

Article

Comparative Life Cycle Assessment of Catalytic Intermediate Pyrolysis of Rapeseed Meal

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Abstract: Biowaste valorization is a means for tackling resource depletion and climate change, which gives rise to environmental benefits and economic growth. One of the most known technological routes to convert biowaste into bioproducts is pyrolysis, which may conduct with and without catalyst application. The purpose of this study was to investigate an early-stage life-cycle assessment (LCA) for catalytic intermediate pyrolysis to valorize rapeseed meal, split over the scenarios using ZSM-5 and zeolite Y catalysts. Four selected environmental impact categories were assessed by IMPACT 2002+ methodology. The results revealed that the ZSM-5 catalytic pyrolysis led to bigger environmental impacts than the pyrolysis utilizing zeolite Y in all compared impact categories except global warming. The scenario that involved zeolite Y had around 20% GHG intensity greater than ZSM-5 pyrolysis. The bulk of GHG emissions mostly involved CO₂ and methane generated from electricity consumption, which was provided by fossil resources. Applying ZSM-5 in the pyrolysis increased environmental burdens in non-renewable energy, respiratory inorganics, and terrestrial ecotoxicity by 140.88 MJ primary, 8.83×10^{-3} kg PM_{2.5} eq. and 125.63 kg TEG soil, respectively. The major driving factor of high value in mentioned categories was the manufacturing process of the ZSM-5 catalyst by utilizing natural gas and chemicals, such as phosphorus trichloride, sodium hydroxide and sodium silicate. Given that catalysts can play a substantial role in the emissions resulting from bio-based products, hence LCAs of pyrolysis should consider the potential influence of catalysts in the valorization processes. This study can predict environmental hotspots in the early stages of bio-waste valorization and show the potential defects of implanted biorefinery at pilot/industrial scales.

Keywords: LCA; intermediate pyrolysis; zeolite catalyst; biochar; pyro-gas; bio-oil

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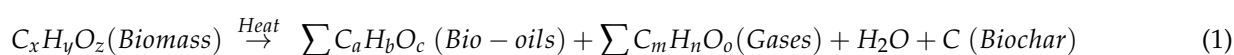
1. Introduction

In recent years, a growing concern about resources depletion owing to the demographic explosion has led to research attempts for finding alternatives for food, energy, and water (FEW) resources [1]. Renewable resources are promising substitutions, which can address not only FEW needs but also mitigate environmental impacts [2]. In the case of energy sources, fossil fuels provide 95% of transportation energy in most countries. Only a few countries have deployed more than 10% of renewable energy sources in the transportation section and most other countries' shares of renewable energies are 4 to 6% or even lower [3]. The current condition brings environmental challenges, particularly in terms of resource depletion and global warming, which shows the importance of renewable energies. Biomass energy has been noticed for substituting fossil fuels with renewable resources in recent years. Biomass refers to organic material, including energy crops and non-food sources, such as biowaste from agricultural, industrial, and household residues that could be a favorable source among all different biomass types due to no threat to food supply

security and potential to implement a circular economy model. Using waste as a feedstock to produce bioenergy is a means for tackling resource depletion and climate change, which gives rise to environmental benefits and economic growth [4]. One of the most attractive processes to produce renewable energies is pyrolysis for biowaste conversion to value-added products, which provides profitable solutions for waste management and improves resource reuse [5]. The first step before starting any combustion and gasification process is the pyrolysis process always [6], involving the thermal decomposition of organic materials with calorific potential into bio-oil, biochar, and combustible/non-combustible gases in the absence of oxygen by using only heat or in the presence of a catalyst [7,8].

Depending on the process configuration and the condition of the reaction, pyrolysis can be classified into slow, intermediate, and fast pyrolysis [9]. In each of the methods of pyrolysis, the feedstock should be prepared in a different form for having an efficient process. Accordingly, in fast pyrolysis, usually, the feedstock should be ground and dried while slow pyrolysis as a classical approach is applicable for handling logs. The feedstock preparation for intermediate pyrolysis could be converting the materials to larger particles than fast pyrolysis. This method is used for particles up to pellet and chip size as well as substrates containing up to 40% of water. It should be noted that intermediate pyrolysis has been presented as a preferable process for non-woody feedstock by some studies. Regarding the end products, it should be said that the liquids, gases, and biochar produced by different pyrolysis routes differ in terms of quantity and quality [10–12].

The non-combustible gases due to the high content of CO₂, CO, H₂, and CH₄ can be converted into electricity or burned to provide heat for the processes as well as valuable chemicals production [13]. Bio-oil or tar as a liquid fraction produced during the pyrolysis is not usable as fuels directly due to the high water content, acidity and viscosity. Upgrading processes are necessary to treat the bio-oil for a product as a fuel or electricity source [14]. Another output of pyrolysis is biochar (charcoal) an environmentally friendly product. Improving the characteristics of biochar converts it to a product for various sustainable benefits, but the most used application of that is as a soil amendment. Biochar as a carbon-rich product can improve the soil condition by sequestering carbon, neutralizing soil acidity, and improving the cation exchange capacity (CEC) [15,16]. Hence, pyrolysis could be a sustainable process with environmental savings by using waste streams to produce high hydrocarbon compounds. Pyrolysis is an endothermic process, which can occur without any external agent in a thermal (conventional) route in a general form as follow [17]:



In catalytic pyrolysis, a catalyst may be used for improving the yields or optimizing the process parameters, such as temperature and/or time of the process [18]. Catalysts can increase the oil and gas yields by further cracking C–C bonds and reduce the overall required energy of the process and the concentration of the impurities [19,20]. Based on the above-mentioned, catalytic pyrolysis could be considered a sustainable valorization platform in biorefineries. IEA Bioenergy—Task 42 has described biorefining as a sustainable technique used to convert biomass into a spectrum of bio-products and bioenergy [21]. However, biorefineries provide a lot of benefits by producing marketable bio-based products, a commercial biorefinery is not zero-pollution. Biorefining activities can vary by different parameters, such as diverse biomass feedstock, installed technology, and other conditions [22]. Therefore, a tool is needed for measuring the environmental benefits and burdens of biorefinery plants. In this context, there is a quantitative methodology known as life cycle assessment (LCA) to identify and assess key environmental impacts of the life cycle of bioenergy and biomaterials [23]. This method is widely applied for a comprehensive assessment of environmental burdens during the life cycle of the products/services [24,25]. To this end, recently a variety of research has conducted LCA for pyrolyzed-derived products to provide environmental savings and improve the challenges of applying the pyrolysis process. These studies have been carried out through different feedstock and technical approaches to implement the LCA. The results presented by Brassard et al. showed the

