is used to compare the recycling routes. For this purpose, possible heat extraction and electricity consumption were derived. Furthermore, a carbon recycling rate was determined for each material flow. This parameter indicates the proportion of carbon contained in the organic condensate and the synthesis gas compared to the amount of carbon introduced into the recycling process by the ETICS.

3. Results

3.1. Mass balance and product yields

3.1.1. Mechanical treatment

The analysis of the crushed material and the selection of the presented particle class, which represents the input material for the downstream air classification, were performed based on the following requirements. The EPS content of the crushed fractions is a subordinate fact for the selection of a suitable particle class, since a targeted enrichment of the valuable material from the crushed material takes place by means of downstream windsifting. The selection of the particle class is rather made according to the achieved recyclable material yield as the decisive criterion. Furthermore, the upper particle size of the pre-concentrate is set at 20 mm. The particle size of individual EPS beads is in the particle size range of 5 to 8 mm, which means that an accumulation of EPS in this particle size range could be assumed. As described in the "Methods" section, screen classification can be used to increase the quality of the sifting result by classifying it beforehand and maintaining the particle size ratio of 3/1. Fig. 3 shows the Y_R for EPS, the Y_M and the percentage of EPS share per fraction for the particle classes <4 mm, 4–11.2, 11.2–20 and >20 from the comminution trials. Looking at the Y_M, it can be seen that the Y_M increases with the decrease in particle size. The largest mass accumulates in the "fine" fraction < 4 mm (median: 60
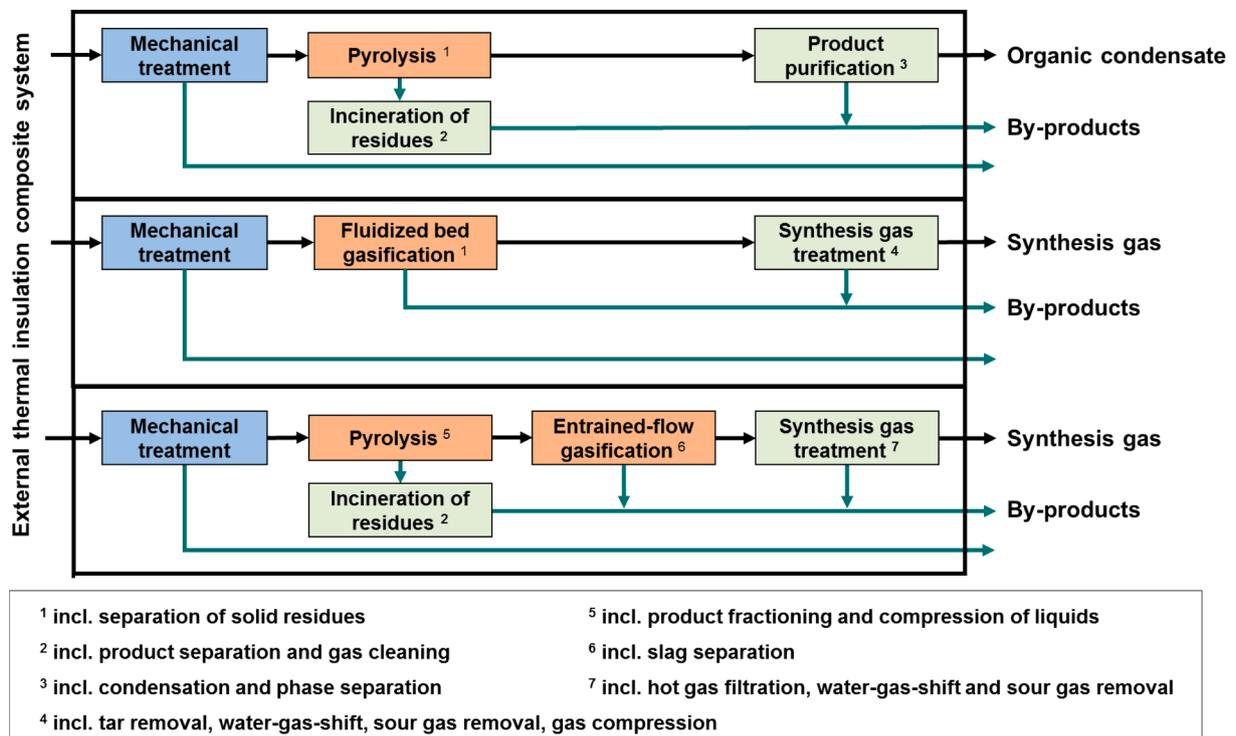
Fig. 2. Design of the investigated process chains.
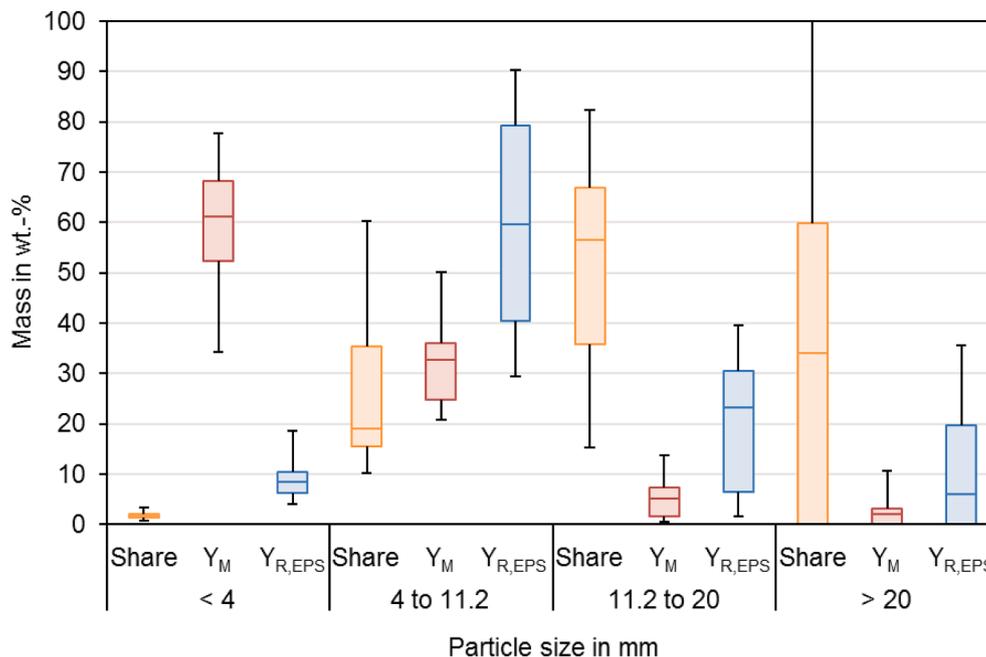
Fig. 3. Comparison of YR of EPS, YM and EPS share per fraction.

