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Valorisation of Plastic Waste *via* Catalytic Hydrocracking: a Technological Survey

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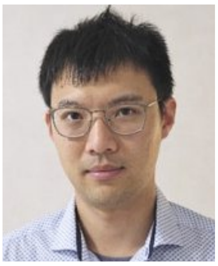
Abstract: In this work, we looked at the most recent advances in the field of plastics hydrocracking from a technology standpoint. A patent search was supplemented by a literature review to evaluate the development of the hydrocracking technology for plastics recycling. We observed that the patent and academic literature output rapidly increased over the past decade, highlighting the recent emergence of this technology to tackle the plastic waste problem. A majority of patents arise from academia, where plastics recycling is a growing research topic. Although innovations for plastics hydrocracking have been reported, most of them disclose the use of catalysts in batch reactors, whereas very few patents describe the process. Continuous operation at pilot scale will be essential to gather process data towards industrialisation and further assess the commercial viability of the plastics hydrocracking technology.

Keywords: Catalysis · Circularity · Hydrocracking · Patent survey · Plastics recycling

Dr. Felix Bobbink holds a PhD from the Ecole Polytechnique Fédérale de Lausanne in the field of sustainable catalysis (2018). After his PhD, he co-founded the company Plastogaz based on one of his inventions. His company develops catalysts for the transformation of plastic waste into value-added products. Dr. Bobbink published over 30 articles (h-index = 20) and is listed as an inventor in 2 patent applications.

Dr. Antoine van Muyden holds a PhD from the Ecole Polytechnique Fédérale de Lausanne in the field of catalysis for biomass valorisation (2020, 14 articles). After his PhD, he co-founded the company Plastogaz based on his invention. Dr. van Muyden leads the development of the catalytic hydrocracking process that he invented together with Dr. Bobbink and Dr. Lee



Dr. Wei-Tse Lee holds a PhD from the Ecole Polytechnique Fédérale de Lausanne in the field of catalysis for plastic transformation. Key results for catalytic hydrocracking developed during his PhD led to publications and a patent invention. He co-founded the company Plastogaz with Dr. Bobbink and Dr. van Muyden to further mature the hydrocracking technology toward real-world applications.

Dr. Christopher Hunston holds a PhD from the Ecole Polytechnique Fédérale de Lausanne in the field of heterogeneous catalysis for biomass conversion, performed at the Paul Scherrer Institute (PSI). Dr. Hunston joined Plastogaz in 2022 as a research engineer, working on the development and scale up of the catalytic hydrocracking process.

1. Introduction

The global plastic waste issue is a growing environmental concern, with over 450 million tons of plastic produced in 2019, of which more than 350 Mt ended up as waste. Approximately 50% of the waste is landfilled and 20% incinerated, and only 9% is recycled.[1] A significant fraction (21%) is poorly managed and eventually ends up in the environment. These alarming numbers show that there is an urgent need, as well as substantial potential for recycling solutions. Considerable efforts have been put in the development and deployment of advanced recycling technologies that can deconstruct plastic waste into its original building blocks without facing the limitations posed by degradation and contamination often associated with traditional recycling methods.[2]

Advanced recycling technologies such as pyrolysis and hydrocracking have enabled recycling plastics that were previously considered non-recyclable. These technologies offer multiple advantages such as reducing greenhouse gas emissions compared to incineration,[2,3] and contributing to the reduction of fossil fuel consumption by utilising plastic waste as a substitute for petroleum-derived feedstocks and products, leading to improved circularity in the plastics industry.

Pyrolysis, which is a thermal degradation process, has made remarkable strides in the field of plastics recycling. By subjecting plastic waste to high temperatures in an inert atmosphere, pyrolysis breaks down complex polymers into smaller molecules, including liquid (pyrolysis oil) and gaseous products. Pyrolysis oil must then be further refined to be used as feedstock for the production of new plastics or other valuable chemicals. Catalytic pyrolysis can help in decreasing downstream purification steps and has thus also seen an increase in popularity over the past decade for the recycling of waste plastics.[4] Pyrolysis offers a versatile solution for plastics that are difficult to recycle through tradition-