role of various parameters, such as biomass, technology, side-products, properties, and marginal technologies in the environmental performance of pyrolysis [26]. The result presented a better environmental performance in ten out of sixteen investigated impact categories and showed the environmental preference of a pyrolysis scenario to leaving the residues on the soil to decay. In 2020, Patel et al. evaluated the greenhouse gas emissions of bio-oil derived from both catalytic and thermal pyrolysis of waste pinewood chips, which could provide significant GHG emission savings compared to the biofuels from first-generation feedstock [27]. Additionally, an LCA study for pyrolysis of municipal solid waste (MSW) as one of the waste management options was performed in 2021 by Chhabra et al. [28]. The research showed that the climate change impact of the process was less than the impact caused by dumping and sanitary landfilling methods. Li et al. confirmed the environmental benefits of pyrolysis of surgical mask waste to produce sustainable fuels and energy through life cycle analysis [29]. Based on the review of previously published studies, there is a growing interest in the LCA of the pyrolysis process in recent years that includes mostly thermal pyrolysis, but a handful of studies have concerned the environmental loads of catalysts in the biofuel supply chain. Gupta et al. provided a comparison of different catalytic pyrolysis in terms of the impact of co-product recycling, catalyst types, and market export on environmental load and process economics [30]. The LCA outcomes showed that using Ni/Al₂O₃ in the pyrolysis made the process more efficient with less environmental impact than pyrolysis with Al₂O₃. Another comparative research carried out by Monteiro et al. in 2022 stated that catalysts do not always improve environmental performance. The study compared the LCA outputs of different pyrolysis routes (catalytic and non-catalytic) for two polymers involving high-impact polystyrene and high-density polyethylene. The obtained results showed the environmental disadvantage caused by using the catalyst technology [31].

Considering the fact that catalysts have an influence on the environment and a few studies that were performed on catalytic pyrolysis, the authors of the present research were motivated to perform a comparative LCA of catalytic pyrolysis. The novel aspects of this study consist of using an early-stage LCA for catalytic intermediate pyrolysis utilizing zeolite socony mobil-5 (ZSM-5) and zeolite Y (ZY) catalysts. Based on previous research, there are very few studies of performing LCA for catalytic pyrolysis, and none of them considered a zeolite Y catalyst as well as rapeseed meal as feedstock. In Europe, the highest market demand for the use of rapeseed is animal feed manufacturing, the food industry, and biodiesel production [32]. As a dominant oilseed crop in Europe, rapeseed accounts for approximately 25% of world production, with Germany, Poland, and France as leading producers in the Europe [33]. The increasing rapeseed oil production coincides with a growth of a by-product from the oil extraction process. This by-product, called rapeseed meal (RM), is a high protein source used as animal feed, which produced 40 million tons and 12.5 million tons in 2020 in the world and the EU, respectively [34].

This study aimed to examine the early-stage LCA for intermediate pyrolysis of post-extraction rapeseed to bio-products split over two distinct scenarios. An early-stage LCA is a very effective tool to identify the wastage of materials and energy in the processes for improving the selection, design, and optimization of a product or process. Through the results of a review, Azapagic in 1999 showed that the emerging life cycle process design (LCPD) tool can help to develop technological innovation by selecting the best alternatives of the process and compounds over the entire life cycle [35]. Accordingly, this study can predict environmental hotspots in the early stages of bio-waste valorization and show the potential defects of implanted biorefinery at pilot/industrial scales.

2. Materials and Methods

This section describes the pyrolysis processes for converting rapeseed meal to bio-products using two different catalysts and LCA methodology applied for assessing the environmental performance. As the analyzed processes are still in the early stages of development, LCA modeling includes some assumptions presented in this part.

2.1. Description of Pyrolysis Experiments

2.1.1. Biomass Preparation

As mentioned in the introduction, due to population growth, the increment of environmental burdens and natural resource depletion, the replacement of renewable sources for energy production is of great importance. Hence, the valorization of rapeseed meal due to their reasonable price and availability all year round could be exploited as an excellent source of bioenergy and bio-materials production. However, many recent LCA studies have been conducted on rapeseed oil production and rapeseed meal as animal feed, but there are no LCA studies on rapeseed meal valorizing through the pyrolysis process. The rapeseed meal sample used in this study was collected from a rapeseed pressing plant in Poland. The RM samples were dried in ambient conditions and sieved (300–750 µm) in the pre-treatment stage of the pyrolysis process. Table 1 shows the characteristics of the rapeseed meal used in this study.

Table 1. Characteristics of rapeseed meal.

Parameters	Value
Proximate analysis (%)	
Moistur	5.66
Volatile Matter	73.99
Ash	6.80
Fixed Carbon	13.55
Ultimate analysis (%)	
Carbon	42.57
Hydrogen	6.40
Nitrogen	5.80
Sulphur	0.61
Oxygen ^a	37.82
Component analysis (%)	
Cellulose	8.91
Hemicellulose	8.09
Lignin	7.23
Extractives	75.77

^a—calculated by difference, Oxygen = 100% – Carbon – Hydrogen – Nitrogen – Sulphur – Ash.

2.1.2. Catalysis

The pyrolysis process and the final yields are affected by parameters including heating rate, feedstock composition, residency type, particle size, process temperature, catalyst type, and catalyst loading mechanism [9,36]. Dutta et al. revealed that developing the catalysts plays a key role in the high efficiency of pyrolysis. Despite the significance of catalysts in the biofuel supply chain, these compounds are usually skipped from the system boundary of LCAs' studies [37]. One of the reasons to exclude the catalysts in the LCA studies is the lack of energy and material data for manufacturing, recycling, and reclaiming those as important components in the design and optimization of the process [38].