wt%). Due to the special comminution effect of the hammer mill on brittle materials, the predominant mass content of this fraction consists of released mineral components, such as plaster, adhesives, and mortar. The proportion of EPS in the particle size class < 4 mm is in median 2 wt %, which corresponds to a Y_R of 6 wt% of the fed EPS mass.

As suspected, the highest Y_R was achieved in the grain size class 4 to 11.2 mm. On median, 59 wt% of the fed EPS mass was discharged into the particle class 4 to 11.2 mm. At the same time, a low Y_M of 33 wt% was achieved compared to the < 4 mm particle class. Despite the high Y_R and the lower Y_M, the median EPS content in this particle class is 19 wt%

which can be attributed to the low density of EPS compared to the other components.

Compared to the particle classes 4 to 11.2 mm and < 4 mm, the proportion of EPS in the particle class 11.2 to 20 mm is significantly higher, with a value of 57 wt%. This can be attributed to the low mass yield of 5 wt% and thus to a low proportion of mineral components. With a median yield of 17 wt%, a comparatively large EPS mass was discharged into this particle size class.

The EPS mass discharged into the particle size class > 20 mm is to be regarded as a loss due to the particle size restriction of the downstream

thermal conversion and is irrelevant for further consideration.

The air classification allows a targeted enrichment of the EPS material, whereby a high EPS content (median value: approx. 95 wt%) and high EPS yield (median value: approx. 99 wt%) were achieved in the pre-concentrate related to wind sifting. The stream classification was carried out at a comparatively low air velocity of around 2 – 3 m/s. Considering the EPS yield for each processing step and calculating the EPS yield for the entire processing chain, a high EPS yield is achieved in the pre-concentrate. (median value: approx. 81 wt%).

The empirically identified parameters served as a basis for the preparation of approx. 40 kg of EPS based ETICS. The preparation of these 40 kg was performed analogously to the previously described experiments. The crushed material was separated from coarse and fine material by sieving and the fraction 4 to 11.2 mm was sifted by air classifier. The generated pre-concentrate was provided for the thermochemical conversion tests. The processing steps were balanced and the balancing of the EPS yield for the entire processing chain showed that compared to the tests with individual test samples, a EPS yield of approx. 86 wt% was achieved with an EPS content of approx. 98 wt% in the pre-concentrate. Thus, the observed values are slightly above the median values of the individual tests. The increase in the recyclable material yield can be credited to less remaining residue in the comminution chamber during continuous comminution of 40 kg of EPS, in contrast to the discontinuous comminution of single test samples in the parameter identification process.

3.1.2. Pre-concentrate characterization

The ash melting point of the mineral component mixture is approx. 1100 °C. Therefore, the ash melting point meets the specified requirements of the gasification technologies described in Table 1. The gravimetric analysis demonstrates that the pyrolysis of EPS follows a one-step thermal decomposition mechanism at about 400 °C, which is consistent with the literature on the degradation of foamed PS (Kannan et al. 2009; Jiao et al. 2012; Mehta et al. 1995). Approximately 85 wt% of volatile products are formed. Concerning the mineral mixture, only minor amounts of volatile components can be determined based on the thermogravimetric analysis, which can presumably be explained by the evaporation of crystal water or binders, as well as calcination processes. Consequently, chemical recycling of EPS by pyrolysis and gasification is possible due to the fulfillment of feedstock specifications.

The plastic-enriched pre-concentrate obtained in the mechanical treatment reveals an EPS content of approximately 98 wt%. From elementary analysis a content of carbon (90,5 wt%), hydrogen (7,8 wt%), bromine (0,7 wt%) and sulfur (0,01 wt%) can be derived leading to a calorific value of approximately 40 MJ/kg. An EPS volume shrinkage can be identified in the range of 80 to 140 °C by heating microscopy. Thermal compacting irreversibly increases the density to approx. 140 g/cm^3, while leading to the shrinking of polymer particles to 1 – 2 mm. Pressing the porous polymer structure does not produce permanently dimensionally stable samples in the laboratory-scale tests due to technical limitations. However, mechanical compacting is proved to be an efficient state-of-the-art option for increasing the density at low operating costs (Seo and Hwang 2006).

