Chemical Recycling

An analysis of research activity based on a review of patent literature

Dec 2022 – Hilde Schoofs, IP Expert essenscia

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1 Introduction

With the European Green Deal and the ambition of Europe to be a world leader in the circular economy in mind, it is worth looking at the potential that Chemical Recycling (CR) holds. Relocation of essential resources and circular plastic strategies pledge to collect more waste and to recycle it, instead of incineration, export or landfilling. Chemical recycling technologies are on the rise.

This report provides an analysis of the innovative activities in the field of Chemical Recycling by a review of the patent activity. Patent data sets were evaluated using "PatentInspiration" whereby insight was obtained with respect to activity over time, main players and technologies used. Patent value analysis was used to obtain the key innovations in the respective data sets.

This report does not cover accounting and traceability for chemical recycling and subsequent recycled content, for which the mass balance allocation method is the most suitable tool.

"Chemical Recycling" breaks down plastics into smaller polymers, oligomers, monomers or feedstocks that can then be used as virgin like materials for the making of new materials, reducing thereby the use of primary resources, among which fossils fuels. More information on what Chemical Recycling is and its subcategories on the next page.

In this report the term "plastic" is used in its broadest sense to refer to a wide range of primarily synthetic or semi-synthetic materials that use polymers as a main ingredient. We looked at plastics and plastic containing materials as a source for waste recovery via CR and have included materials like plastic, rubber, textiles, carpets, photovoltaic cells, electrical and electronic equipment etc.

2 Chemical Recycling in a nutshell

Circular economy of plastics may be achieved in various ways, after the end of use of a product, (i) if the product has been designed to be used once, for a short or long period of time, or (ii) if it has been designed for repeated use. Depending on the use and the consumer type, a product reaching its end of use may reach its end of life, and may be discarded as waste.

Polymers and plastics are versatile materials which need suitable technologies to be refitted in new materials, among which chemical recycling processes.

Commercially applied chemical recycling technologies according to the ECHA 2021 report on Chemical Recycling (CR) include[1]:

- **Chemolysis**: Process that breaks down polymers into monomers, oligomers and other chemical substances. Virgin-grade quality monomers can be achieved with the necessary purification techniques. There are different types of chemical depolymerization depending on the chemical agent being used. Alcoholysis, aminolysis, ammonolysis, glycolysis, hydrolysis, methanolysis, solvolysis etc are a few examples[2].

- **Pyrolysis** (also known as conventional pyrolysis or thermal cracking): One of most widely known chemical recycling techniques according to literature. Thermal cracking under the absence of oxygen breaks down the polymer macrostructure and gives monomers and fuel-type products.

- **Catalytic cracking**: another form of pyrolysis that involves the use of catalysts. This process is limited to pure polymers such as polyolefins and polystyrene. Examples of catalysts used include zeolites and silica-alumina.

- **Gasification**: another thermal treatment but one that involves partial oxidation of plastic waste. This process leads to detailed polymer breakdown and leads to a mixture of hydrocarbons and syngas. Syngas can be used as alternative energy source or for the production of simple molecules like ethanol, methanol, ethylene.

Developing chemical recycling techniques according to this report inlude[1]:

- **Hydrocracking** (also known as hydrogenation): a pyrolysis technique wherein hydrogen is added to the pyrolysis process.

- **Pyrolysis with in-line reforming**: yet another pyrolysis technique, developed to obtain tar-free hydrogen from plastic waste. After the pyrolysis process, the pyrolysis product is reformed in another reactor.

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[1] Chemical Recycling of Polymeric Materials from Waste in the Circular Economy, Final Report prepared for the European Chemicals Agency, August 2021

[2] Ragaert et al., 2017. Mechanical & chemical recycling of solid plastic waste. *Waste Management, 69,* 24-58

- **Plasma gasification**: any thermal process where the necessary heat is produced by thermal plasma, for instance using plasma torches[3].

- **Micro-wave assisted pyrolysis**: dielectric materials or absorbents (like activated carbon, silicon dioxide or graphene) are added that absorb microwave energy and allow to reach the temperature needed:

- **Plasma pyrolysis**: a technology that makes use of a combination of plasma and conventional pyrolysis techniques to completely breakdown monomers to produce syngas, composed mainly of CO, H_2 and small amounts of higher hydrocarbons.

Of the developing chemical recycling techniques mentioned herein, plasma pyrolysis and microwave-assisted pyrolysis would have been used at laboratory scale only thus far, whereas the other techniques mentioned would already be operated at (limited) commercial scale.

We further wished to explore **Enzymatic/Biologic depolymerization**, or the use of enzymatic activity to break down polymer chains to monomers or digestion.

The above information was used to break down the larger patent data set into more narrow data sets to gain more insights in the major chemical recycling technologies described in patent literature, tendencies, and players.

Note: Solvent purification is included in the general data set, as it was not easy to filter out patents that relate to removal/purification of polymers without including a polymer degradation step. Solvent purification was excluded from the more detailed data sets, unless indicated otherwise.

[3] Al Rayaan, 2021. Recent Advancements of thermochemical conversion of plastic waste to biofuel – A review. *Cleaner Engineering and Technology*, 100062

3 Search strategy and data sets

In order to explore the patent landscape of Chemical Recycling (CR), a number of different patent data sets were created using "PatentInspiration" and "Orbit's FamPat".

First, the following search strings were used to search in title, abstract and claims (ti, ab, clms):

- Chemical recycling and plastic/polymer OR chemical recover+ AND recover+ of plastic/polymer (ti, ab, clms)
- Plastic/polymer recycle+ and techniques (like gasification, pyrolysis, depolymerisation, hydrolysis etc) (ti, ab, clms)
- Contamina+/impur/pollut+ near techniques AND waste/end-of-life/post-consumer (full text) + codes for solid waste management (plastics/rubber/polymer recycling)

Second, we focused on specific types of waste materials in combination with commonly used CR techniques like gasification, pyrolysis, depolymerisation, hydrolysis etc. Here we looked at full text. In the below the term "codes" refers to the commonly used CR techniques (cfr point 2 above)

- PVC near techniques (full-text) and codes for technologies for solid waste management / recovery of plastics/polymers
- Polyurethane near techniques (full-text) and codes
- Polypropylene near techniques (full-text) and codes
- Polyolefins / ?DPE near techniques (full-text) and codes
- Rubber near techniques (full-text) and codes
- PET near techniques (full-text) and codes
- Polystyrene near techniques (full-text) and codes

This led to a large data set of over 5,300 patent families over the 20 years. This data set was then further cleaned out to remove irrelevant patents. A final data set of around 3,750 patent families was maintained.

More focused data sets were created to explore Chemolysis, Pyrolysis, Catalytic Cracking and Gasification as well as Means for Biological/Enzymatic Degradation. These were obtained as follows:

Chemolysis: a subset of patents related to Chemolysis was created by looking for patents that have ["chemical*" or "chemoly" or "solvoly*" or "glycoly*" or "hydrolys*" or "alcoholy*" or "methanoly*" or "aminoly*" or "ammonoly"] in title, abstract or claims (ti, ab, clms). A subset of 680 patents relating to Chemolysis was created.

Pyrolysis: a subset of patents related to Pyrolysis was created by looking for patents that have ["pyrolys*" or "pyrolyt*" or "thermal crack*" or "cracked thermal*" or "hydropyrolys" or "hydrocrack*" or "hydrogenation" or "micro0wave" or "plasma pyrolys" or "in-line reform"] in title, abstract or claims (ti, ab, clms). A subset of 830 patents relating to Pyrolysis was created.

Catalytic cracking: a subset of patents related to Catalytic Cracking was created by looking for patents that have ["catalytic crack" or "cracked catalytic*" or "catalytically crack*" or "catalytic pyrolys*" or "catalytically pyrol*"] in title, abstract or claims (ti, ab, clms). A subset of 71 patents relating to Catalytic Cracking was created.

Gasification: a subset of patents related to Gasification was created by looking for patents that have ["gasification" or "gasify" or "gasifie*" or "plasma gasification" or "syngas"] in title, abstract or claims (ti, ab, clms). A subset of 187 patents relating to Gasification was created.

Text pattern analysis in title, abstract and claims (ti, ab, clms) was used to gain more insights into the Waste type being treated, the Polymer type being treated and the Means used for Biological/Enzymatic depolymerization:

Waste type: "battery" or "batterie*" or "fuel cell*" or "tire" or "tyre*" or "demolition waste" or "fibre*" or "fiber*" or "fibrou*" or "textile" or "carpet*" or "packaging" or "plastic*" or "municipal waste" or "industrial waste" or "waste*" or "concrete" or "glass" or "paper*" or "electronic" or "electric" or "circuit board*" or "photovoltaic cell*" or "biomass"

Polymers involved: "polyamide" or "polyvinyl" or "poly-vinyl" or "polystyrene" or "polylactic" or "polyolefin" or "polyethylene" or "polypropylene" or "polyester" or "PET" or "thermoplastic" or "rubber" or "polyurethane". Another approach is to search for "poly*" via text pattern analysis

Means for Biological/Enzymatic depolymerization: "hydrolyz*" or "hydrolase*" or "enzyme*" or "petase*" or "esterase*" or "micro-organism*" or "microorganism*" or "consorti*" or "strain*" or "bacteri*" or "bacill*" or "cellulos*" or "deligni*" or "biodegrad*" or "biomass*"

4 Overall analysis of patents in the field of CR

The large data set as set out above (of about 3,750 patent families) was used to make an overview analysis of the field of Chemical Recycling (CR).