As was demonstrated, catalysts can increase the reaction rate by lowering the activation energy of the reaction initiation step. In the general classification, catalysts are categorized into two main types: heterogenous and homogenous [18]. The catalysts used in this study belong to the zeolite family of the heterogenous class, ZSM- 5 and zeolite Y. Zeolite is the most common and best catalyst for the cracking process due to its high thermal stability, crystallized structure, and ability to reach out to the reactants trapped in the pores [39]. In 2019, Abu Suleiman et al. presented the composition of both catalysts, as given in Table 2 [40].

Table 2. Chemical composition of ZSM-5 and ZY (Data adapted from [40]).

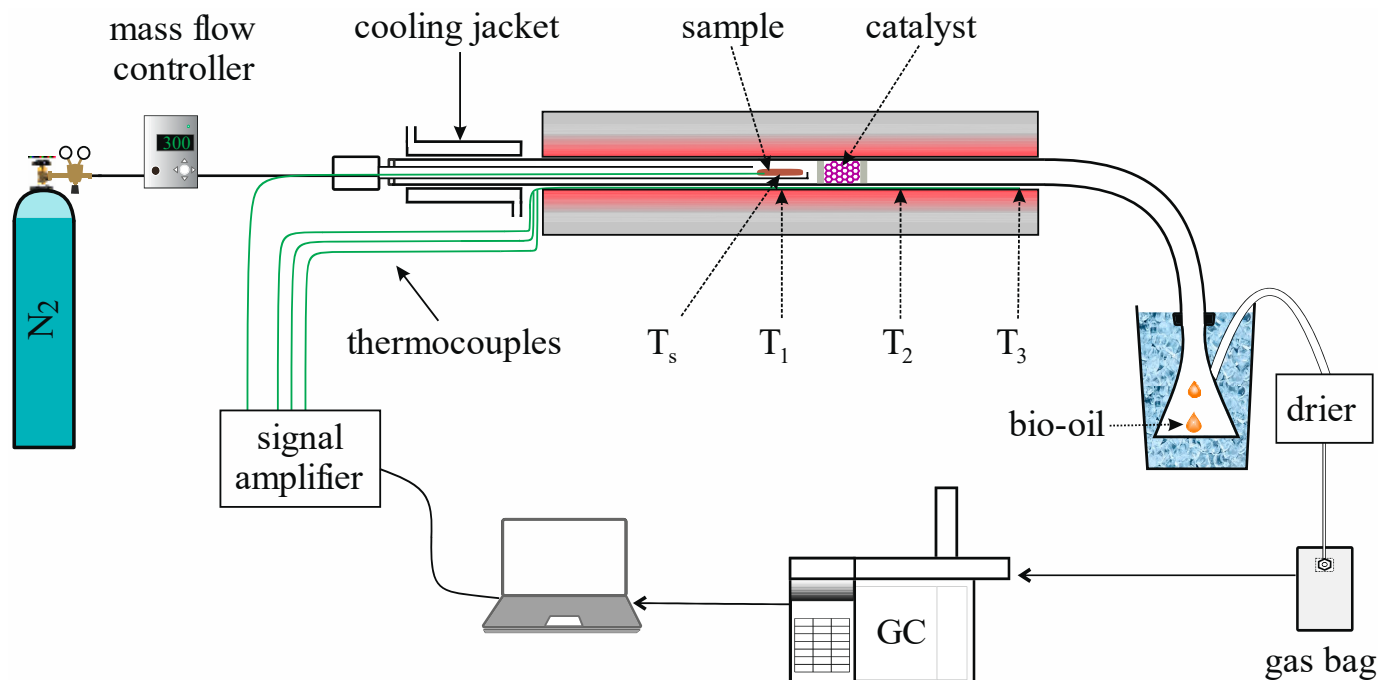
Framework Code	Zeolite Type	SiO ₂ %	Al ₂ O ₃ %	Si/Al %	Na ₂ O %
MFI (Pentasil family)	ZSM-5	92.13	5.67	13.8	2.21
FAU (Faujasite family)	Zeolite Y	60.04	28.30	1.8	11.64

According to information in the literature, when ZSM-5 is used as a catalyst, it is often combined with fillers and binders, such as silica gel and kaolin, to form a catalyst [41,42]. In this study, to distinguish ZSM-5 and the mixture of ZSM-5 with fillers and binders, the first was referred to as a ZSM-5 compound and the latter as a ZSM-5 catalyst.

2.1.3. Pyrolysis

In present study, the catalytic pyrolysis of post-extraction rapeseed meal to biochar, bio-oil, and pyro-gas was considered based on the experiments conducted by Jerzak et al. in 2022 [10]. The experiment started by placing the catalyst (1.5 g) in a cylindrical fixed bed reactor, which was externally heated to reach 500 and held at a stable temperature for 1 h. During the experiment, the temperature was retained by a control unit linked to a K-type thermocouple and pyrolysis chamber.

Afterward, the RM sample (1.5 g) was set on a boat in the area of the water cooler, and the reactor was under a nitrogen flow purge at a rate of 100 mL/min for 5 min. Once the reactor has reached the optimum operation conditions, the RM boat was fed into the central heating zone of the pyrolizer. After finishing the pyrolysis process, the sample boat was returned to the cooling water zone. The hot volatiles streamed to the ice tank where the aqueous and oil phases were condensed, and the non-condensed dried gases were accumulated in a Tedlar bag for gas chromatography analysis. Figure 1 presents the constructed apparatus for rapeseed meal pyrolysis in this study.

**Figure 1.** Intermediate pyrolysis system adapted from [10].

2.2. Life Cycle Assessment Methodology

LCA is a standardized method to assess the environmental burdens associated with products, services, or processes from the extraction of unprocessed materials to the end of their useful life [43]. According to ISO 14040 and 14044 standards, LCA is based on four main phases [44,45]: (1) goal and scope, all methodological parameters, such as functional unit, system boundary, etc., are defined in this step; (2) inventory analysis, which concerns the foreground and background data collection for inputs and outputs of the processes; (3) impact assessment, the data inventory is evaluated to measure and express the environmental impacts; and (4) interpretation as the last stage for presenting the results.

2.2.1. Goals and Scope Definition

This section implies establishing goals to provide accurate aims, methods, outcomes, and planned applications that are optimally aligned. The goal of the study is to identify, measure, assess, and compare the LCA results of defined scenarios for pyrolyzing rapeseed meal. The scope of the LCA covers the intermediate pyrolysis within a “gate–gate” system boundary, shown in Figure 2, in which a chain of material/energy streams occurs to produce the final products of interest.