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al methods due to contamination, mixed materials, or complex structures. However, the critical challenges remain the safe removal of halogens and a better control of the pyrolysis oil quality.[5] Hydrocracking, on the other hand, uses hydrogen and a catalyst to deconstruct plastic waste. This catalytic process involves H_2 pressure and high temperature to break down the molecular bonds of plastic polymers. This results in the production of lighter hydrocarbon compounds such as naphtha, that can be directly fed to a steam cracker to produce ethylene, an essential building block in the plastics industry. Inserting hydrogen at the beginning of the reaction is beneficial, as there is no need for downstream refining thanks to a good reaction and product control, as illustrated in Fig. 1. However, plastics hydrocracking is more sensitive to impurities due to the presence of a catalyst. A recent LCA (life cycle assessment) study comparing pyrolysis and hydrocracking for the production of fuels from waste plastics found that hydrocracking is better in terms of environmental impact in 10 of the 11 impact categories.[6]

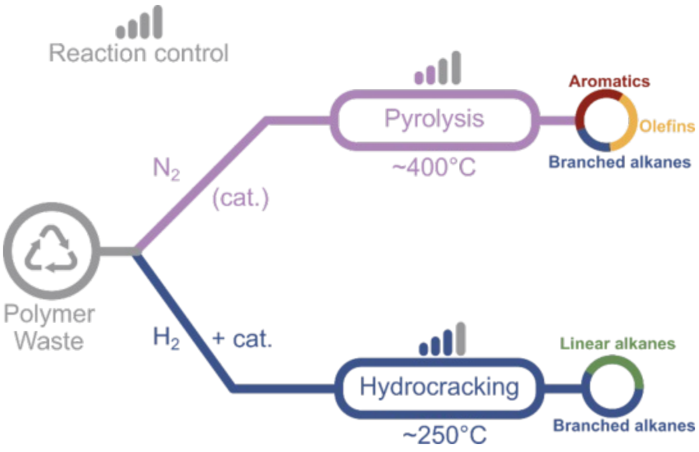

Fig. 1. Schematic description of the pyrolysis and hydrocracking processes.

In this contribution, we examine the state-of-the-art in the field of plastic waste hydrocracking, with a particular focus on patents. We explore the various processes that have been developed for converting plastic waste into useful products. Our goal is to provide a comprehensive overview of the current state of the plastic hydrocracking technology and identify areas for future research and development.

2. Literature and Patent Search Results

To follow the recent academic and industrial innovations of the hydrocracking technology for plastics recycling, we performed a patent search on the topic: 'plastic material conversion into liquids using hydrogen and a catalyst', as well as a literature survey. Our findings are presented and discussed below. The detailed search strategy can be found in the Methodology section.

The search yielded 64 patents, of which 35 refer to catalytic hydrocracking of plastic waste (with or without solvent). 13 patents describe pyrolysis processes, with a 8 further patents reporting advances on pyrolysis oil upgrading, categorised as hydrotreatment. Fig. 2 shows the timeline of the published patents. It is interesting to note that the first record was published in 1929, describing coal liquefaction (*i.e.* hydrocracking). Other than this pioneering account, most patents in the field of hydrocracking were published in the last two decades, with a significant proportion published in the last four years. Although hydrocracking is an established technology in the petroleum refining industry, it is relatively new in the field of plastic waste valorisation.

The novelty of the plastics hydrocracking technology also indicates that, to the best of our knowledge, there are no large-scale demonstration or commercial plants operating yet. The technology is at the prototype or pilot scale, representing a technical readiness level (TRL) of 4-6. On the contrary, pyrolysis is more advanced with several demonstration or commercial plants running and can be considered at a TRL of 6-9.[7] Although no demonstration plants have been built yet for plastics hydrocracking, there are strong signs that the technology will develop rapidly. Many academic institutions have been publishing on plastics hydrocracking recently, as emphasised by the significant increase over the past four years in Fig. 3. Although these figures are rapidly rising, they remain well behind the research output for pyrolysis, by at least one order of magnitude. Research groups based in the USA are the primary contributors to the field of plastics hydrocracking, accounting for nearly 30% of the published research papers. The main share of these publications originates from cross-institutional collaborations through research consortia such as iCOUP,[8] CPI[9] and BOTTLE.[10] The main academic research discussed herein is summarised in Table 1.