4.1 CR Patent Activity

Patent activity in CR took on near 2005. In 2007 we see a first peak in activity, with Japan as one of the major filing countries around that time. As of 2018, patent activity in the CR domain is steadily growing worldwide [Figure 1]. In 2022 we have already more filings than the year before and it is expected that another 100-200 patent publications will follow before the year's end.

Figures 2A&B show you where most of the patents are being filed. This graph provides information on the strategies of the actors in the sector. They either file in their home country or they file where production facilities and key markets are. This graph is based on patent publication numbers and is dependent on the practices of the national/regional patent Offices (when they publish/republish). We seem to have key players in China, Japan, the US, Korea and Europe. Figure 2A shows the top 50, Figure 2B the top 10. Germany, France and Italy figure amongst the top 10.

Figure 3 shows with which Patent Offices patents are being filed. This figure indicates that in Japan activity is declining, after the peak in activity around 2017. In China and other countries patent activity is increasing as of 2018.

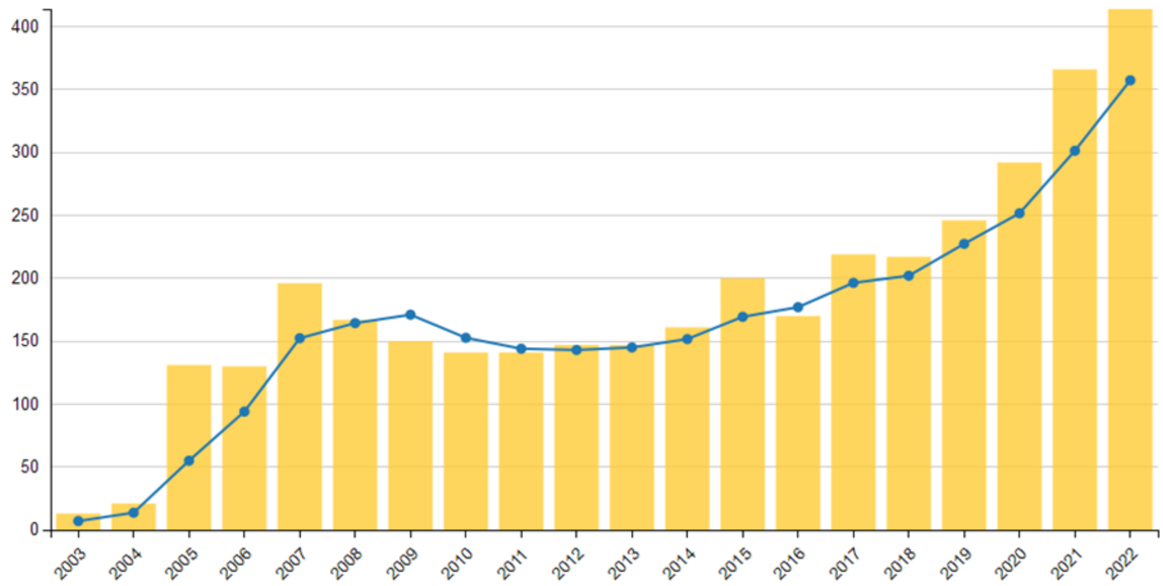

Figure 1: Number of patent publications by publication year (period 2003-2022).

Left: Figure 2A: Top 50 publication countries (without EP and WO)
Right: Figure 2B: Top 10 publication countries (without EP and WO)

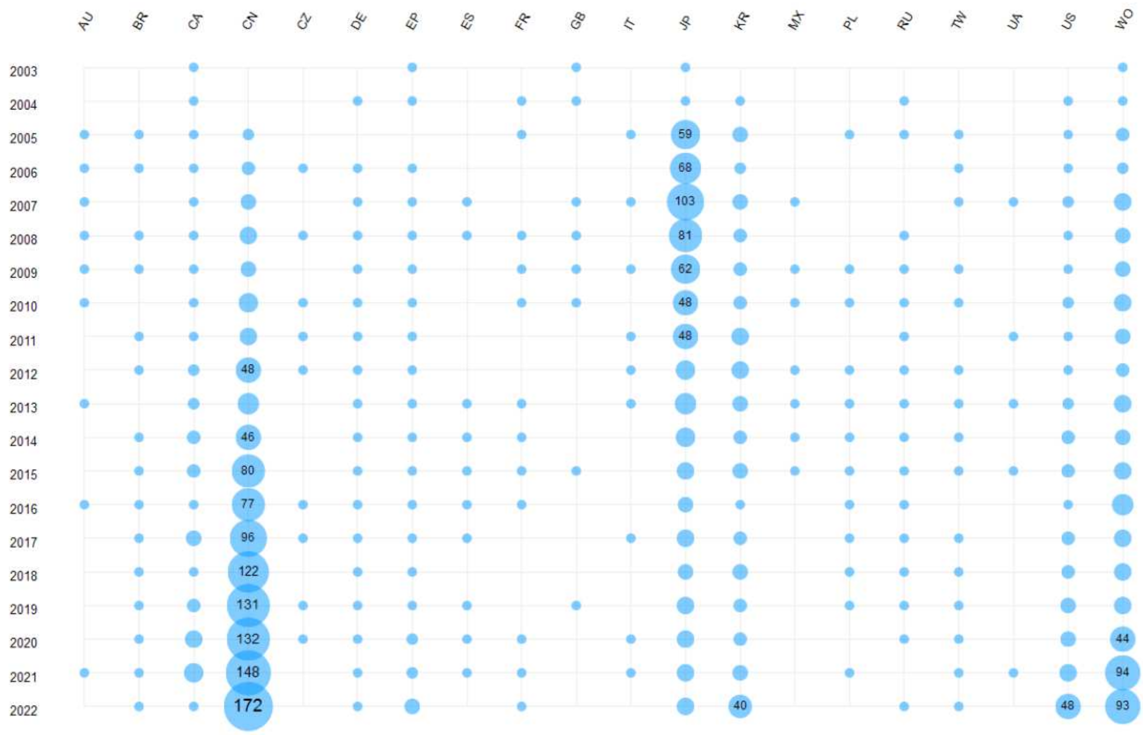

Figure 3: Where are most of the CR patents filed
(with which national or regional Patent Office).
EP stands for filings with the EPO[4], WO stands for filings with WIPO[4]

[4] EPO: European Patent Office, WIPO: World Intellectual Property Office

Table 1 below lists the number of patent publications by applicant country. These numbers give an indication of where the top applicants (companies + academics) are based (in which country/region).

Applicant Country	No of patent publications	% (total =3,750)
China	1212	32
Japan	881	24
Europe (total of)	555	15
US	428	11
Korea	344	9
Canada	66	2
Russia	50	1,5

Table 1: Patent publications by applicant country

We see that Europe takes the third place. When we look at individual countries (instead of at Europe as a whole), we see that Germany, France and Italy figure in the top 10 [Figure 2B]. Belgium figures in the top 15 of active players.

Overall, the CR patent landscape is scattered. There are over 1,000 applicants (filing entities) of which most have filed 1 patent only. Chinese applicants mainly file utility models (or short patents[5]) and most do not file outside of China. Also most Korean applicants file domestically only.

4.2 Top 10 applicants

In the top 10 of filing entities (companies + academics) figure many Japanese & US companies, with 1 US company as true top filer: EASTMAN CHEMICALS CO [Table 2].

In the top 10 we have 1 French company (CARBIOS) and 1 Belgian company (SOLVAY). No academic entities figure in the top 10, only companies. Table 2 below lists on what the European companies have filed their patents on.

[5] Utility models have a lifetime of 10 years only (and not 20 years) and have a lower standard of inventive step

Top	Applicant	Country	No. of patent families	Focus is on
1	Eastman Chemicals Co	US	70	
2	Matsushita Electric Works Ltd	JP	31	
3	Carbios	**FR**	26	Biological/Enzymatic degradation
4	Procter and Gamble	US	26	
5	JFE Steel Corp	US	25	
6	Toshiba KK	JP	25	
7	Sharp KK	JP	22	
8	Solvay	**BE**	21	Thermal & Catalytic cracking, Solvolysis, Solvent purification
9	Taiheyo Cement Corp	JP	19	
10	Toray Industries	JP	18	

Table 2: Top 10 applicants (companies + academics)

Top 10 Academic entities have residence in China, Japan, the US, Korea and Europe (same countries as for the top 10 Companies):

- Korea: KOREA RES INST CHEM TECH [KR], KOREA ENERGY RESEARCH INST [KR],
- China: UNIV CENTRAL SOUTH [CN], UNIV JIANGNAN [CN], INST PROCESS ENG CAS [CN], UNIV NANJING, UNIV QINGDAO TECHNOLOGICAL [CN],
- Europe: CENTRE NAT RECH SCIENT [FR], FROUNHOFER GES FORSCHUNG [DE]
- Japan: UNIV SHIZUOKA [JP]
- the US: UNIV CALIFORNIA [US]

Table 3 below gives an idea of the Belgian CR players and their domain of expertise. The list was extracted from the general data set of 3,75 patents studied and covering the past 20 years.