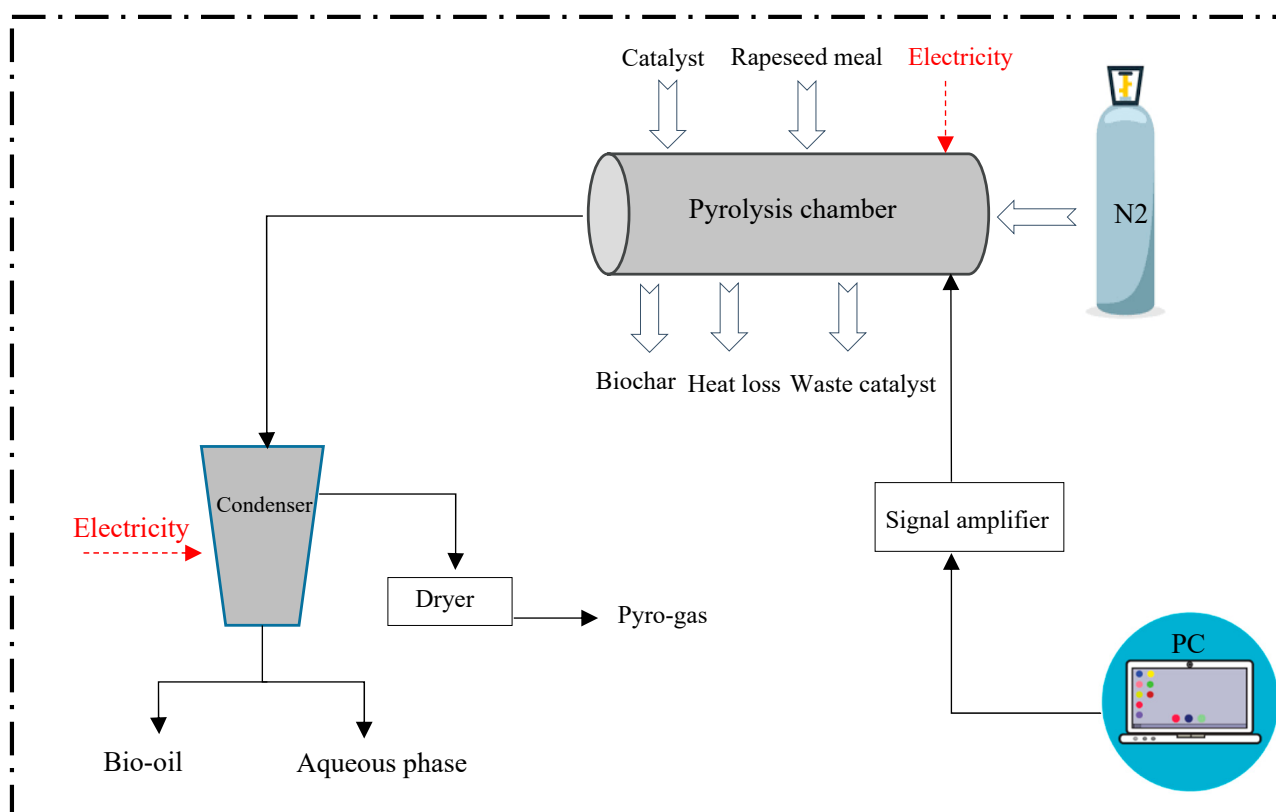


Figure 2. System boundary of valorizing the RM via pyrolysis.

Two scenarios have been modeled based on different catalyst usage. One scenario presented the RM valorization process by using a ZSM-5 catalyst, and the other one investigated the pyrolysis, including the ZY catalyst. SimaPro version 8.5.2.0 LCA software [46] was used to model the processes, and functional unit (FU) was defined as the conversion of 1 kg of RM substrate. The manufacturing of equipment was excluded from the system boundary, and the focus of the study was on the operational system. Additionally, the upgrading process, usage, and end-of-life of the products are out of the defined system boundary. The utilized electricity for the processes was calculated based on the electricity grid of Poland.

2.2.2. Inventory Analysis

Life cycle inventory (LCI) contains input/output data of all processes concerned in the system boundary. Therefore, LCI is an influential stage in that the accuracy of LCA output is dependent on the quality and sufficiency of its content [47].

In this study, the details of rapeseed meal valorization were acquired from laboratory experiments. The LCIs were prepared according to the optimized quantity of the materials, chemicals, and energy used in the experiments, the literature, and the reports issued by Argonne national laboratory [41,48] and Ecoinvent database v3 [49]. Table 3 describes the data used in this study to clarify the quality of the inventories.

Table 3. Sources and types of data used in the study.

Data Source	Data Type
i. Literature	i. Measured
ii. Experiments	ii. Calculated
iii. Consultation	iii. Estimated
	iv. Average value

For generating the LCIs, limited data regarding material and energy consumption in the catalyst production caused calculations and estimates based on publicly available information. Furthermore, in the analysis, the “zero burden assumption” was considered, in which upstream environmental loads were not involved in the analysis. The major energy/material inputs required for pyrolysis conversion are summarized and presented in Table 4.

Table 4. Material and energy flows involved in catalyzed pyrolysis system.

	Value	Unit
<i>Pyrolysis process</i>		
RM	1	kg
ZSM-5/Zeolite Y	100	g
N ₂	240	dm ³
Electricity	7.86	kWh
<i>* Substances/energy per ton ZSM-5 catalyst production</i>		
ZSM-5 compound	0.5	ton
SiO ₂ gel	0.25	ton
Kaolin	0.25	ton
Natural gas	60.22	mmBtu
Electricity	0.36	mmBtu
<i>* Substances/energy per 1 kg ZY production</i>		
Quartz	1.291	kg
Bauxite	0.489	kg
Rock salt	1.212	kg
Limestone	0.878	kg
Sodium silicate solution	4.498	kg
Aluminum hydroxide	0.389	kg
NaOH	0.220	kg
Soda	0.670	kg
Energy consumption	46.259	MJ

* Data adapted from [38,48].