Prof. Anderson's early work on catalytic coal liquefaction at the University of Utah showed that liquids could be produced in high yields with the use of a catalyst.[11] Keeping the same approach, he reported the deconstruction of virgin and post-consumer PE into liquid fractions with a $TiCl_3$ or H-ZSM-5 catalyst.[12,13]

At the University of Manchester, Prof. Garforth followed up on these early reports, doing research on polymer waste recycling since the 1990s. He investigated fluidised bed cracking[14] before moving on to hydrocracking in the 2000s,[15] generating several publications[14-19] and patents.[20, 21]

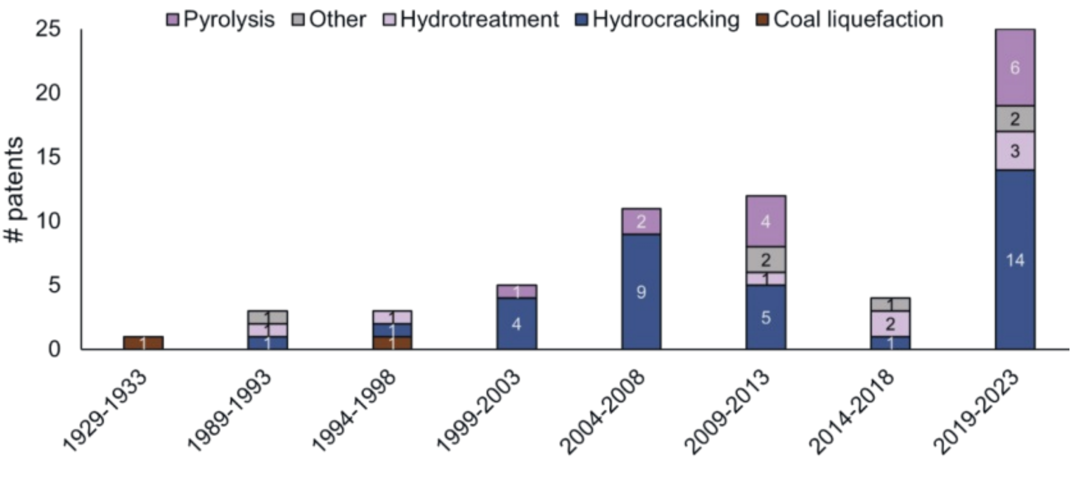

Fig. 2. Timeline of patent publication separated by technology.

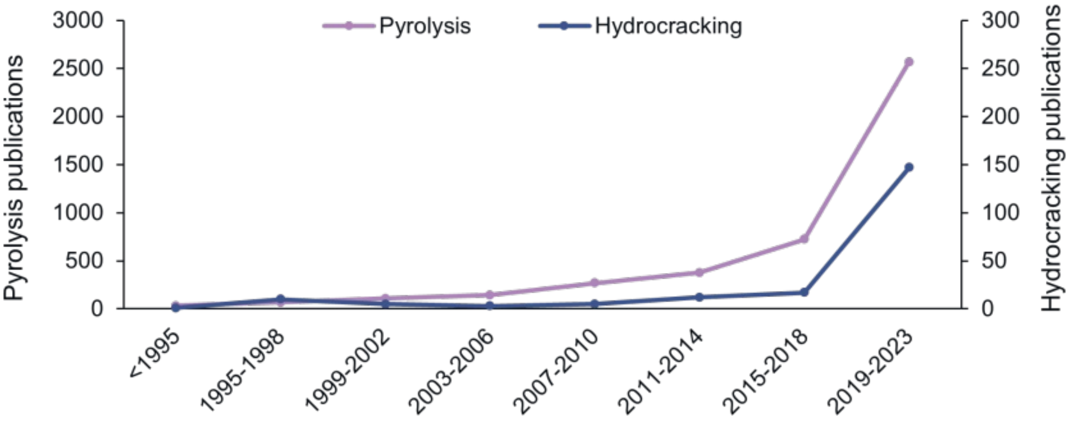

Fig. 3. Publication output for plastic hydrocracking and pyrolysis – data from Web of Science.

In the USA, BOTTLE's goal is to provide catalytic and biocatalytic technologies to create value from waste plastics, bringing chemical and biological technologies together to valorise plastic waste.[22] They also work on hydrocracking catalysts, with descriptions of Ru-supported catalysts for the production of liquid alkanes,[23,24] as well as selective propane production over Co/ZSM-5, together with methane suppression.[25] CPI, based at the University of Delaware, also focus on catalyst development for polyolefin hydrocracking, with several reports on Pt,[26-28] Ru[29,30] and Ni[31,32] catalysts. They have also applied for two patents in the field, the first one based on a hydrocracking process to convert PP-based waste into lubricants over a Ru/TiO_2 catalyst.[33] The second one covers hydrocracking catalysts.[34] The iCOUP consortium, led by the Ames Laboratory, develops technologies to upcycle plastic waste, with a strong focus on hydrogenolysis catalysts. For example, they use Pt nanoparticles on strontium titanate (Pt/$SrTiO_3$) to valorise plastic waste.[35-38] They also take advantage of mesoporous architectures in silica to synthesise Pt catalysts that are selective to waxes during polyolefin hydrogenolysis.[39,40] Longer hydrocarbon chains are targeted, such as lubricants,[41]