3.1.3. Thermochemical conversion

Table 2 shows the mass balance that is determined via the pyrolysis experiments and gasification simulations of the thermally compacted pre-concentrate. The main products of the respective product fractions are also indicated. The elemental analysis of the organic condensates shows almost exclusively hydrocarbons, with carbon and hydrogen contents of 91.6 wt% and 7.5 wt%, respectively. GC-FID analysis confirms a high aromatic content. The proportion of styrene in the organic condensate is 51.2 wt%, so 34 wt% of the EPS in the pre-concentrate can be recovered as styrene monomer. This is in line with the literature (Artetxe et al. 2015; Faravelli et al. 2001). The coke and reinforcing fabric-containing solid residue can be separated from the sand particles by sieving with minor technical effort. While the sand can then be reused, the residue must undergo further treatment to be converted into disposabIe products, as its carbon content is about 87 wt%. Further

Table 1
Comparison of the characteristic properties and requirements of the evaluated thermochemical conversion processes (Stapf et al. 2019).

Parameter	Intermediate pyrolysis	Fluidized bed gasification	Entrained-flow gasification
Feedstock requirements:			
Physical condition	solid	solid	liquid, gas, dust
Maximum particle size in mm	< 100	< 50	< 0.1
Ash melting point in °C	-	> 950	< 1.100
Reactor characteristics:			
Temperature in °C	450-550	900-950	1150-1250
Pressure in bar	ambient	ambient	40-60
Operating supplies	heat transfer medium	bed material, gasification agents (steam, oxygen or air)	gasification agents (steam, oxygen)
Products	permanent gas, condensate, solid residue (coke, ash, metals, minerals)	raw synthesis gas, solid residues (coke and ash, metals, minerals)	raw synthesis gas, residues (slag)

Table 2
Mass balance and main products of the thermochemical conversion processes including product separation and conditioning.

	Product	Mass fraction in wt. %		Main products
Pyrolysis	Input:			
	Pre-concentrate	100		-
	Output:			
	Organic condensate	69.0		Aromatic and aliphatic hydrocarbons
	Aqueous condensate	0.1		H_2O
	Permanent gas	19.1		CO_2, CO, light hydrocarbons
	Solid residues	7.8		Coke, minerals, reinforcement fabric, ash
	Balance loss	4.0		-
Fluidized bed gasification	Input:	**2.1/ 1**	**1/1**	
(H_2/CO 2.1/ 1 and 1/1)	Pre-concentrate	32.9	43.4	EPS, mineral content, reinforce-ment fabric
	Steam	42.3	23.8	H_2O
	Oxygen	24.8	32.8	O_2
	Output:			
	Synthesis gas	44.4	77.8	H_2, CO
	Solid residue	1.8	2.4	Coke, minerals, reinforcement fabric, ash
	Tar	1.3	1.3	-
	Wastewater	11.0	0.5	H_2O, solved sour gas
	Off-gas	41.5	18.0	CO_2
Entrained-flow gasification	Input:	**2.1/ 1**	**1/1**	
(H_2/CO 2.1/ 1 and 1/1)	Pyrolysis condensate	33.5	33.5	Hydrocarbons
	Steam	9.2	9.2	H_2O
	Quench water	24.5	24.5	H_2O
	Oxygen	32.8	32.8	O_2
	Output:			
	Synthesis gas	44.4	63.6	H_2, CO
	Solid Residue	0.6	0.6	Coke, minerals, reinforcement fabric, slag
	Wastewater	4.9	18.2	H_2O, solved sour gas,
	Off-gas	50.1	17.6	CO_2

treatment also applies to the low-calorific permanent gas which consists mainly of CO_2 and CO. These components are presumably the result of only partial outgassing from the porous structure or mineral components. Nevertheless, the permanent gas also contains up to 10 wt% of light hydrocarbons such as methane or ethane. Hence, its conditioning is necessary prior to its discharge from the process. The carbon content of 5 wt% indicates the presence of organic compounds in the aqueous condensate phase.

For the two gasification variants, the mass balances with different H_2/CO ratios of the synthesis gas are shown in Table 2. In the case of fluidized bed gasification, the pre-concentrate can be used as feedstock in the same way as in the pyrolysis concept, whereas in the case of the entrained flow gasification, the condensate from the upstream pyrolysis is fed into the gasifier. For both gasification technologies, it can be seen that the yield of synthesis gas is higher for an H_2/CO ratio of 1/1, because only a low proportion of hydrogen is present in both the condensate and the pre-concentrate. If a molar ratio of 2.1/1 in the synthesis gas is to be achieved in the gas conditioning, more steam must be converted to hydrogen in the water–gas shift reactor, which results in an increased formation of CO_2 and a lower CO content. This also accounts for the higher off-gas amount after the gas cleaning at a molar ratio of 2.1/1. The mass of the gasification agents added can vary in fluidized bed gasification depending on the synthesis gas composition, since a different amount of steam is needed to shift the water–gas equilibrium. In entrained-flow gasification, quench water is introduced in addition to steam for rapid cooling of the products after the gasification reaction. The amount of water or steam added is therefore subject to process-related restrictions and is less limited by the shift in the water–gas equilibrium.

3.2. Balancing energy in scaled processes

3.2.1. Mechanical treatment

Based on the results, an operational scale processing plant was designed to estimate the energy consumption per ton. As previously described, the EPS mass flow accounts for a small mass fraction of approx. 13 wt% of the feed. The impurities such as mortar, plaster, adhesive or reinforcement account for the larger mass fraction of approximately 87 wt%. The majority of the impurity fraction is separated by mechanical processing. This is necessary to achieve the goal of a pre-concentrate with a high EPS content. The mass flow in the pre-concentrate is approx. 13 wt% of the feed and has an EPS content of approximately. 98 wt%.