2.2.3. Impact Assessment

The environmental impact of products/services is calculated in the life cycle impact assessment (LCIA) stage via a selected impact assessment method. This phase contains various methods for measuring the impact categories of the environmental impacts within the system boundary. [50,51]. Based on the LCI of the inputs and outputs of the processes, the environmental impact categories were measured by the IMPACT 2002+ methodology. The methodology included 17 midpoint impact categories and 4 endpoint categories by

linking LCI results via several midpoint categories to several damage categories. All midpoint scores in IMPACT 2002+ are described in units of a reference substance and related to the four damage-oriented categories comprising ecosystem quality, human health, climate change, and resources [52].

Out of the categories with the midpoint approach in IMPACT 2002+ methodology, four impact categories were investigated in this study presented in Table 5. The selection of the categories was based on the main processes reflecting environmental issues related to the pyrolysis system. These categories are classified into different types according to the midpoint indicators and then characterized into the common equivalent units, which mirror their contributions to the midpoint impacts. The normalized results are weighted and aggregated to deliver a single-score LCA result, which is an inclusive and a convenient form to show the outcomes.

Table 5. Selected impact categories in this study.

Impact Category	Midpoint Reference Substance	Abbr.
Respiratory inorganics	kg PM _{2.5} into air _{eq}	RIOs
Terrestrial ecotoxicity	kg Triethylene glycol into soil _{eq}	TE
Global warming	kg CO ₂ equivalent	GW
Non-renewable energy	MJ primary	NRE

3. Results and Discussion

The findings showed that applying ZY had better environmental performance than the ZSM-5 pyrolysis process. Following, the results are broken down in the text and figures to show the contribution to the total impacts from pyrolysis performed by two catalysts.

3.1. Environmental Impacts of the Catalytic Pyrolysis

Figure 3 presents the contribution of each of the scenarios to the overall environmental burdens of the processes through the IMPACT 2002+ method. This signifies that applying ZSM-5 in the pyrolysis process leads to bigger environmental burdens in all compared impact categories (except global warming (GW)) than the process utilizing ZY. The amount of GW caused by zeolite Y pyrolysis was 1.29 millipoint (mPt), which showed a higher value than GW emissions released by the ZSM-5 pyrolysis process.

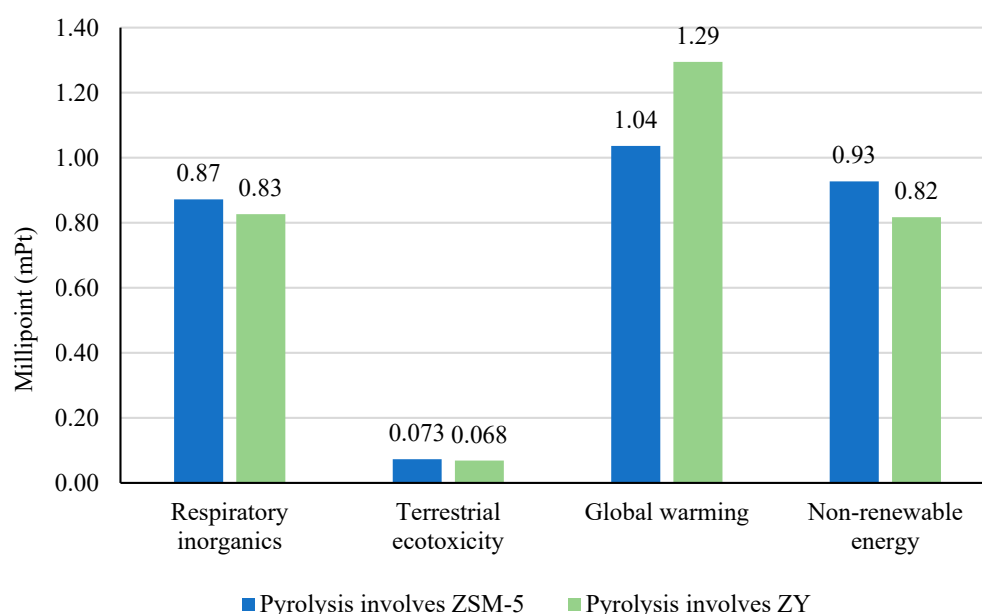


Figure 3. Single score LCA results for two different catalytic pyrolysis processes.

Global warming (GW)

In both scenarios, the bulk of GHG emissions within the conversion process came from the electricity generation for running the process. Most global warming caused by GHG emissions involved CO₂ and methane. Figure 4a,b shows the share of GHG emissions in both scenarios. The results showed that global warming potential was reliant on the extent of CO₂ emissions (12.51 kg CO₂ eq. in ZY pyrolysis and 9.91 kg CO₂ eq. in ZSM-5 pyrolysis) depending on the fossil fuels burned during the processes. Although methane has a smaller contribution to GW than carbon dioxide, its amount in the ZSM-5 pyrolysis is 0.27 kg CO₂ eq., which is higher than (0.23 kg CO₂ eq.) methane emitted from pyrolysis by ZY. The main reasons for the increase were natural gas, phosphorus trichloride, and sodium silicate consumption during the different stages of ZSM-5 production. The LCA study conducted by Benavides et al. revealed that catalysts can contribute remarkably to biofuel GHG emissions. They confirmed the catalysts' influence on GHG emissions depends on the catalysts' production and their loading rates during the pyrolysis process [38].