Table 3: List of CR patents of the past 20 years with contribution of Belgian inventors

Publication No.	Title	Standard applicant	Espacenet URL	Priority year
EP1186618A1	Controlled rheology polypropylene heterophasic copolymers	ATOFINA RES	https://worldwide.espacenet.com/patent/search?q=EP1186618A1	2000
CA2972505A1	Process and system for treating municipal solid waste materials and producing multiple products	DECIDE NV	https://worldwide.espacenet.com/patent/search?q=CA2972505A1	2017
WO2010118955A1	Chemical recycling of pla by alcoholysis	GALACTIC SA	https://worldwide.espacenet.com/patent/search?q=WO2010118955A1	2009
WO2010118954A1	Chemical recycling of pla by hydrolysis	GALACTIC SA	https://worldwide.espacenet.com/patent/search?q=WO2010118954A1	2009
WO2011015433A1	Purification of a conventional polymer flow contaminated with pla	GALACTIC SA	https://worldwide.espacenet.com/patent/search?q=WO2011015433A1	2009
WO2011029648A1	Method for stereospecifically recycling a pla polymer mixture	GALACTIC SA	https://worldwide.espacenet.com/patent/search?q=WO2011029648A1	2009
WO2021180893A1	Process for recovering styrene monomer from a styrene-containing polymer	INDAVER	https://worldwide.espacenet.com/patent/search?q=WO2021180893A1	2020
WO2021023889A1	Improved method of recycling polyurethane materials	RECTICEL	https://worldwide.espacenet.com/patent/search?q=WO2021023889A1	2019

*Patents that relate to Solvent Purification (without any chemical reaction step) were excluded from this Table. Belgian applicants which have filed patents relating to Solvent Purification in the period 2000-2022 are INERGY AUTOMOTIVE and SOLVAY.

Table 3 - continued: List of CR patents of the past 20 years with contribution of Belgian inventors

Publication No.	Title	Standard applicant	Espacenet URL	Priority year
WO2017167948A1	Process for converting plastic into waxes by cracking and a mixture of hydrocarbons obtained thereby	SOLVAY	https://worldwide.espacenet.com/patent/search?q=WO2017167948A1	2016
WO2017103022A1	Process for producing waxes and liquid fuels from waste plastic	SOLVAY	https://worldwide.espacenet.com/patent/search?q=WO2017103022A1	2015
WO2017103012A1	Use of a catalyst composition for the catalytic depolymerization of plastics waste	SOLVAY	https://worldwide.espacenet.com/patent/search?q=WO2017103012A1	2015
WO2017103018A1	Use of a catalyst composition for the catalytic depolymerization of plastics waste	SOLVAY	https://worldwide.espacenet.com/patent/search?q=WO2017103018A1	2015
WO2006030020A1	Process for the treatment of a mixture of abs and of ps	SOLVAY	https://worldwide.espacenet.com/patent/search?q=WO2006030020A1	2004
WO2005100461A1	Process for the solvent treatment of a plastic	SOLVAY	https://worldwide.espacenet.com/patent/search?q=WO2005100461A1	2004
WO2016058062A1	Cyclo-depolymerisation of polybutadiene	UNIV LEUVEN KATH	https://worldwide.espacenet.com/patent/search?q=WO2016058062A1	2014

*Patents that relate to Solvent Purification (without any chemical reaction step) were excluded from this Table. Belgian applicants which have filed patents relating to Solvent Purification in the period 2000-2022 are INERGY AUTOMOTIVE and SOLVAY.

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4.3 Major Fields, Major CPC codes and Major Technologies

Before diving deeper into the CR patent landscape, it is worth looking at the Major Fields wherein CR patents are filed & at the major CPC codes attributed to them.

4.3.1 Major Fields

Figure 4 below depicts an analysis of the Fields encountered within the patent pool of 3,750 CR patents studied. This kind of analysis is valuable if processes or functions are being explored, or if problems need to be solved. It gives an abstract idea of the possible ways to realize something[6].

The graph provides some insights in the techniques that are involved. The darker or the brighter the colour, the more patents in the segment. Because of a lack of clarity in the domain, and different meanings being given to certain terms, this graph provides a rough indication only.

Figure 4: Fields of activity. The darker/brighter the colour, the more patents in this segment

[6] Based on information provided by PatentInsight

4.3.2 Major CPC Codes

Patent Classification Codes are another way to get a quick insight in the technology behind patents. Classification codes are assigned by Patent Officers at filing, in order to classify patents according to their technical content.

Figure 5 below depicts the Major CPC [7] codes (top 10) attributed to patent families filed in the CR domain.

Table 4 below provides some details about the CPC subcodes and classification used within the Y02W30/00 patent classification.

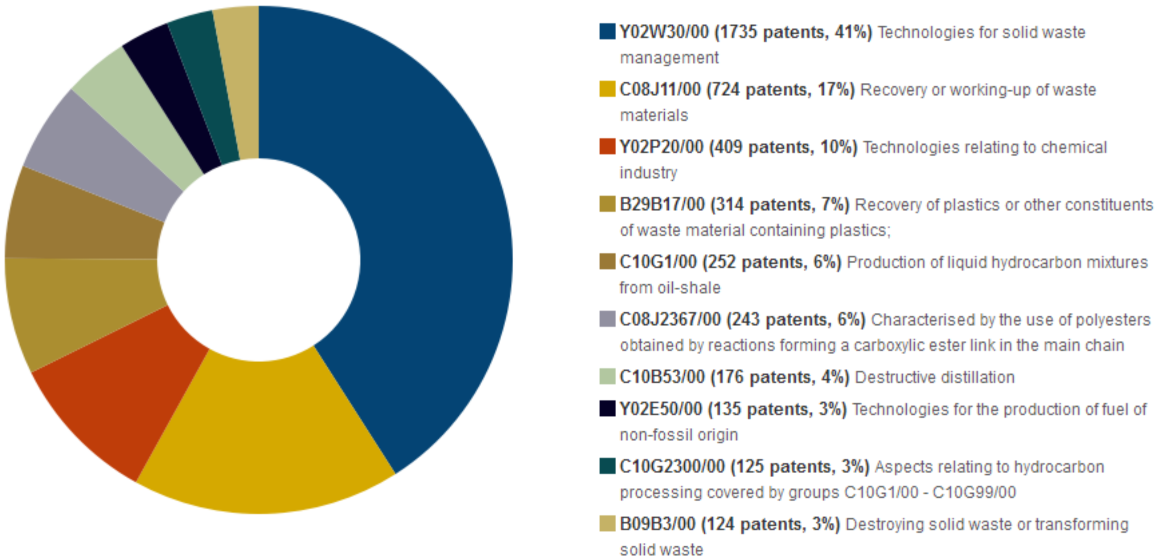

Figure 5: MainGroup CPC Codes (Top10) associated with CR

[7] CPC stands for Cooperative Patent Classification, a joint effort of the EPO (European Patent Office) and the USPTO (United States Patent & Trademark Office) to classify patents in a same way

Within the Y02W30/00 MainGroup, the following CPC codes are of interest in the context of this study:

Y02W30/52	Reuse, Recycling, Recovery
Y02W30/54	Metal recycling
Y02W30/54	Construction waste demolition
Y02W30/60	Glass recycling
Y02W30/62	Plastics recycling
Y02W30/64	Paper recycling
Y02W30/66	Fibre-containing textile recycling
Y02W30/68	Rubber recycling
Y02W30/70	Other than plastics or rubber
Y02W30/80	Packaging reuse
Y02W30/82	Electrical or electronic
Y02W30/84	Batteries
Y02W30/86	Fuel cells
Y02W30/90	Different types of waste
Y02W30/91	Fillers for concrete
Y02W30/95	Building materials
Y02W30/96	Organic materials like rubber, PS[8]

Table 4: Different Y02W30/00 subcodes and what they cover

4.3.3 Major Technologies involved over time

Some of the technologies used in Chemical Recycling (CR) and their activity over time are depicted in Figure 6. This Figure gives us an idea of when the first patents around a certain technology were published, and if the technology evolved or not. In the second place, this mapping can provide an insight in how mature certain technologies are, in particular when patent insights are combined with commercial or literature insights.

Following techniques were looked at: (1) Chemolysis, (2) Pyrolysis, (3) Catalytic Cracking and (4) Gasification.

Pyrolysis is dominating the picture, followed by Chemolysis. There are less patents about Catalytic Cracking and Gasification. The first patents on Chemolysis published in 2005 and the first Pyrolysis patents in 2004. For the rest we see the same pattern as for the overall CR patent landscape: a first patent activity peak is seen around 2007 and we see a sharp increase in activity the past few years.