Based on the waste operational scale processing plant of ETICS in Germany and the conducted tests, the energy demand was estimated. The waste volume of EPS-containing ETICS was estimated using data from Lindner et al. (Lindner et al. 2017), and Albrecht and Schwitalla (Albrecht and Schwitalla 2015). From the ETICS waste volume in Germany in 2016, waste EPS was estimated to be approximately 120.000 t. With 80 million inhabitants, this results in a per capita production of 1.5 kg/E*a. Due to the decentralized nature of ETICS waste, a decentralized processing structure was selected and an annual plant throughput of 15.000 t was assumed, which represents a catchment area of approx. 10 Mio. inhabitants. The annual quantity of 15.000 tons corresponds to a plant throughput of 9 t/h at 1.600 operating hours per year.

The design provides for pre-shredding by means of a counter comb shredder to narrow down the particle size distribution and to homogenise the material flow. Connected to the pre-shredding, comminution using hammer mills is applied in redundant circuitry as second step. The adjustment of the required particle class for stream classification is reached using tension shaft screens. After stream classification, press devices are provided to increase the bulk density of the pre-concentrate produced up to 350 kg/m^3 and to prepare it for the transport. The results of the mechanical treatment tests were used to estimate the energy demand of a mechanical processing plant for EPS–containing ETICS waste on an operational scale based on the designed volume and mass flows.

The aggregates and conveying equipment were designed accordingly. For the estimated mechanical treatment plant, a connected load of approx. 725 kW is required. Related to the mass of the pre-concentrate, this corresponds to a specific energy demand of approx. 745 kWh/t or 2682 MJ/t.

3.2.2. Thermochemical conversion

The electrical energy demand of the reactors is assumed to be 3 % of their thermal capacity. This estimation includes the reactor itself as well as electrical energy required for the associated periphery, e.g. downstream or upstream processes (condensers, pumps, etc.) or measurement and control devices. Additional energy-intensive processes (air separation for the production of oxygen, process steam generation for gasification etc.) are excluded. Hence, it is important to consider the endothermic pyrolysis process and the compression of synthesis gas from 1 to 40 bar ahead of raw synthesis gas purification of the fluidized

Table 3

Mass and energy flows of input streams, intermediates, by-products, and final products in the evaluated chemical recycling concepts.

in t/h (if not stated otherwise)	Pyrolysis	Fluidized bed gasification		Entrained-flow gasification	
		H2/CO 2.1/1	H2/CO 1/1	H2/CO 2.1/1	H2/CO 1/1
Input:					
- ETICS	92.0	92.0	92.0	92.0	92.0
- Air	17.0	-	-	17.0	17.0
- Oxygen	-	6.8	6.8	6.3	6.3
- Steam	-	11.6	4.9	6.5	6.5
Intermediates:					
- Pre-concentrate	9.0	9.0	9.0	9.0	9.0
- Pyrolysis vapors	8.2	–	–	8.2	8.2
- Pyrolysis solid residue	0.7	–	–	0.7	0.7
- Pyrolysis permanent gas	1.8	–	–	1.8	1.8
- Aqueous condensate	0.0	–	–	0.0	0.0
- Organic condensate				6.4	6.4
- Raw synthesis gas		19.8	19.8	14.4	14.4
Products:					
- Organic condensate	6.4	–	–	–	–
- Synthesis gas		12.1	16.1	8.5	12,2
By-Products:					
- Mineral-rich reject	83.0	83.0	83.0	83.0	83.0
- Incineration residue	0.1	–	–	0.1	0.1
- Incineration flue gas	19.4	–	–	19.4	19.4
- Gasification residue		0.5	0.5	–	–
- Gasification slag		–	–	0.1	0.1
- Gasification tar		0.4	0.3	–	–
- Gas treatment off-gas		11.3	3.7	9.6	3.4
- Gas treatment waste water		3.0	0.1	0.9	3.5
Energy Balance:					
Electrical energy demand	in MW$_{el}$	in MW$_{el}$	in MW$_{el}$	in MW$_{el}$	in MW$_{el}$
- Mechanical treatment (incl. compacting)	6.7	6.7	6.7	6.7	6.7
- Reactor and periphery	3.0	3.0	3.0	5.5	5.5
- Synthesis gas compression	–	2.1	3.6	–	–
Potential heat recovery					
- Medium pressure steam	–	–	–	5.8 t/h	5.8 t/h
- District heating	1.9 MW	–	–	1.9 MW	1.9 MW

bed gasifier. The electrical energy demand and potential heat recovery are listed in Table 3.

For a central large-scale pyrolysis process, the energy demand is estimated based on experimental data from the pyrolysis of extruded polystyrene (XPS) pre-concentrate which was generated in an identical mechanical treatment. This is comparable to the EPS concentrate given its very similar chemical and physical composition. The energy demand determined for the pyrolysis of the XPS material is approximately 1.7 MJ/kg feedstock. This corresponds to an energy demand of 4–5 % of the feedstock's calorific value. From literature data, a calorific value-related mean energy demand for the pyrolysis of 4 – 5.5 % for pyrolytic cracking is available by differential scanning calorimetry (Cafiero et al. 2015; Ippolito et al. 2019). However, the energy demand measured in the experimental tests includes physical and chemical endothermic processes like phase transition, heating the feedstock to the pyrolysis temperature, reaction enthalpy and evaporation of the products but neglects reactor heat losses. For the recycling concept, the endothermic pyrolysis needs to be balanced by the exothermic incineration of pyrolysis byproducts, considering a secondary fuel cofiring, if necessary.