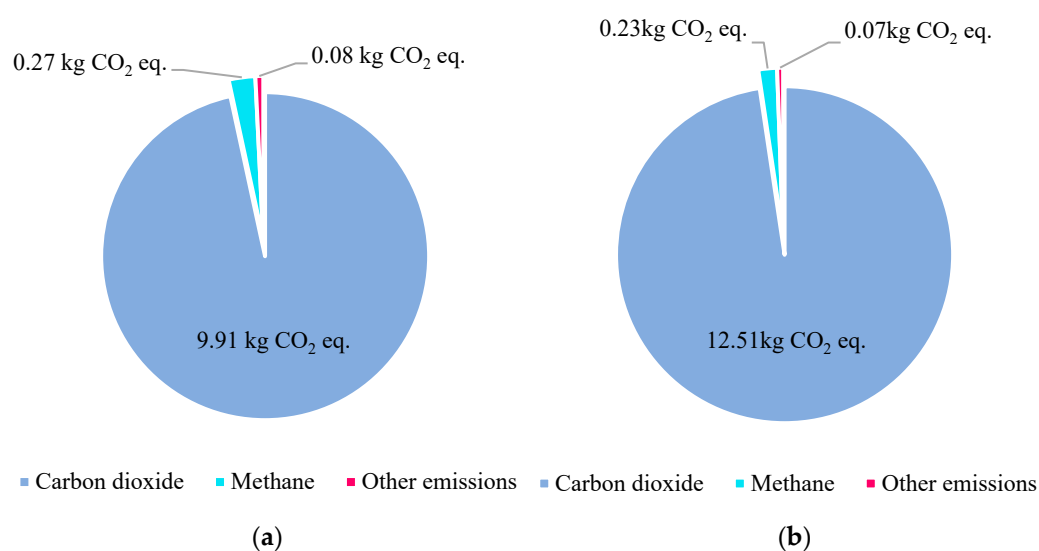


Figure 4. (a) Contribution of GHG emissions in pyrolysis involved ZSM-5 catalyst. (b) Contribution of GHG emissions in pyrolysis involved zeolite Y catalyst.

As aforementioned, the scenario involving ZY is the dominant contributor to global warming (around 20%) in comparison to the ZSM-5 catalytic pyrolysis. For the pyrolysis with ZY, the GW was 12.81 kg CO₂ eq. per 1 kg pyrolyzed rapeseed meal due to the bigger amount of energy needed for the ZY catalyst production provided by fossil resources. The GHG emissions of the ZY catalyst production were dominated by the production of soluble sodium silicate and electricity. The emissions of those two sources were estimated to be 2.08 and 1.79 kg CO₂ eq., respectively. Furthermore, aluminum hydroxide, by representing 0.45 kg CO₂ eq. emission, contributes to the global warming impact category during the ZY catalyst production. Other materials flow, such as soda and deionized water consumption, had an extremely small potential to release GHG emissions during their life cycle, which included 0.03 kg CO₂ eq. and 0.05 kg CO₂ eq., respectively.

Non-renewable energy (NRE)

The non-renewable energy impact category accounts for consuming primary depletable energy resources, mostly including crude oil, natural gas, coal, or uranium. Dang et al. carried out an LCA study to compare non-renewable energy demand and global warming potential for 1 MJ pyrolyzed biofuel from corn stover with conventional fuel. The research demonstrated the reduction in net non-renewable energy by 147.5% and a net GWP reduction of 119.4% in pyrolysis scenarios in comparison to gasoline and diesel scenarios [53].

In the present study, the NRE impact category quantifies the amount of non-renewable energy employed for each of the proposed scenarios relative to one another. This was reflected in the current study by an NRE value of 140.88 MJ primary in ZSM-5 pyrolysis compared to 124.18 MJ primary resulting from ZY pyrolysis. In both scenarios, the main contributor to the NRE was electricity production due to utilizing coal in the grid mix of electricity in Poland.

Apart from the amount of NRE caused by electricity production, using the ZY catalyst in the pyrolysis offered NRE environmental savings of 18.60 MJ primary in comparison to pyrolysis with ZSM-5. The higher value of non-renewable energy in the ZSM-5 catalytic pyrolysis was influenced by the production processes of the ZSM-5 catalyst. Table 6 demonstrates the input flow of the catalytic pyrolysis that caused the superiority of the NRE value resulting from the ZSM-5 production. Accordingly, after electricity as a common input in both catalytic pyrolysis, the consumption of natural gas plays a key role to raise the value of the NRE impact category to 11.77 MJ primary in ZSM-5 pyrolysis. The third contributor to the NRE value (2.50 MJ primary) was phosphorus trichloride used in the production of TPAOH (tetrapropylammonium hydroxide) as a raw material of the ZSM-5 component.

Table 6. NRE resulted from material and energy inputs in pyrolysis process included ZSM-5 catalyst.

Material/Energy	Value (MJ Primary)
Electricity	118.99
Natural gas	11.77
Crude alumina (Al ₂ O ₃)	0.18
Ethylene (C ₂ H ₄)	1.80
Heat	0.13
Kaolin	0.08
Phosphorus trichloride (PCl ₃)	2.50
Silica sand (SiO ₂)	0.02
Sodium hydroxide (NaOH)	0.61
Sodium silicate (Na ₂ SiO ₃)	1.60
Sulfuric acid (H ₂ SO ₄)	0.88
Syngas	0.51
Ammonia	0.16
Deionized water	0.03
Nitrogen (N ₂)	1.62

Respiratory inorganics (RIOs)

Respiratory inorganics is a midpoint impact category contributing to human health damages. The RIOs are defined as the total emission expressed as particulate matter equivalent (PM_{2.5} eq.), which can cause respiratory diseases. Steele et al. investigated human respiratory effects, including emissions, such as SO₂, PM < 2.5 µm, PM > 10 µm, and NO₂, from the fast pyrolysis process of pine chips and residual fuel oil (RFO). The results showed that the contribution of RFO in the human respiratory effect was 32% more than pyrolyzed bio-oil because of the high release of SO₂ [54].

In the current research, based on the LCA results from analyses, the respiratory inorganics impact category had a very close value in both scenarios, 8.83×10^{-3} kg PM_{2.5} eq., for using ZSM-5 in the process, and 8.37×10^{-3} kg PM_{2.5} eq., for pyrolysis with ZY. The main determinant of respiratory inorganics for the scenarios is the emission of sulfur dioxide, followed by particles (PM_{2.5}) and nitrogen oxides. Figure 5 illustrates the individual processes' input contribution to inorganic emissions from pyrolysis. Contribution analysis revealed that, in the case of using ZSM-5 in pyrolysis, the major part (90.76%) of the respiratory inorganic emitted from electricity consumption included low- and medium-voltage electricity produced by Poland's electricity network. Phosphorus trichloride (2.88×10^{-4} kg PM_{2.5} eq.), sodium silicate (1.61×10^{-4} kg PM_{2.5} eq.), sodium hydroxide

(7.56×10^{-5} kg PM_{2.5} eq.), and nitrogen (6.78×10^{-5} kg PM_{2.5} eq.) were in the next ranks to release the inorganic emissions to the air.