Table 1. Summary of academic publications on direct plastics hydrocracking

Institution or Consortium	Feed	Conditions[a]	Catalyst (% cat/feed)	Product yields
University of Utah	Coal	300 °C, 135 bar, 3h	$ZnCl_2$ (n.a.)	71wt% liquids[11]
	PE	420 °C, 69 bar, 1h	$TiCl_3$ (5%)	46wt% C_{5-27}[12]
	PE	420 °C, 69 bar, 1h	H-ZSM-5 (4%)	38wt% C_{5-27}[12]
University of Manchester	PE	310 °C, 55 bar, 0.5h	1%Pt/USY (10%)	56wt% C_{5-12}[15]
	Mix	330 °C, 20 bar, 0.5h	5%/Beta (10%)	22wt% liquids[18]
	PE	330 °C, 20 bar, 1h	5%NiS/Beta (10%)	38wt% liquids[18]
BOTTLE	PP	225 °C, 20 bar, 16h	5%Ru/C (14%)	68wt% C_{5-32}[24]
	PE	200 °C, 30 bar, 16h	5%Ru/FAU (7%)	67wt% C_{5-28}[23]
	PE	250 °C, 40 bar, 20h	5%Co/ZSM-5 (7%)	97wt% C_{2-4}[25]
CPI	PE	250 °C, 30 bar, 2h	0.5%Pt/WO_3/ZrO_2 (10%)	30wt% C_{4-20}[27]
	PE	250 °C, 30 bar, 2h	0.5%Pt/WZ (5%) + HY (5%)	92wt% C_{5-16}[28]
	PE	250 °C, 50 bar, 2h	5%Ru-MoZr (2.5%)	65wt% C_{4-35}[29]
	PE	300 °C, 30 bar, 2h	NiAl (10%)	47wt% C_{2-35}[32]
	PE	300 °C, 30 bar, 2h	15%Ni/SiO_2 (10%)	65wt% C_{6-35}[31]
iCOUP	PP	300 °C, 12 bar, 24h	1wt%Pt/$SrTiO_3$ (10%)	95wt% liq./wax[35]
	PE	300 °C, 12 bar, 96h	11wt%Pt/ $SrTiO_3$ (2%)	97wt% liq./wax[38]
	PE	300 °C, 9 bar, 20h	mSiO_2/0.1%Pt/SiO_2 (1%)	33wt% liq./wax[39]
	PP	250 °C, 14 bar, 6h	0.6%Pt/SiO_2 (1%)	8wt% C_{9-35}[40]
	PE	280 °C, no H_2, 24h	1.5%Pt/γ-Al_2O_3 (100%)	80wt% liq./wax[42]
EPFL	PE	300 °C, 50 bar, 3h	2.5%Ru/HY (6%)	96wt% gases[44]
	PS	350 °C, 50 bar, 4h	2.5%Ru/HY (6%)	98wt% gases[44]
	Mix	350 °C, 50 bar, 4h	2.5%Ru/HY (6%)	99wt% gases[44]
	PE	375 °C, 45 bar, 16h	2.5%Ni/ZSM-5 (6%)	38wt% C_{5-20}[45]
	PE	375 °C, 45 bar, 16h	2.5%Co/ZSM-5 (6%)	40wt% C_{5-20}[45]

[a]all entries were performed in batch setups.

waxes and alkylaromatics,[42,43] because of their higher market value.

Prof Dyson's group at EPFL has also investigated plastic waste hydrocracking catalysts for the generation of grid compatible natural gas at first,[44] before targeting the production of saturated liquid hydrocarbons.[45] A patent has also been filed for these innovations.[46]

3. Selected Patents

The most representative patents from the plastics hydrocracking search are summarised in Table 2 and described below. It is important to note that the patent search yields all published entries, regardless of their status (application, granted, cancelled, rejected, *etc.*).