For entrained flow gasification, the simulation of the gasifier can be accessed regarding potential heat recovery. The process heat can be recovered in the form of a superheated medium-pressure steam (here 16 bar, 250 °C), which is of high exergetic value.

Due to the tar-containing raw synthesis gas, heat recovery is technically more challenging in the case of fluidized bed gasification. Also, the process temperature is comparably lower than in the entrained-flow gasification system. The possibility of heat extraction is therefore neglected. The amount of electrical energy needed for the synthesis gas compression is calculated based on an isentropic two-stage compression.

3.3. Comparison of the scaled recycling concepts

Table 3 depicts the mass and energy balance for the three chemical recycling concepts. For all of them, a pre-concentrate of 9.0 t/h is introduced into the thermochemical conversion process. This results in 92.0 t/h of ETICS that are to be introduced into multiple mechanical recycling systems, requiring approximately 6.7 MW of overall electrical energy for mechanical processing. The main part of the feedstock leaves the system as a mineral-rich reject regarding a chemical recycling concept focusing on thermochemical recycling of the carbon-containing plastics. Fig. 4 shows the carbon recycling rates for the different concepts. In mechanical processing, about 12 wt% of the carbon contained in the ETICS is already discharged from the system with the mineral-rich rejects. In all concepts, the thermochemical process steps also convert some of the pre-concentrate into undesirable by-products such as coke containing solid residue, CO_2 containing flue- or off-gas, or tar, which reduce the yield of the target products synthesis gas and organic condensate. Nevertheless, in all process chains, at least 54 wt% of the carbon contained in the ETICS is converted into these target products.

The experimentally determined pyrolysis energy demand is approximately balanced by the amount of energy that can be generated during the combustion of the pyrolysis residues, enabling the autothermal operation without cofiring. Furthermore, the surplus of low-temperature heat can potentially be recovered for generating district heat (water 6 bar, 100 °C).

Particularly concerning the concept of combined pyrolysis and entrained-flow gasification, further possibilities for material flow optimization emerge. Minor shifts in the product distribution due to feedstock-related adjustments of reaction conditions appear promising in pyrolysis. Thus, a permanent gas with a higher hydrocarbon content could also be fed into the entrained-flow gasifier. This would result in a higher yield of synthesis gas and fewer by-products, while the autothermal operation may no longer be guaranteed. Compared to fluidized bed gasification, entrained-flow gasification is more efficient in terms of higher carbon conversion rates and negligible tar formation. However, lower synthesis gas yields and carbon recycling rates are achieved in the combined concept. The recycling of the plastic fraction into organic condensate or synthesis gas offers the possibility of producing a variety of products with virgin material quality in petrochemical processes from waste material that has been so far mostly thermally recycled. Concerning the variety of different products, synthesis gas and pyrolysis oil with a significant styrene amount can only be compared to limited extent in terms of their product value.

4. Study limitations

The results of this study are subject to certain limitations regarding the transferability of the results due to the chosen system design. The investigated feedstock is a model EPS-ETICS, which contains all essential components and mineral impurities, but rarely occurs as a mono-fraction in real waste streams. Rather, a mixture of various mineral and organic constituents with toxic impurities (e.g. HBCD) is to be expected, so that, for example, extruded polystyrene or polyurethane-based ETICS should also be considered as feedstock in a future studies.

Transferring the results to an operational scale requires further development of the underlying approach and the developed process steps, in particular on continuous-large scale operation and product refinement. The processing of the pyrolysis oil in accordance with the specifications for subsequent product synthesis must be demonstrated. The removal of impurities in product refinement should be experimentally tested and further developed with regard to impurities such as bromine. In addition to the separation of the styrene monomer content, a possible use for the remaining organic condensate should also be verified. Rodriguez et al. propose the processing of the remaining condensate via fluid catalytic cracking to substitute fossil feedstocks in refineries (Rodríguez et al., 2019). Since the application as steam cracker feedstock is excluded due to a high aromatic content (Kusenberg et al., 2022) this approach appears promising. Also, gasification data was based on an equilibrium simulation, the transferability has to be proven by means of large-scale gasification tests.

In addition, the gasification and pyrolysis parameters are based on common values. By further optimizing the process parameters, yield improvements to valuable products can be expected, as shown by the variation of the H2/CO ratio in the gasification simulation. At best, such an optimization should not be carried out for individual process steps. Besides the important interface of mechanical treatment and thermochemical conversion, transport logistics of feedstocks, intermediates and products or feedstock availability, as well as various options for covering the energy demand should be considered. Only the extension of the system boundary enables the derivation of meaningful techno-economic and -ecological parameters on the basis of which an overall technical optimization can be carried out.

5. Conclusions

The technical feasibility of the thermochemical conversion of polystyrene plastics originating from ETICS and the effectiveness of the pre-conditioning by mechanical processing adapted to the specific material were demonstrated in the present paper.

The objective of mechanical pre-conditioning, i.e. to produce a pre-concentrate with a maximum content of EPS, was fulfilled in a process combination of comminution, screening, windsifting and compacting. The machine parameters identified by laboratory tests resulted in an EPS yield in the pre-concentrate of approx. 86 wt% with an EPS content of approx. 98 wt%. Since most of the ETICS mass is mineral material (approx. 86 wt%), processing and management of this material stream must be further optimized.