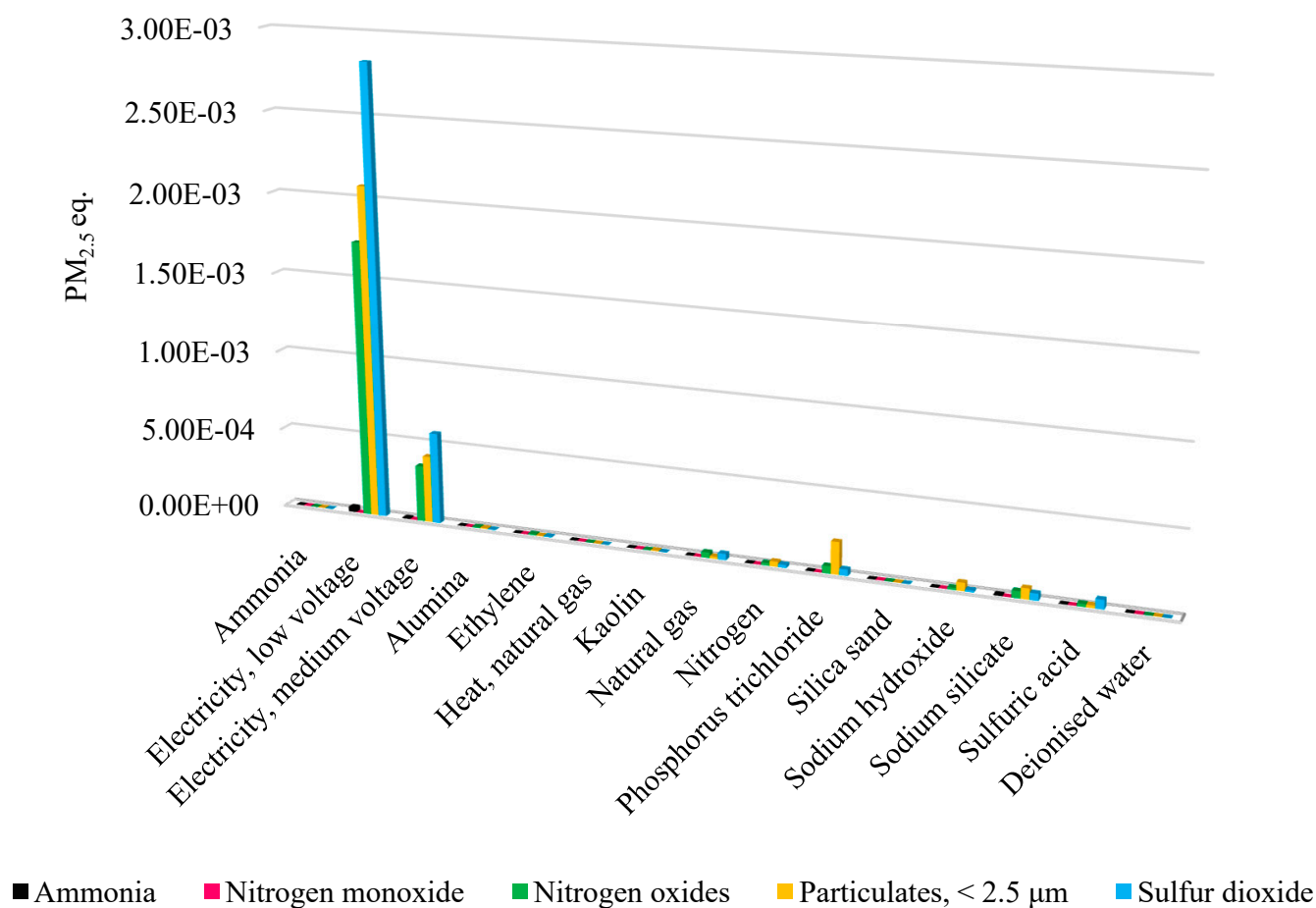


Figure 5. Inputs contribution to inorganic emissions of pyrolysis with ZSM-5 catalyst.

Terrestrial ecotoxicity (TE)

Terrestrial ecotoxicity represents the release of substances with ecotoxic effects on air, water, and soil. Analysis of the TE impact category revealed that catalyst consumption caused the highest amount of terrestrial ecotoxicity in pyrolysis. The result is also consistent with the comparative LCA study carried out by Gupta et al. Their research showed that the highest value of terrestrial ecotoxicity potential belonged to catalysts' (Al₂O₃ and Ni/Al₂O₃) consumption in pyrolysis in comparison to non-catalytic pyrolysis [30]. Pyrolysis with ZSM-5 (125.63 kg TEG soil) resulted in a more intensive impact than ZY (118.52 kg TEG soil). The main contribution to emissions was electricity application during both pyrolysis and catalysts manufacturing. Table 7 depicts the number and value of emitting substances to the air, water, and soil from the pyrolysis scenarios. Based on the results in the above table, 44.81% of all 366 emitted substances are emitted to the soil while the emissions to the water have the lowest share, 19.95%. The main heavy metals with ecotoxic effects on air and soil, including aluminum, copper, cadmium, arsenic, chromium, lead, mercury, and nickel, were presented in Table 8. In both pyrolysis scenarios emitted, aluminum has the highest pollution load on the soil. The main reasons were the consumption of electricity, sodium silicate, natural gas, nitrogen, phosphorus trichloride, sodium hydroxide, and sulfuric acid in the ZSM-5 pyrolysis process. In the case of using ZY in the pyrolysis, most of the aluminum was emitted from electricity, nitrogen, soda, and sodium hydroxide utilized in the process.

Table 7. The number and value of the substance's emission in two scenarios.

Compartment	Number of Substances	Pyrolysis Involved ZSM-5 (kg TEG Soil)	Pyrolysis Involved Zeolite Y (kg TEG Soil)
Soil	164	85.73	84.04
Air	129	39.90	34.47
Water	73	6.84×10^{-7}	1.30×10^{-7}

Table 8. Main heavy metals emitted to the air and soil in the catalytic pyrolysis scenarios.