The University of Manchester has two granted patents on plastic waste hydrocracking, the first one describing a continuous process for plastic waste recycling.[20] They claim the continuous introduction of plastic waste feedstock into a dechlorination chamber to treat Cl-contaminated feedstocks if needed, before feeding the dechlorinated feedstock together with H_2 into a hydrocracking reactor containing a zeolite catalyst. Reactive extrusion is also described, well-mixed polymer feed and catalyst are fed to an extruder containing a hydrogen addition port and a vacuum port. The second one focuses on a synthesis procedure for modified zeolites, used as catalysts in the hydrocracking process.[21] CPE Lyon had a patent granted in 1999 on polymer degradation into hydrocarbons over a Zr-based catalyst in the presence or absence of solvent.[47] The main focus of the patent was on the catalyst; no description of the process is provided. The patent was discontinued in 2005 in the USA and 2008 in Europe.

Several patents arose from the iCOUP research consortium with a particular focus on hydrocracking catalysts, as described from academic publications in the previous section. A Pt-based catalyst with a silica core and a mesoporous silica shell is described in one patent,[48] a Pt/$SrTiO_3$ catalyst is described in other patents,[49,50] in which a particular focus is set on the synthesis and use of these catalysts for lubricant production. Regarding the hydrocracking process, a global description is provided in a more recent patent application.[51] They disclose an integrated process for the production of lubricants from waste plastics, which are first melted in a hydrocracking reactor in the presence of their Pt/$SrTiO_3$ catalyst and under a flow of H_2. They adapt the temperature, pressure, and reaction time to convert most of the feedstock into liquid lubricants. The product-catalyst slurry is then fed to a distillation column, where the lubricants are purified, the catalyst is washed and recycled into the reactor. Lighter hydrocarbon fractions, such as naphtha, can also be recovered.

EPFL has one patent on the conversion of plastics to methane or liquid hydrocarbons.[46] The invention describes the continuous transformation of waste plastics to light hydrocarbons (naphtha) in the presence of a zeolite-based catalyst and hydrogen.

Tohoku University in Japan provide a description of a catalyst synthesis procedure for a hydrocracking process, contacting metal-oxide-supported Ru catalysts with polyolefins having melting points below 250 °C, at temperatures above 100 °C.[52] They do not provide additional information on the process. In China, Dalian University of Technology have a patent on a tandem hydrogenolysis/alkylation process to convert waste plastics and naphthenic solvents to waxes and long-chain cycloalkanes over modified Nb_2O_5 or ZSM-5 catalyst.[53]

Looking at innovations from the industry, ENFC Co., a South Korean company, filed a patent in 2012 for a continuous hydrocracking process,[54] extruding waste plastics into a catalytic decomposition reactor at 150-450 °C. The reactor is equipped with an H_2 diffuser for homogeneous gas distribution. However, there is no mention of pressure, suggesting that the reaction was performed under atmospheric conditions. Although the innovation looked promising, it was withdrawn in 2015. An invention by Honeywell in 2015 described a batch reactor for plastics hydrocracking to waxes with broad claims, such as the use of hydrogenation catalysts and a pressure above 0.5 bar.[55] The patent was discontinued in the USA and in Europe. LyondellBasell have filed a patent on hydrodepolymerisation of plastic waste.[56] However, there is no mention of a continuous process, and in all examples provided the polymer feedstock is blended with white oil or pyrolysis oil. SABIC have a patent on a catalytic process to treat

Table 2. Summary of hydrocracking patents

Institution or Company	**Patent**	**Conditions**	**Catalyst**	**Status**[a]	**Year**
CPE Lyon	EP0840771B1[47]	25–300 °C, 0.01–500 bar	Zr/SiAl	Cancelled	2008
Uni of Manchester	EP2649121B1[20] EP2437886B1[21]	220 °C 35–60 bar	Zeolite Pt/USY	Granted Granted	2017 2019
iCOUP	US2022111356A1[48] US11499110B2[49] US11596935B2[50] US2023109450A1[51]	150–450 °C, 1–69 bar 150–350 °C, 7–14 bar 100–500 °C, 1–68 bar 200–450 °C, 12–35 bar	Pt/SiO_2 Pt/$SrTiO_3$ Pt/$SrTiO_3$ Pt/$SrTiO_3$	App. Granted Granted App.	2022 2022 2023 2023
EPFL	EP3884013A1[46]	140–300 °C, 40–200 bar	Ru/Y	App.	2021
Dalian Uni of Technology	CN112779045B[53]	200–400 °C, 5–50 bar	Rh/Nb_2O_5, Pt/ZSM-5	Granted	2021
Tohoku Uni	JP7304615B2[52]	>100 °C	Ru/MO	Granted	2023
ENFC Co.	US2013136665A1[54]	250–450 °C	Zeolite	Cancelled	2015
Honeywell	US2015247096A1[55]	300–500 °C, >0.5 bar	Zeolite	Cancelled	2019
LyondellBasell	EP4032963A1[56]	300–450 °C, 150–300 bar	Ni/SiAl	App.	2022
SABIC	US10717936B2[57]	n.a.	Zeolite	Granted	2020
Beijing Petrochem	CN108456328B[58]	150–300 °C, 40–70 bar	Sn or Ti / ZSM-5 or Y	Granted	2020