The process chain analysis reveals that a carbon recycling rate between 53 and 68 wt% is achieved. In this study of polystyrene based plastics, the concepts with only one thermochemical conversion step, i.e. stand-alone pyrolysis for direct use of the pyrolysis oil as liquid petro-
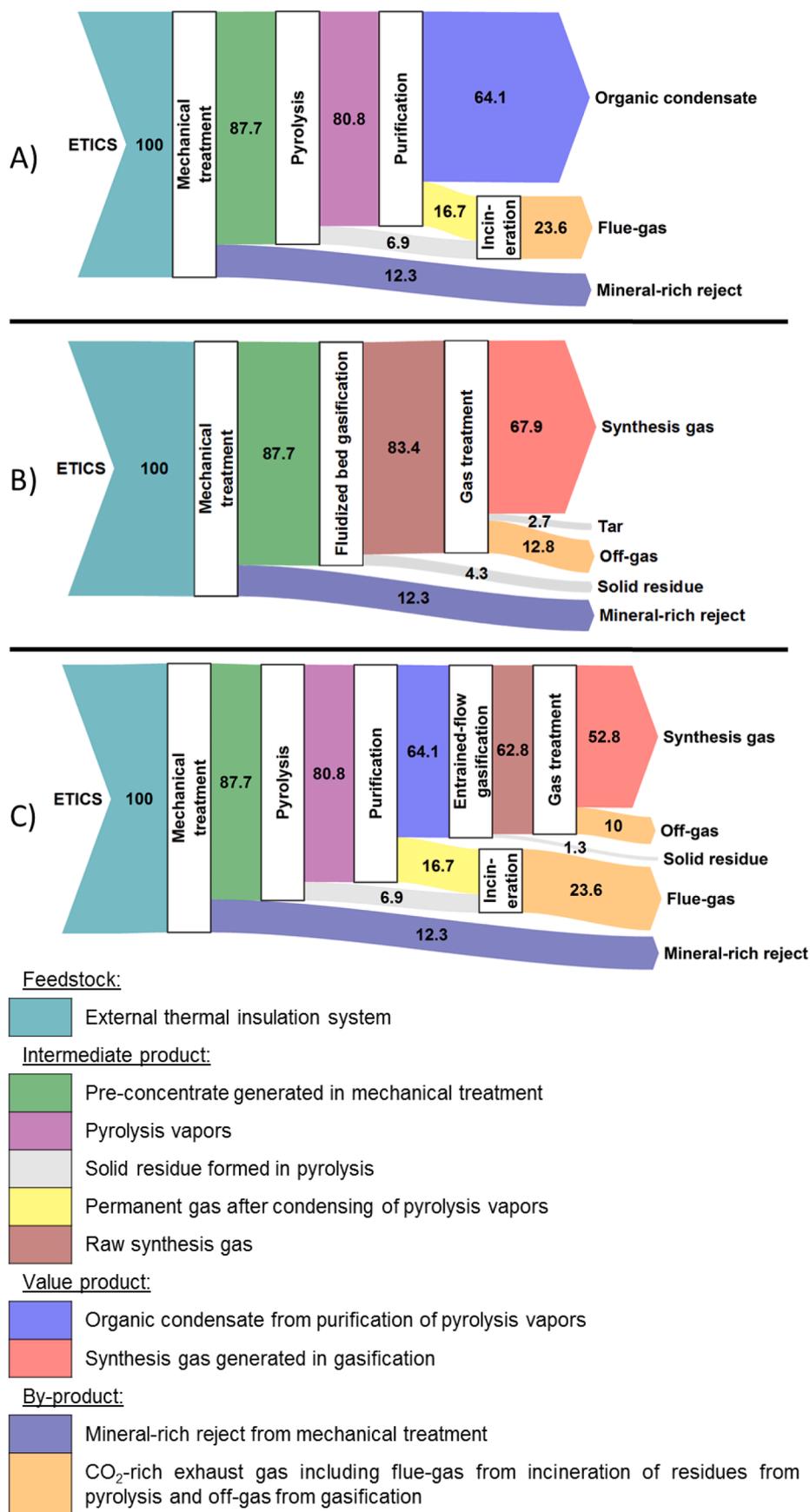
Fig. 4. Comparison of the carbon recycling rate (in wt. %) for different concepts including intermediate pyrolysis (A), fluidized bed gasification (B), and the hybrid concept (C) with organic condensate and synthesis gas (H2/CO = 1/1) as final products.

synthesis gas, result in higher recycling rates and product yields than the combined concept with pyrolysis and entrained flow gasification, where liquid pyrolysis products are converted to synthesis gas. On the contrary, stand-alone pyrolysis and fluidized bed gasification have lower technology readiness and achieving the required product quality is expected to demand more extensive post processing than large scale entrained flow gasification. Because of the high styrene monomer content generated in pyrolysis, recycling of the monomer should be considered as utilization. For gasification optimization, a feedstock adapted H_2/CO ratio in the synthesis gas should be focused on.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

Achilias, D.S., Kanellpoulou, I., Megalokonomos, P., Antonakou, E., Lapps, A.A., 2007. Chemical recycling of polystyrene by pyrolysis: potential use of the liquid product for the reproduction of polymer. Macromol. Mater. Eng. 292, 923–934. https://doi.org/10.1002/mame.200700058.