Substance	Compartment	Pyrolysis Involved ZSM-5 (kg TEG Soil)	Pyrolysis Involved Zeolite Y (kg TEG Soil)
Aluminum	Air	17.07	14.71
Aluminum	Soil	33.34	32.41
Arsenic	Air	0.67	0.63
Cadmium	Air	0.36	0.33
Cadmium	Soil	0.16	0.16
Chromium	Air	0.83	0.65
Chromium	Soil	0.92	0.91
Chromium VI	Soil	8.96	8.96
Copper	Air	5.27	4.76
Copper	Soil	20.37	20.33
Lead	Air	0.50	0.44
Mercury	Air	2.76	2.77
Nickel	Air	2.11	1.94
Nickel	Soil	0.37	0.36
Zinc	Air	10.21	8.13
Zinc	Soil	21.40	20.71

3.2. Catalyst Impact on Life Cycle Assessment

In general, the results showed that catalyst production was the major contributor to environmental burdens that occurred during the pyrolysis process. In 2022, Gupta et al. revealed the same statement that resulted from a comparison between non-catalytic and catalytic pyrolysis [30]. Additionally, they confirmed that overall environmental load is affected by the types of catalysts due to environmental impacts caused during their production. Between the two catalysts used in this study, the higher magnitude of environmental impacts (three categories out of four selected impact categories) belonged to the ZSM-5 catalyst. The reason for this result was the production process of the ZSM-5 catalyst. Catalyst synthesis led to emissions highly dependent on energy and materials utilization in the various processes of manufacturing. For instance, TPAOH (tetrapropylammonium hydroxide) as an input to prepare ZSM-5 compound contributed to emissions by phosphorus trichloride, sodium hydroxide, ethylene, ammonia, energy, and syngas. As aforementioned, the ZSM-5 catalyst is a combination of a ZSM-5 compound with fillers and binders. For this purpose, silica gel (SiO_2) and kaolin as filler and binder are needed to convert the ZSM-5 component into a catalyst by using energy. Figure 6 shows the environmental loads resulting from the production of the ZSM-5 catalyst. The results showed that utilizing silica gel caused the highest value in all impact categories because its production included sulfuric acid, sodium silicate water, and electricity. This result correctly confirms the role of the ZSM-5 catalyst in raising the environmental impact categories in previous sections.

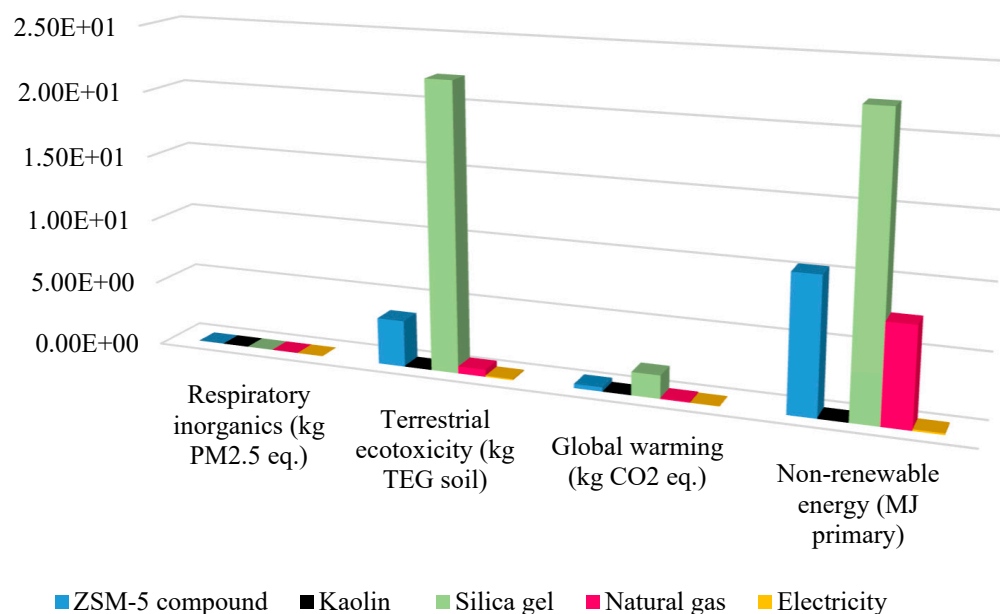


Figure 6. Contribution of the ZSM-5 catalyst production processes in selected impact categories.

4. Limitations of the Study

The limitation of the current study is connected to data uncertainty in two different classes. One of them is the efficiency of the processes in the laboratory, in which inventories are prepared based on the experiments, containing measurement uncertainty. RM conversion to the bioproducts by catalytic pyrolysis at a laboratory scale could be different from an industrial plant due to conducting the experiments in a well-controlled condition, which may not happen in a real-scale plant. Furthermore, a part of the uncertainty might arise from marginal technology or material alternates in a commercial plant. These differences cannot be accurately captured in an early-stage LCA.

The second limitation of this research was the lack of full data on the material and energy used for catalyst production. That could be a major reason to exclude the catalysts in previous LCA studies, which Benavides et al. revealed in a similar statement in 2017. The lack of data on catalyst manufacturing makes the researchers consider public information to estimate the catalyst components and production process, which causes uncertainty in the study.

5. Conclusions

The study focused on LCA modeling for two pyrolysis scenarios by using ZSM-5 and ZY catalysts in a laboratory scale. Four selected impact categories, including GW, NRE, RIOs, and TE, were estimated by life cycle assessment. Global warming, which was attributed mainly to the consumption of electricity was the highest among all categories in both scenarios.

The use of a more polluting catalyst (ZSM-5) provided the ZY catalytic scenario to be more environmentally friendly. Consumption of ZSM-5 in the pyrolysis led to higher environmental burdens in NRE, RIOs, and TE categories. The major driving factor of high value in the mentioned categories was the manufacturing process of the ZSM-5 catalyst by utilizing a big amount of natural gas and chemicals, such as phosphorus trichloride, sodium hydroxide, and sodium silicate. Therefore, switching to a renewable energy source for providing electricity and heat can mitigate the environmental impacts. Additionally, biochar, bio-oil, and syngas utilization as a replacement for energy needed in pyrolysis could be a solution to reduce impacts and, subsequently, production costs. Because of the absence of sufficient data on catalysts' manufacturing, this LCA analysis was carried out based on the literature and reports of catalyst production. Conducting more research and

sensitivity analysis through catalysts' loading, lifetimes, and specific composition could help to define LCA result uncertainties. To summarize, the result of this study would be beneficial for decision-makers and stakeholders in Europe regarding waste management and circular economy in the bioenergy industry.

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