[a] App.: Patent is in the application procedure

mixed plastics, the main invention being based on pyrolysis with a zeolite catalyst.[57] Their claims also include the use of hydrogen, which would allow versatility between a pyrolysis and hydrocracking process. Again, there is no mention of pressure in their invention. Beijing Institute of Petrochemical Technology, have a granted patent on plastic waste conversion in the presence of a zeolite (HZSM-5 or HY) modified by Sn or Ti, but with no mention of a continuous process.[58]

4. Summary and Outlook

The academic and patent publication data presented in this contribution shows that there is a rise in plastics hydrocracking output. In academia, more and more institutions dedicate research to hydrocracking with the goal to recycle plastics, leading to increased patent applications in the field. However, most of the innovations relate to catalyst formulations for a better reaction control, and less to the process itself. This is evidenced by the experiments performed for these publications, all of them are done in batch setups. Batch reactors remain good instruments to screen catalysts and feedstocks. Yet, process data such as conversion and selectivity will significantly vary when switching to a continuous process, which is essential towards the industrialisation of such processes. The same limitations apply to patents from industry. Indeed, catalyst formulation is often explained, whereas the process description often remains vague.

The development of a continuous plastics hydrocracking process will most probably arise at the intersection of academia and industry, with start-ups trying to build on a technology discovered in an academic lab. For example, at Plastogaz, an EPFL spin-off,[59] we work on the development and scale up of plastics hydrocracking, assessing the technology on a continuous pilot unit capable of treating up to 5 kg/h of plastic waste. This will help us gain fundamental understanding of the continuous process and generate the data required for the design of a plastics hydrocracking demonstration plant.

Critical aspects in the development of this advanced recycling technology will be gathering several hours on stream of continuous hydrocracking to assess catalyst lifetime. The evaluation of different sources/grades of post-consumer plastic waste will also be essential to have a realistic projection of the streams to target and the pre-processing required. The right balance has to be found between versatility towards feedstock, catalyst lifetime and product specifications. The feedstock aspect will be crucial, as better product design will improve the recyclability of packaging (*e.g.* mono- *vs.* multi-layered materials). Although before mentioning recycling, it is important to first reduce the consumption of plastic materials, second to design materials for reuse, and eventually recycle. These pillars of recycling will help in achieving our goals towards a circular, more sustainable economy.

5. Methodology

A patent search was conducted on the topic of plastic material conversion into liquids using hydrogen and a catalyst. The search was conducted together with the Swiss Federal Institute of Intellectual Property (IPI). The keywords search structure was the following:

OR PLASTIC+, POLYMER+, POLY_ETHYLEN+, POLY_PROPYLEN+, POLY_STYREN+, POLY_OLEFIN+, POLY_ESTER+, POLY_VINYL_CHLORID+, CARBONACEOUS
OR CONVER+, RECYCL+, UP_CYCLING+
OR HYDROGEN, H_2
OR PRESSURE+, MPA, BAR, PA
OR CATALY+, ZEOLITE, METAL+, OXIDE+
HYDROCRACK+
OR FUEL, METHANE, HYDROCARBON, NAPHTHA, +ALKAN+

Additionally, the following patent classifications were targeted:

C08J11/16, C10G, C10G1/00, C10G1/06, C10G1/10 in the International Patent Classification (IPC) system, and Y02W30/62 in the Cooperative Patent Classification (CPC) system.

The academic literature survey was conducted through Web of Science with the following search keywords for hydrocracking:

plastic* OR polymer OR polyolefin*
ANDhydrogenolysis OR hydrocracking
ANDwaste OR post-consumer

For pyrolysis, 'hydrogenolysis OR hydrocracking' was replaced by 'pyrolysis'

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