[8] Polystyrene

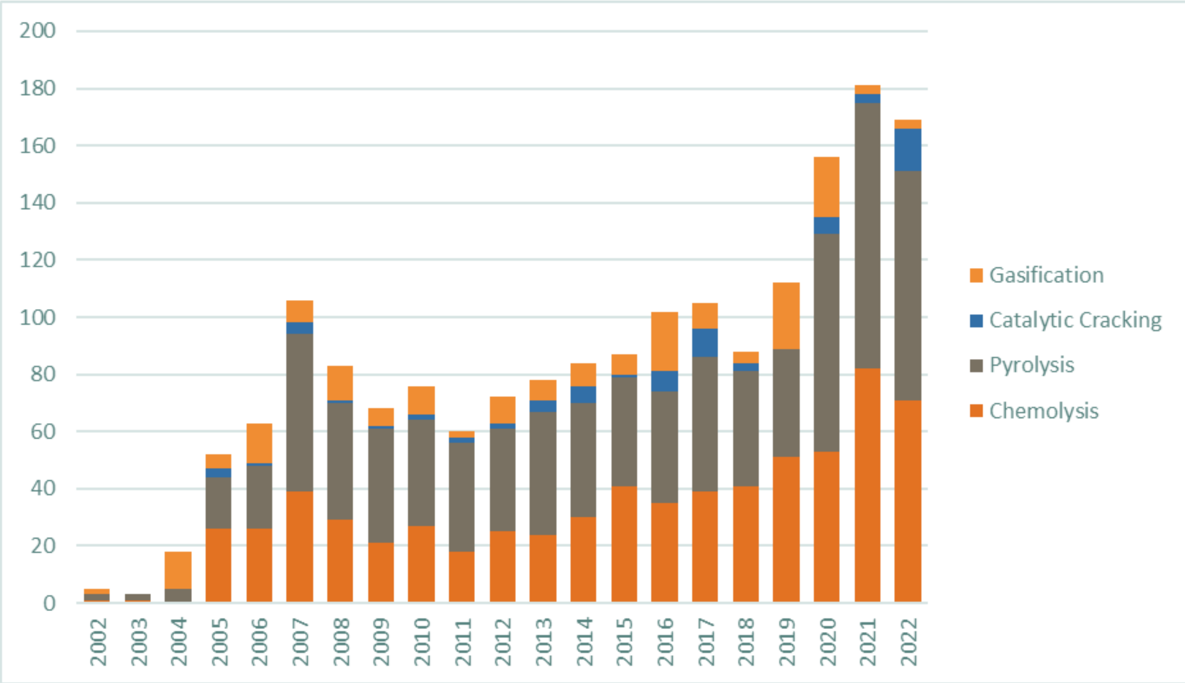

Figure 6: Major CR Technologies with time

In the next Chapter we look in more details at the major CR technologies. We start with Chemolysis, then Pyrolysis, next Catalytic Cracking and Gasification. We end with taking a closer look at Biological Approaches described in CR patent literature.

5 Selection of topics

As discussed in Chapter 1, Chemolysis, Pyrolysis, Catalytic Cracking and Gasification are commercially exploited CR technologies[9]. It is hence expected that this reflects itself in a high number of patent publications on these subjects. Dedicated patent data sets were created for each of them & studied to know more about the main players and key patents.

5.1 Chemolysis

Chemolysis is one of the more important technologies used in Chemical Recycling (CR) and this is confirmed by the high number of patent publications found on this topic. A subset of 680 patents on chemolysis was created & studied, and further cleaned where necessary.

According to literature, chemolysis has the advantage that polymers can be broken down to the monomer level, and the technique is commercially operational for PET. Though the energy and operation cost is lower than for pyrolysis and gasification, high volumes are required for chemolysis to be cost-effective. As the technique is sensitive to contaminants, it is said to be suitable for homogenous plastics only[9].

Figure 7 shows that the interest in Chemolysis from a patent perspective is continuously growing, and is peaking the last 2 years. The onset is in 2005. The trend follows more or less that of the overall CR field. It can be expected that the final number of patent publications in 2022 will be as high as in 2021 (see the blue trend line).

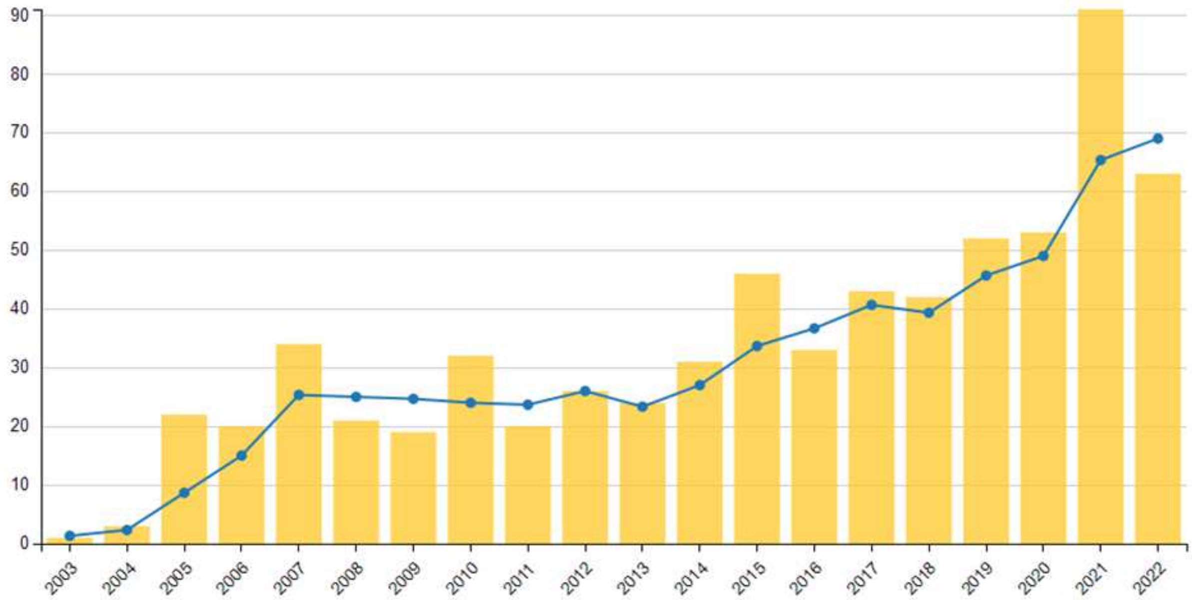

Figure 7: Number of patent publications by publication year (period 2003-2022) - Chemolysis

[9] ECHA – Chemical Recycling of Waste in the Circular Economy

Table 5 below shows the top 10 filing Companies. We see that EASTMAN CHEMICALS CO leads the list by far. The other companies listed in the top 10 come from various parts of the world though Asian companies seem to dominate this list. The only European firm in the top 10 is IFP ENERGIES NOW, a French company.

Table 6 below show the top 5 filing Academics. This time Chinese players dominate this list. There are no European Academic figuring in this list. The total of Academics filing in this field lies around 100.

Company	Country	No. of patent families
EASTMAN CHEM CO	US	33
CENTRAL PLASTIC UNION NEW MAT TECH HUBEI CO LTD	CN	7
NITTO DENKO CORP	JP	6
HUVIS CORP	KR	5
SION TECH CO LTD	KR	5
GREENMANTRA RECYCLING TECH LTD	CA	5
IFP ENERGIES NOW	FR	5
TORAY INDUSTRIES	JP	4
IBM	US	4

Table 5: Top 10 filing Companies within Chemolysis

Academic Entity	Country	No. of patent families
UNIV ZUNYI NORMAL	CN	6
INST PROCESS ENG CAS	CN	5
FAR EAST COLLEGE	TW	4
UNIV JIANGNAN	CN	4
UNIV ZHEJIANG SCIENCE & TECH	CN	4

Table 6: Top 5 filing Academics within Chemolysis

If we look at the top 10 applicant countries then we see that Germany, France and Belgium figure in the top 10 [Figure 8]. France is due to CARBIOS and IFP ENERGIES NOW mainly, the latter focusing on PET recycling. Belgium is mainly due to GALACTIC SA, having filed patents on the depolymerization of polylactic acid (PLA) and RECTICEL, having filed a patent on the depolymerization of polyurethanes (PU).

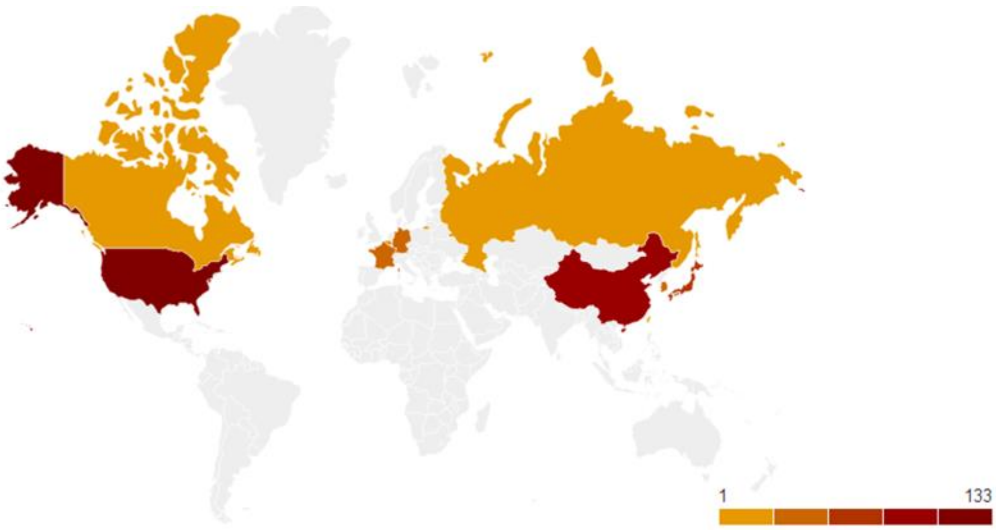

Figure 8: Top 10 publication countries (without EP and WO)

Most patent filings are done in China, the US and Japan. When we take a closer look at which applicants filed when, then we see that Japan had a high patent activity near 2007, but knew a decline since then; while patent activity is taking on only now in the US [Figure 9]. EASTMAN CHEMICALS CO has been heavily investing in this technology the past few years (with 30 patent publications in 2020-2021).