Albrecht, W., Schwitalla, C., 2015. Rückbau, Recycling und Verwertung von WDVS. Moglichkeiten der Wiederverwertung von Bestandteilen des WDVS nach dessen Rückbau durch Zuführung in den Produktionskreislauf des Dammstoffs bzw. Downcycling in dir Produktion minderwertiger Güter bis hin zur energetischen Verwertung, Fraunhofer IRB Verlag, Stuttgart.

Arena, U., 2012. Process and technological aspects of municipal solid waste gasification. A review. Waste Manage. 32 (4), 625–639. https://doi.org/10.1016/j.wasman.2011.09.025.

Arena, U., Zaccariello, L., Mastellone, M.L., 2009. Tar removal during the fluidized bed gasification of plastic waste. Waste Manage. 29 (2), 783–791. https://doi.org/10.1016/j.wasman.2008.05.010.

Artetxe, M., Lopez, G., Amutio, M., Barbarias, I., Arregi, A., Aguado, R., 2015. Styrene recovery from polystyrene by flash pyrolysis in a conical spouted bed reactor. Waste Manage. 45, 126–133. https://doi.org/10.1016/j.wasman.2015.05.034.

Bockhorn, H., Hornung, A., Hornung, U., Weichmann, J., 1999. Kinetic study on the non-catalysed and catalysed degradation of polyamide 6 with isothermal and dynamic methods. Thermochim. Acta 337, 97–110. https://doi.org/10.1016/S0040-6031(99)00151-3.

Cafiero, L., Fabbri, D., Trinca, E., Tuffi, R., Vecchio Ciprioti, S., 2015. Thermal and spectroscopic (TG/DSC–FTIR) characterization of mixed plastics for materials and energy recovery under pyrolytic conditions. J. Therm. Anal. Calorim. 121 (3), 1111–1119. https://doi.org/10.1007/s10973-015-4799-2.

Chen, D., Yin, L., Wang, H., He, P., 2014. Pyrolysis technologies for municipal solid waste: a review. Waste Manage. 34 (12), 2466–2486. doi:10.1016/j.wasman.2014.08.004.

Ciuffi, B., Chiaramonti, D., Rizzo, A.M., Frediani, M., Rosi, L., 2020. A critical review of SCWG in the contect of available gasification technologies for plastic waste. Appl. Sci. 10 (18), 6307–6342. https://doi.org/10.3390/app10186307.

DIN EN 15169:2007-05: Characterization of waste - Determination of loss on ignition in waste, sludge and sediments.

DIN EN 66165:2016-08: Particle size analysis - Sieving analysis - Part 1: Fundamentals.

Dogu, O., Pelucchi, M., Van de Vijver, R., Van Steenberge, P., D'hooge, D.R., Cuoci, A., Frassoldati, A., Faravelli, T., Van Geem, K.M., 2021. The chemistry of chemical recycling of solid plastic waste via pyrolysis and gasification: state-of-the-art, challenges, and future directions. Prog. Energy Combust. Sci. 84, 100901. https://doi.org/10.1016/j.pecs.2020.100901.

European Union, 2006. Regulation (EU) 2006/1907 of the European Parliament and the Council concerning the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH), establishing a European Chemicals Agency. Official Journal of the European Union. OJ L 396, 1.

Faravelli, T., Pinciroli, M., Pisano, F., Bozzano, G., Dente, M., Ranzi, E., 2001. Thermal degradation of polystyrene. J. Anal. Appl. Pyrol. 60 (1), 103–121. https://doi.org/10.1016/S0165-2370(00)00159-5.

Geyer, R., Jambeck, J.R., Law, K.L., 2017. Production, use and fate of all plastics ever made. Sci. Adv. 3 (7), e1700782 https://doi.org/10.1126/sciadv.1700782.

Han, J., Kim, H., 2008. The reduction and control technology of tar during biomass gasification/pyrolysis: an overview. Renew. Sust. Energ. Rev. 12 (2), 397–416. https://doi.org/10.1016/j.rser.2006.07.015.

Heller, N., Flamme, S., 2020. Waste management of deconstructed External Thermal Insulation Composite Systems with expanded polystyrene in the future. Waste Manage. Res. 38 (4), 400–407. https://doi.org/10.1177/0734242X20904413.

Ippolito, N.M., Cafiero, L., Tuffi, R., Vecchio Ciprioti, S., 2019. Characterization of the residue of a commingled post-consumer plastic waste treatment plant: a thermal, spectroscopic and pyrolysis kinetic study. J. Therm. Anal. Calorim. 138 (5), 3323–3333. https://doi.org/10.1007/s10973-019-09003-z.

Jang, M., Shim, W.J., Han, G.M., Rani, M., Song, Y.K., Hong, S.H., 2017. Widespread detection of a brominated flame retardant, hexabromocyclododecane, in expanded polystyrene marine debris and microplastics from South Korea and the Asia-Pacific coastal region. Environ. Pollut. 231, 785–794. https://doi.org/10.1016/j.envpol.2017.08.066.

Jiao, L., Xu, G., Wang, Q., Xu, Q., Sun, J., 2012. Kinetics and volatile products of thermal degradation of building insulation materials. Thermochim. Acta 547, 120–125. https://doi.org/10.1016/j.tca.2012.07.020.

Kannan, P., Biernacki, J.J., Visco, D.P., Lambert, W., 2009. Kinetics of thermal decomposition of expandable polystyrene in different gaseous environments. J. Anal. Appl. Pyrol. 84 (2), 139–144. https://doi.org/10.1016/j.jaap.2009.01.003.