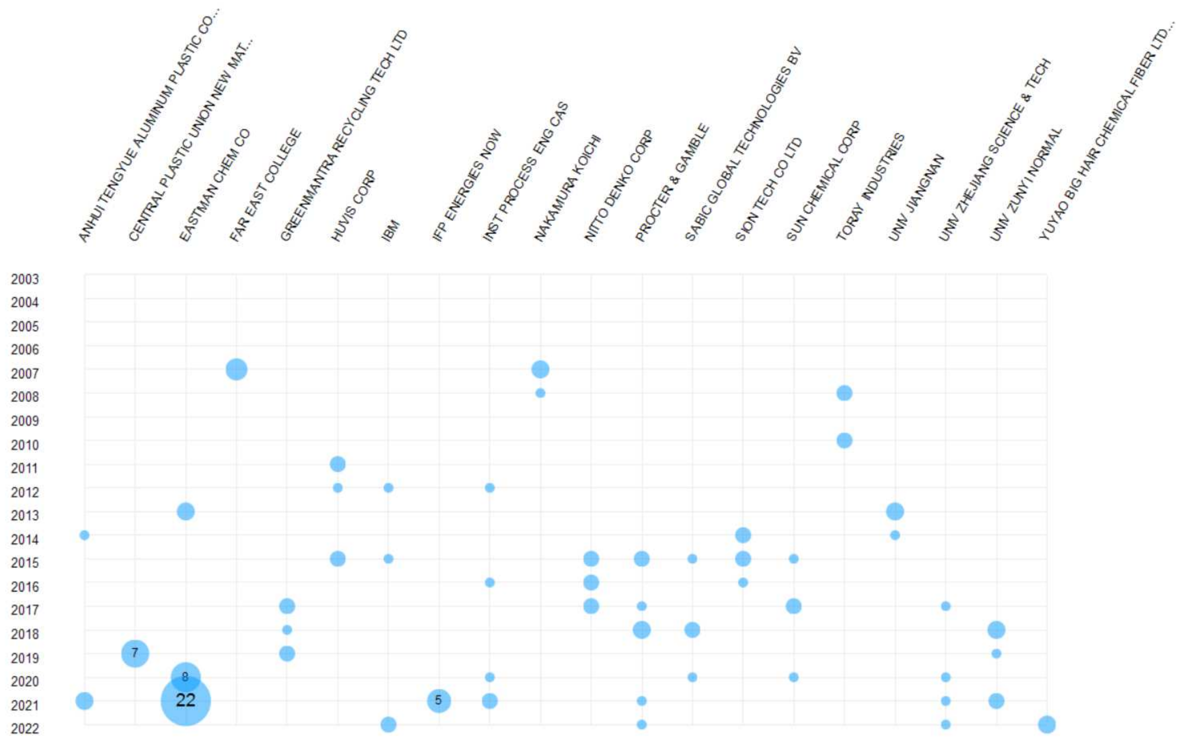

Figure 9: Patent activity of Chemolysis applicants with time

Using a patent value analysis, patents were scored based on the size of the patent family and the number of forward patent citations. A high score is obtained if the family is large, which would be an indicator of the market significance of the patent, and if the number of forward citations is large, which is an indicator of the significance of the technology described. From the analysis done, the patents with a high value score as well as patents that have a large number of citations were selected and listed in Table 7 below.

Value	Score	Publication Date	Size Family	Size Forward Citations
US2007045456A1	85	01 Mar 2007	48	111
WO2016062695A1	68	28 Apr 2016	14	31
EP3112406A1	62	04 Jan 2017	19	8
WO2018069794A1	57	19 Apr 2018	6	15
US2015105532A1	57	16 Apr 2015	8	18

Table 7: Most valuable Chemolysis patents

US2007045456A1 from XYLECO INC [US] relates to the use of a microorganism to produce fuel (ethanol) from fibrous materials with lignin as by-product. WO2016062695A1 from CARBIOS [FR] discloses a peptide with polyester degrading activity[10]. WO2020152317A1 from SABIC GLOBAL TECHNOLOGIES BV [NL] is about the making of polyethylene from mixed plastics. US2015105532A1 from IBM [US] is one the depolymerization of PET bottles via alcoholysis.

[10] The first 2 patents relate to Biological/Enzymatic depolymerization approaches

5.2 Pyrolysis

Pyrolysis is one of the most important technologies used in Chemical Recycling (CR) and this is confirmed by the hight number of patent publications found on this topic. A subset of 830 patents on Pyrolysis was created and studied, and further cleaned where necessary.

According to literature, pyrolysis has the advantage that this technique can be used for difficult to depolymerize plastic waste and for heterogeneous mixtures of plastics. High caloric value fuel can be produced for use in gas engines. Downsides would be a high energy requirement (due amongst others to the need for high temperatures), complexity of reactions & low tolerance to PVC (because of chlorinated compounds being created during pyrolysis). Like for chemolysis, high volumes are required for pyrolysis to be cost-effective[11].

Figure 10 shows that the interest in Pyrolysis is continuously growing, and is peaking the last 3 years. The onset is in 2005. The trend follows more or less that of the overall CR field. It can be expected that the final number of patent publications in 2022 will be at least as high as in 2021 (see the blue trend line).

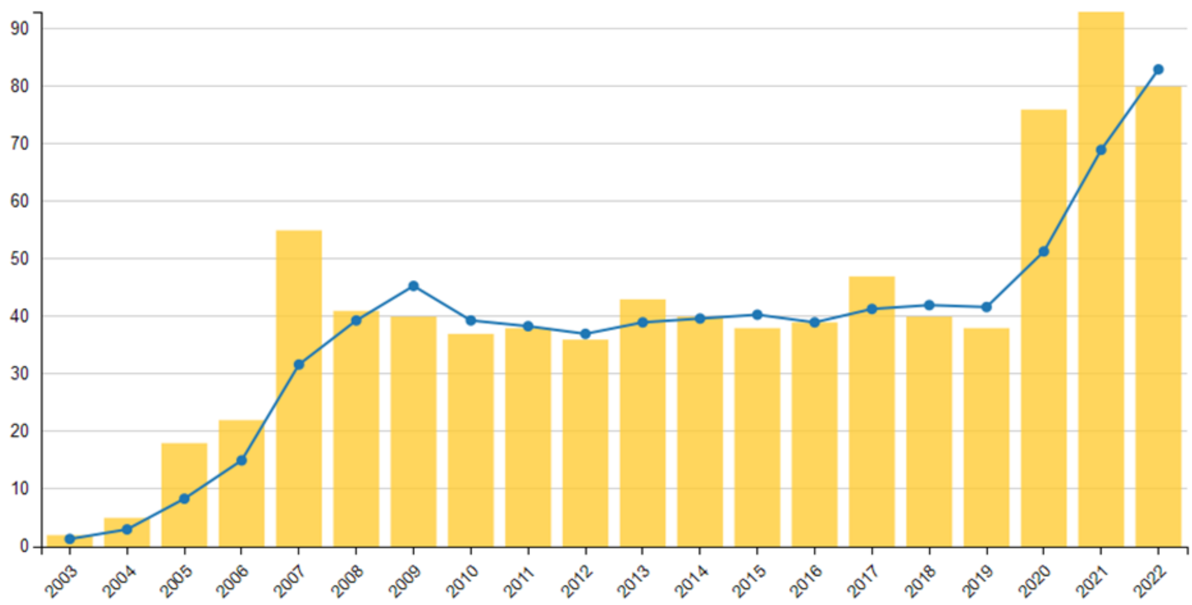

Figure 10: Number of patent publications by publication year (period 2003-2022) - Pyrolysis

[11] ECHA – Chemical Recycling of Waste in the Circular Economy, Table 5.2

Table 8 below shows the top 10 filing companies within Pyrolysis. Again, EASTMAN CHEMICALS CO is leading the list by far. Toshiba KK, one of the overall top filers [see Table 2], also figures in this list. It are primarily US and Japanese companies that dominate the Top 10. The only European company in the Pyrolysis Top 10 is SABIC GLOBAL TECHNOLOGIES BV, a Dutch company .

Table 9 below show the top 5 filing Academics. This time Korean Academics dominate this list and no European Academic figures in this list. The total of Academics filing in this field lies around 100.

Company	Country	No. of patent families
EASTMAN CHEM CO	US	46
JFE CHEMICAL CORP	JP	15
TOSHIBA KK	JP	13
SABIC GLOBAL TECHNOLOGIES BV	**NL**	10
JFE STEEL CORP	JP	8
CHEVRON USA CORP	US	8
BRIDGESTONE CORP	JP	7
MITSUI SHIPBUILDING ENG	JP	7
UOP LLC	US	7
NIPPON STEEL CORP	JP	5

Table 8: Top 10 filing Companies within Pyrolysis

Academic Entity	Country	No. of patent families
KOREA ENERGY RESEARCH INST	KR	9
UNIV SEOUL IND COOP FOUND	KR	6
UNIV SHANGHAI 2ND POLYTECHNIC	CN	5
UNIV MASSACHUSETTS	US	4
KOREA RES INST CHEM TECH	KR	4
UNIV CENTRAL SOUTH	CN	3

Table 9: Top 5 filing Academics within Pyrolysis

If we look at the top 10 applicant countries then we see that Germany, Spain and the Netherlands figure in the top 10 [Figure 11]. The Netherlands is mainly due to SABIC GLOBAL TECHNOLOGIES BV with focus on the production of polyolefins and pyrolysis oil from waste plastic and dechlorination of pyrolysis oil made from mixed plastics.

Most patent filings are done in China, the US and Japan. When we take a closer look at which applicants filed when, then we see again that Japan had a high patent activity near 2007, but knew a decline since then while patent activity is taking on only now in the US [Figure 12]. EASTMAN CHEMICALS CO has been heavily investing in this technology the past few years (with 44 patent publications from 2020 till now).

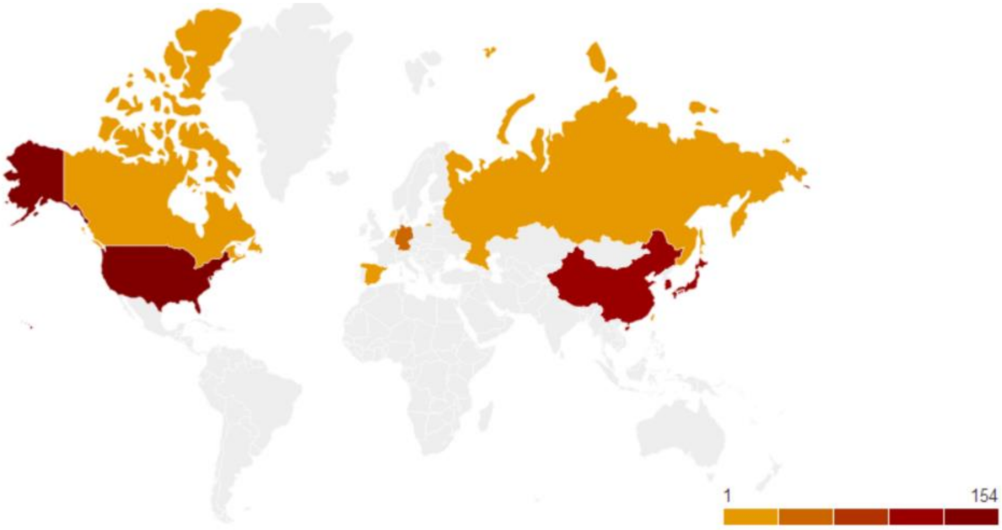

Figure 11: Top 10 publication countries (without EP and WO)

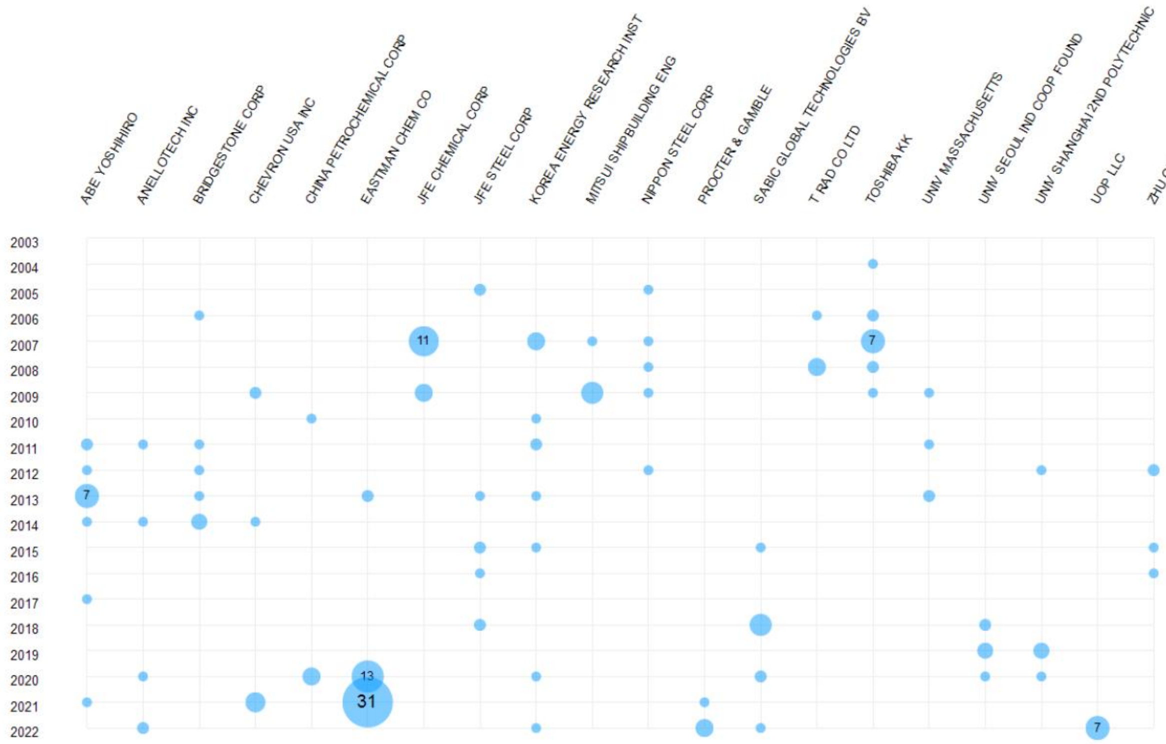

Figure 12: Patent activity of Pyrolysis applicants with time

Using a patent value analysis, patents were scored based on the size of the patent family and the number of forward patent citations. A high score is obtained if the family is large, which would be an indicator of the market significance of the patent, and if the number of forward citations is large, which is an indicator of the significance of the technology described. From the analysis done, the patents with a high value score as well as patents that have a large number of citations were selected and listed in Table 10.

Value	Score	Publication Date	Size Family	Size Forward Citations
US2009227823A1[12]	73	10 Sep 2009	14	171
WO2011097648A2	69	11 Aug 2011	36	25
WO2020152317A1	58	30 Jul 2020	20	3
US2020248082A1	55	06 Aug 2020	12	4
CA3019392A1	53	05 Oct 2017	30	1
CA3133879A1	45	24 Sep 2020	15	0

Table 10: Most valuable Pyrolysis patents

US2009227823A1 from the UNIVERSITY OF MASSACHUSETTS [US] has the highest score and is related to the catalytic pyrolysis of solid biomass, more in particular lignocellulosic biomass, to convert it into biofuel, aromatics and olefins. Pyrolysis is done in a fluid bed using high ratios of catalyst over hydrocarbons. The biofuels thus produced are said to be compatible with existing liquid transportation fuels, such as gasoline and diesel, and can be produced at high enough rate.

WO2011097648A2 from FULCRUM BIOENERGY INC [US] relates to processes and equipment for economically converting municipal solid waste into ethanol and syngas.

CA3019392A1 from RES POLYFLOW LLC [US] relates to the production of solid fuels formed from mixed solid waste, in particular mixed plastic waste.

US2020248082A1 from EASTMAN CHEMICALS CO [US] is one of the youngest patents with a relatively high patent value score and a large patent family. The patent describes the making of polyethylene from plastic waste.

Other 2020 patents with high family size are WO2020152317A1 from SABIC GLOBAL TECHNOLOGIES BV [NL] and CA3133879A1 from MICROWAVE SOLUTIONS GMBH [CH] resp. WO2020152317A1 relates to a process for the preparation of polyethylene from waste plastic feedstocks. CA3133879A1 relates to a pyrolysis method for recovering silica from a polymer waste material containing silica, like a rubber or plastics waste material containing silica.

[12] Patent that relates to catalytic cracking, see section 5.3

A high family size points to the importance given by the filing entity to this technology. The forward citation score for these patents is low but this might be due to the fact that this is relatively new technology.

The above demonstrates that the US and Europe score well according to this patent value analysis. Though most of the high value patents are from companies, the patent from the University of Massachusetts has the highest overall score.

In a last step, we looked at Developing Chemical Recycling Techniques listed in the ECHA 2021 report on Chemical Recycling[13]: Hydrocracking, Pyrolysis with in-line reforming, Microwave-assisted pyrolysis and Plasma pyrolysis as Developing chemical recycling techniques. The patent literature reflects this: Of the 833 patents studied, only 25 related to Microwave Pyrolysis which technique appears to be used for the depolymerization of a variety of waste materials such as polyesters, polyolefins, polystyrene and WEEE[14]. Applicants include PYROWAVE [US], MICROWAVE SOLUTIONS [CH] and a few Chinese petrochemical companies. About 10 patents related to Plasma Pyrolysis, most of them filed by Chinese applicants and not filed outside of China. About 30 patents related to Hydrocracking, filed by Japanese companies such as JFE CHEMICAL CORP and MITSUI SHIPBUILDING. This confirms the low economic interest in these technologies.

5.3 Catalytic cracking

Catalytic cracking is mentioned as one of the more important technologies used in Chemical Recycling (CR), yet we find a low number of patents published on this topic. A subset of 71 patents on catalytic cracking was created and studied, and further cleaned where necessary.

According to literature, compared to conventional pyrolysis, catalytic cracking has a reduced production cost and energy consumption (as lower operating temperatures can be used & reaction times are shorter), and gives higher oil yields. Some of the major downsides of this technique would be the absence of a suitable reactor technology and the sensitivity to contaminants. Presence of nitrogen and chloride can deactivate the catalyst and inorganic components can block catalyst pores[15].

Despite the low number of Catalytic Cracking patents found [71 only], some high value patents have been filed within this technological segment [see Table 12, US2009227823A1]. Plotting the number of patents by publication year, we saw a lot of variation over the years but this may be due in part to the low number of patents in this data set [Figure not shown]. What can be noticed though is a high patent activity in 2022 compared to the previous years [evident from Figure 6 too].