Kusenberg, M., Eschenbacher, A., Djokic, M.R., Zayoud, A., Ragaert, K., De Meester, S., Van Geem, K.M., 2022. Opportunities and challenges for the application of post-consumer plastic waste pyrolysis oils as steam cracker feedstocks: to decontaminate or not to decontaminate? Waste Manage. 138, 83–115. https://doi.org/10.1016/j.wasman.2021.11.009.

Lindner, C., Weih, D., Fischer, E., 2017. Aufkommen und Management von EPS- und XPS-Abfallen in Deutschland 2016 in den Bereichen Verpackung und Bau. Conversio Market & Strategy GmbH, Frankfurt.

Lindner, C., Schmitt, J., Hein, J., 2020. Stoffstrombild Kunststoffe in Deutschland 2019. Conversio Market & Strategy GmbH, Mainaschaff.

Mehta, S., Biederman, S., Shivkumar, S., 1995. Thermal degradation of foamed polystyrene. J. Mater. Sci. 30 (11), 2944–2949. https://doi.org/10.1007/BF00349667.

Moens, E.K.C., De Smit, K., Marien, Y.W., Trigilio, A.D., Van Steenberge, P.H.M., Van Geem, K.M., Dubois, J., D'hooge, D.R., 2020. Progress in reaction mechanism and reactor technologies for the thermochemical recycling of poly(methyl methacrylate). Polymers 12, 1667. doi:10.3390/polym12081667.

PlasticsEurope, 2020. Plastics - The Facts 2020. An analysis of European plastics production, demand and waste data. Available from: <https://plasticseurope.org/de/wissenshub/> (accessed 3 February 2022).

PolyStyreneLoop B.V., 2019. Guideline on collection and pretreatment of PS foam for PolyStyreneLoop recycling. Available from: <https://polystyreneloop.eu/downloads/> (accessed 3 February 2022).

PolyStyreneLoop B.V., 2020. The circular economy in action. Closed-loop recovery of polystyrene foam and bromine. Available from: <https://polystyreneloop.eu/downloads/> (accessed 3 February 2022).

Pretz, T., Feil, A., Julius, J., 2017. Aufbereitung fester Abfallstoffe. In: Kranert, M. (Ed.), Einführung in die Kreislaufwirtschaft. Springer Fachmedien Wiesbaden, Wiesbaden, pp. 189–253.

Ragaert, K., Delva, L., Van Geem, K.M., 2017. Mechanical and chemical recycling of solid plastic waste. Waste Manage. 69, 24–58. https://doi.org/10.1016/j.wasman.2017.07.044.

Rahimi, A., García, J., 2017. Chemical recycling of waste plastics for new materials production. Nat. Rev. Chem. 1 (6) https://doi.org/10.1038/s41570-017-0046.

Rani, M., Shim, W.J., Han, G.M., Jang, M., Song, Y.K., Hong, S.H., 2014. Chemosphere 110, 111–119. https://doi.org/10.1016/j.chemosphere.2014.02.022.

Rewindo GmbH, 2020. Kunststofffenster-Recycling in Zahlen 2019. Available from: <https://rewindo.de/infomaterial/> (accessed 3 February 2022).

Riedel, W., Oberhaus, H., Frossel, F., Haegele, W., 2010. Warmedamm-Verbundsysteme, Von der Thermohaut bis zur transparenten Warmedammung. second rev. ed. Waldshut-Tiengen, Fraunhofer IRB Verlag, Stuttgart.

Rodríguez, E., Gutiérrez, A., Palos, R., Vela, F.R., Arandes, J.M., Bilbao, J., 2019. Fuel production by cracking of polyolefins pyrolysis waxes under fluid catalytic cracking (FCC) operationg conditions. Waste Manage. 93, 162–172. https://doi.org/10.1016/j.wasman.2019.05.005.

Sancho, J.A., Aznar, M.P., Toledo, J.M., 2008. Catalytic air gasification of plastic waste (polypropylene) in fluidized bed. Part I: Use of in-gasifier bed additives. Ind. Eng. Chem. Res. 47 (4), 1005–1010. https://doi.org/10.1021/ie071023q.

Seo, J.M., Hwang, B.B., 2006. A reappraisal of various compacting processes for wasted expandable polystyrene (EPS) foam. Mater. Sci. Forum. 532–533, 261–264, doi:10.4028/www.scientific.net/MSF.532-533.261.

Stapf, D., Seifert, H., Wexler, M., 2019. Thermische Verfahren zur rohstofflichen Verwertung kunststoffhaltiger Abfalle. In: Thiel, S., Thomé-Kozmiensky, E., Quicker, P., Gosten, A. (Eds.), Energie aus Abfall. Thomé-Kozmiensky Verlag, Neuruppin, pp. 357–375.

Sto SE & Co. KGaA, 2017. Technisches Merkblatt: StoLevellClassic. Available from: <https://www.sto.de/s/> (accessed 3 February 2022).

Tomasi Morgano, M., Leibold, H., Richter, F., Seifert, H., 2015. Screw pyrolysis with integrated sequential hot gas filtration. J. Anal. Appl. Pyrol. 113, 216–224. https://doi.org/10.1016/j.jaap.2014.12.019.

Zeller, M., Netsch, N., Richter, F., Leibold, H., Stapf, D., 2021. Chemical recycling of mixed plastic wastes by pyrolysis – pilot scale investigations. Chem. Ing. Tech. 93 (11), 1763–1770. https://doi.org/10.1002/cite.202100102.