Table 11 lists the Top 3 applicants amongst respectively companies and academics: 1 Belgian company in this top 3, SOLVAY

[13] ECHA – Chemical Recycling of Waste in the Circular Economy, Table 5.2
[14] Waste electrical and electronic equipment
[15] ECHA – Chemical Recycling of Waste in the Circular Economy, Table 5.2

Applicant	Country	No. of patent families
UNIV QINGDAO TECHNOLOGICAL	CN	6
ANELLOTECH INC	US	5
UNIV MASSACHUSETTS	US	4
SOLVAY	**BE**	4

Table 11: Top 10 filing Entities within Catalytic Cracking

As mentioned before, a patent value analysis was done to score patents based on the size of the patent family and the number of forward patent citations. A high score is obtained if the family is large, which would be an indicator of the market significance of the patent, and if the number of forward citations is large, which is an indicator of the significance of the technology described. From the analysis done, the patents with a high value score as well as patents that have a large number of citations were selected and listed in Table 12 below.

Value	Score	Publication Date	Size Family	Size Forward Citations
US2009227823A1[16]	73	10 Sep 2009	14	171
US2013060070A1	63	07 Mar 2013	6	36
US2007173673A1	60	26 Jul 2007	6	39
US2013324772A1	60	05 Dec 2013	6	23
CN111825514A	53	27 Oct 2020	7	2
CA2773311A1	47	17 Mar 2011	18	0

Table 12: Most valuable Catalytic Cracking patents

What stands out is the following: (i) most of the above have been filed by Academics and (ii) there is a top scorer.

Four out of the six patents listed in Table 12 are from the UNIVERSITY OF MASSACHUSETTS [US]. US2009227823A1, which has the highest score, deals with the catalytic cracking of solid biomass (more in particular lignocellulosic biomass) to convert it into biofuel, aromatics and olefins[17]. US2013060070A1, US2013324772A1 and CA2773311 are 3 other patents from that same University on the making of fluid hydrocarbons. These 4 patents published between 2009 and 2013.

US2007173673 is from the KITAKYUSHU FOUNDATION [JP] and describes the catalytic cracking of plastic with an oil recovery rate of over 50%, though the cracking is at low temperature and cracking & dechlorination take place in the same reaction vessel.

CN111825514A from ZHEJIANG KEMAO ENVIRONMENT TECH CO LTD [CN] is one of the few patents originating from China with a high patent value score. This patent describes a catalytic cracking process of plastic that maximizes ethylene and propylene production.

[16] Information on the patent given in section 4.1
[17] See also section 5.2

5.4 Gasification

Also Gasification is mentioned as an important technology used in Chemical Recycling (CR).

According to literature, Gasification is mainly used for the production of syngas. The technique can be used for mixed plastics and allows for a detailed polymer breakdown but has the disadvantage that further upgrading is needed before use. High feedstock volumes are needed for the technique to be feasible. A few of the disadvantages besides high cost and energy consumption are the production of noxious NO_x, the formation of tar leading to operational and efficiency issues, and the sensitivity to contamination[18].

We see that less patents have been filed around Gasification than around Pyrolysis. A subset of 187 patents around this topic was created and studied. Due to the lower number of patents in this technology segment little to nothing can be said about trends see, though it appears that there is a decline after the peak in 2020 [Figure 6 and Figure 13]. This needs to be confirmed.

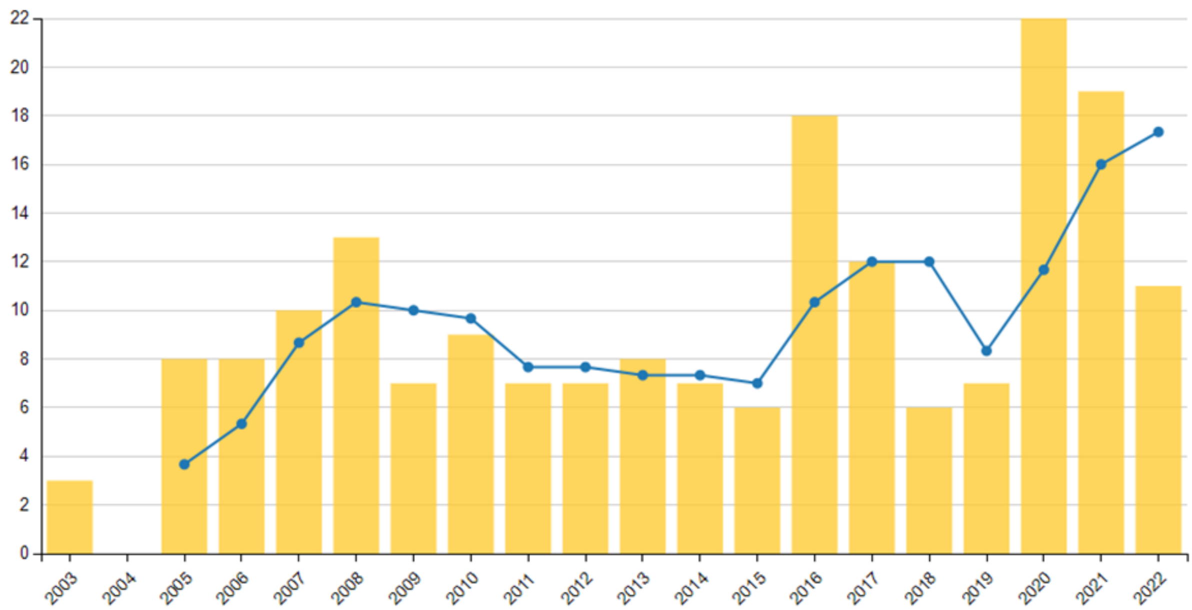

Figure 13: Number of patent publications by publication year (period 2003-2022) – Gasification

Table 13 below lists the top 5 applicants (companies + academics). We see once more that EASTMAN CHEMICALS CO [US] is leading the list as top scorer. For the rest Japanese and Chinese applicants dominate the picture. Most of the applicants have 1 patent on their name only, so the field is scattered and immature.

[18] ECHA – Chemical Recycling of Waste in the Circular Economy, Table 5.2

Applicant	Country	No. of patent families
EASTMAN CHEM CO	US	22
JFE STEEL CORP	JP	6
UNIV QINGDAO TECHNOLOGICAL	JP	6
TAKAHASHI KENZO	JP	4
TSURUMI KAZUYUKI	JP	3
FUJIAN ZHENGREN ENV PROT CO LTD	CN	3

Table 13: Top 10 filing entities within Gasification

If we look at the top 10 applicant countries then we see that Germany and Poland figure in the top 10 [Figure 14].

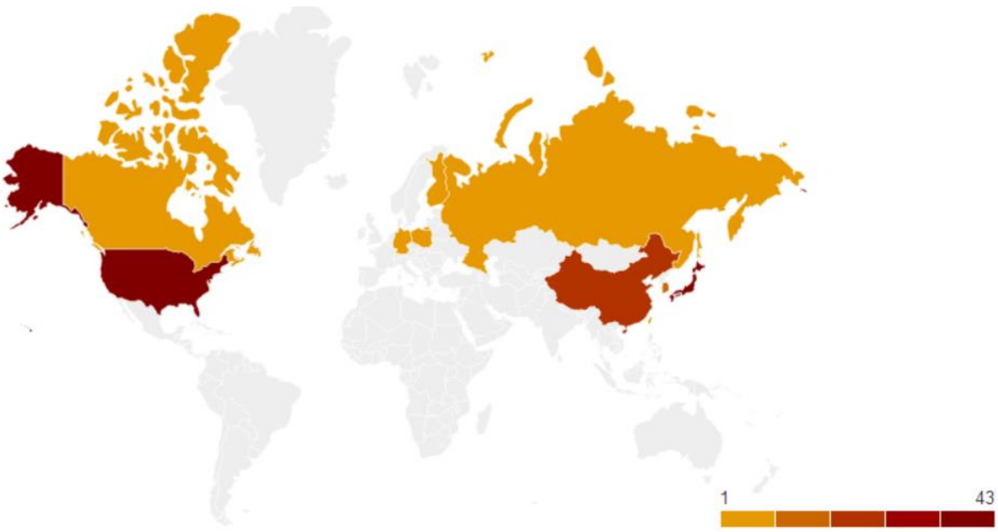

Figure 14: Top 10 publication countries (without EP and WO)

Most patent filings are done in China, the US and Japan. When we take a closer look at which applicants filed when, then we see once more that Japan had a high patent activity near 2007, but knew a decline since then while patent activity is taking on only now in the US [Figure 15]. EASTMAN CHEMICALS CO has been heavily investing in this technology the past few years (with 22 patent publications from 2019 till now).

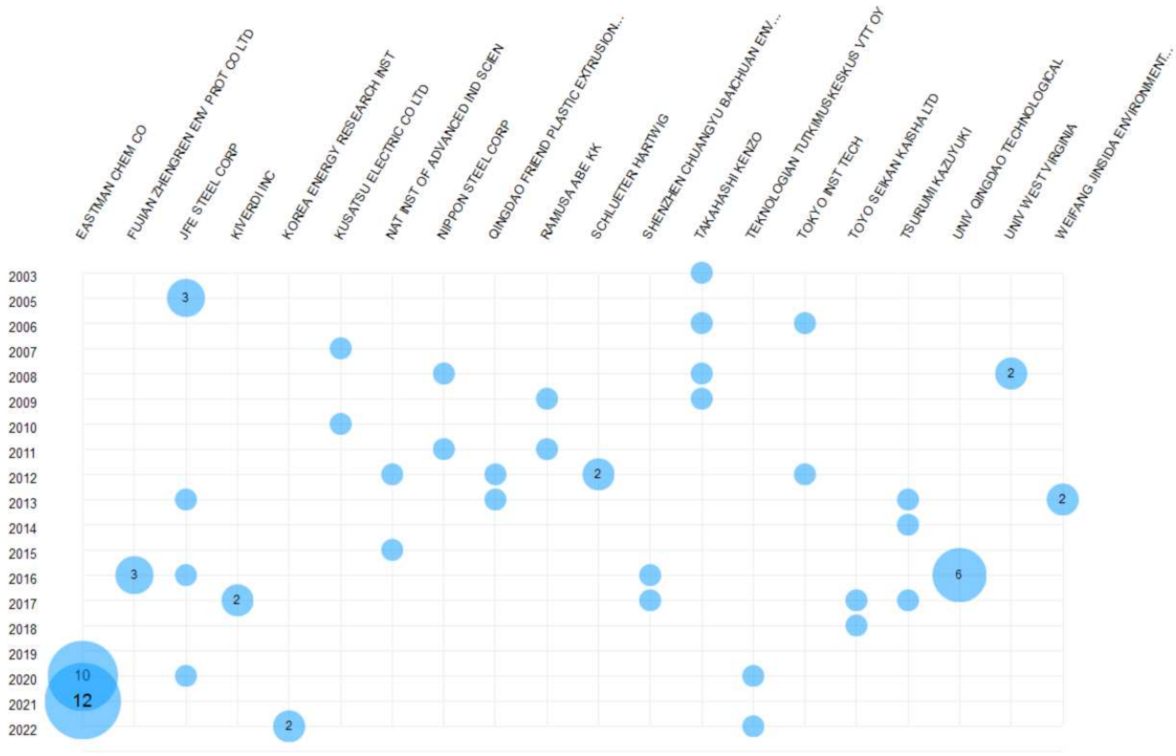

Figure 15: Patent activity of Gasification applicants with time

Using a patent value analysis, patents are scored based on the size of the patent family and the number of forward patent citations. A high score is obtained if the family is large, which would be an indicator of the market significance of the patent, and if the number of forward citations is large, which is an indicator of the significance of the technology described. From the analysis done, the patents with a high value score as well as patents that have a large number of citations were selected and listed in Table 14.

Value	Score	Publication Date	Size Family	Size Forward Citations
WO2011097648A2	75	11 Aug 2011	36	25
US2016122674A1	67	05 May 2016	29	7
US2016122673A1	61	05 May 2016	17	7
US2007173673A1	58	26 Jul 2007	6	39
US2020248082A1	58	06 Aug 2020	12	4
CA2475015A1	51	14 Aug 2003	16	6

Table 14: Most valuable Gasification patents

WO2011097648A2 from FULCRUM BIOENERGY INC [US] discloses processes for economically converting municipal solid waste into ethanol. US2016122674A1 and US2016122673A1, both from ECOGENSUS [US] relate to solid fuel/syngas prepared from mixed solid waste. US2020248082A1 is from EASTMAN CHEMICALS [US AND is about the feed location for gasification of plastics and solid fossil fuels, CA2475015A1 from the UNIVERSITY OF CALIFORNIA [US] relates to the production of synthetic transportation fuels from carbonaceous materials using self-sustained hydro-gasification.

We see that the important patents in this field are filed by US companies, which is not surprising as Chinese and Japanese patents - though some have been granted - rarely were filed outside of the domestic country.

On the total of 187 patents studied only 5 related to plasma gasification, so there seems to be little interest therein.

5.5 Biological and enzymatic degradation of polymers

Figure 16 below shows that the interest in biological solutions for Chemical Recycling (CR) is increasing with the years & in particular as of 2018. In 2022 the actual number of patent publications is already higher than in 2021.

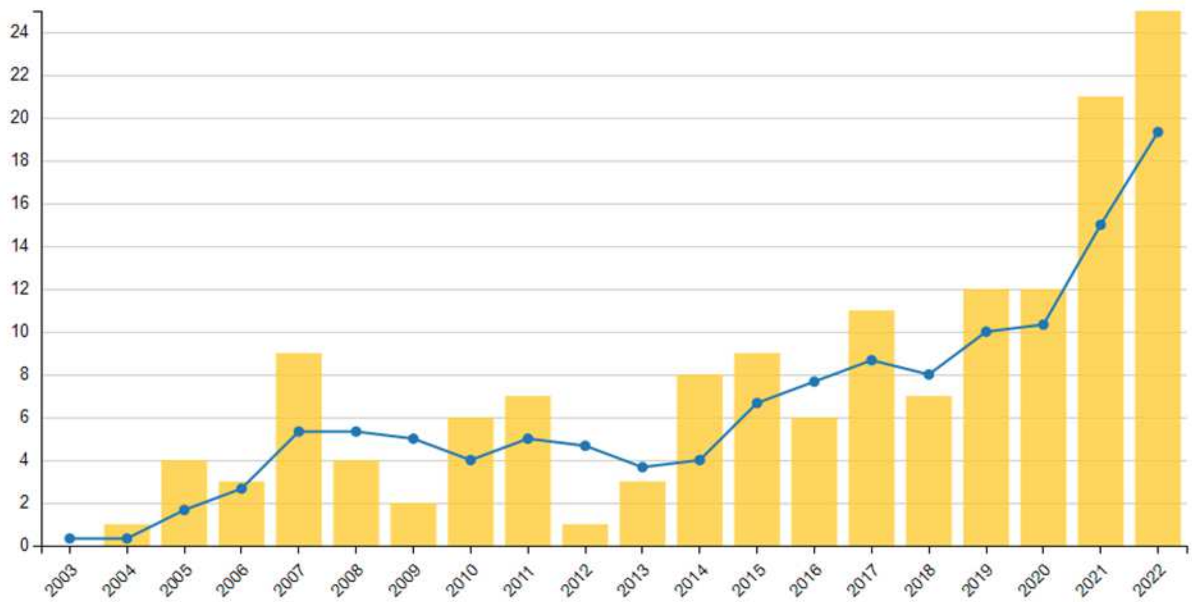

Figure 16: Number of patent publications by publication year (period 2003-2022) – Biological/Enzymatic depolymerization

Table 15 below lists the top 5 applicants (companies + academics). CARBIOS [FR] is the main player in this field, followed by Japanese and US players. DU PONT has filed patents on the biodegradation of aliphatic-aromatic co-polyesters by enzymatic treatment. ALLIANCE SUSTAINABLE ENERGY [US] is focusing on enzymes and plastic degrading fusion proteins for polymer degradation. NESTLÉ'S patent covers the enzymatic recycling of polyurethanes via cutinases, lipases and esterases.

Applicant	Country	No. of patent families
CARBIOS	FR	16
TOYO SEIKAN KAISHA LTD	JP	6
TEIJIN FIBERS LTD	JP	4
JAPAN SCIENCE & TECH AGENCY	JP	3
DU PONT	US	3
NAGARAJAN VASANTHA	IN	3
ALLIANCE SUSTAINABLE ENERGY	US	3
NESTLE SA	US	3

Table 15: Top 10 filing entities within Biological/Enzymatic depolymerization

If we look at the top 10 applicant countries then we see that France, Germany and Italy figure in the top 10 [Figure 17].

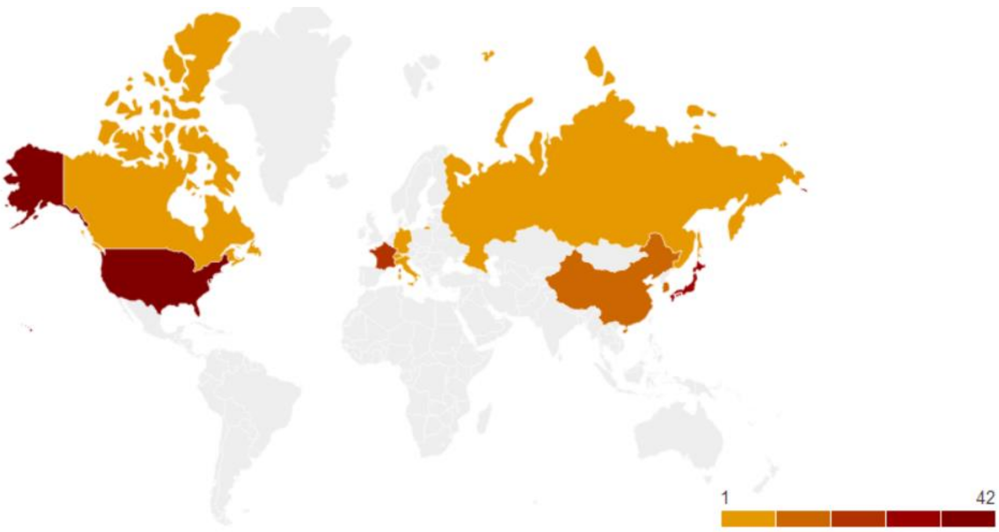

Figure 17: Top 10 publication countries (without EP and WO)

In France we have CARBIOS focusing on novel esterases from bacterial origin. Also in France, the CENTRE NAT RECH SCIENT [CNRS, FR] and the UNIV POITIERS [FR] filed a patent on Bacterial strains that can degrade polylactic acid [PLA]. In Germany COVESTRO has a patent on urethanases for the enzymatic degradation of polyurethanes. And in Italy, INNOVAZIONE E SVILUPPO SOSTENIB has a patent on plastic degrading bacterial solutions.

CARBIOS has been filing patent applications on esterases for chemical recycling since 2013 while other European and US players started filing in this field only recently. Again, we see a patent activity in Japan that peaked in 2007 but slowed down thereafter. DU PONT also seems to have stopped its patent activities after 2006 [Figure 18 below], at least for now.

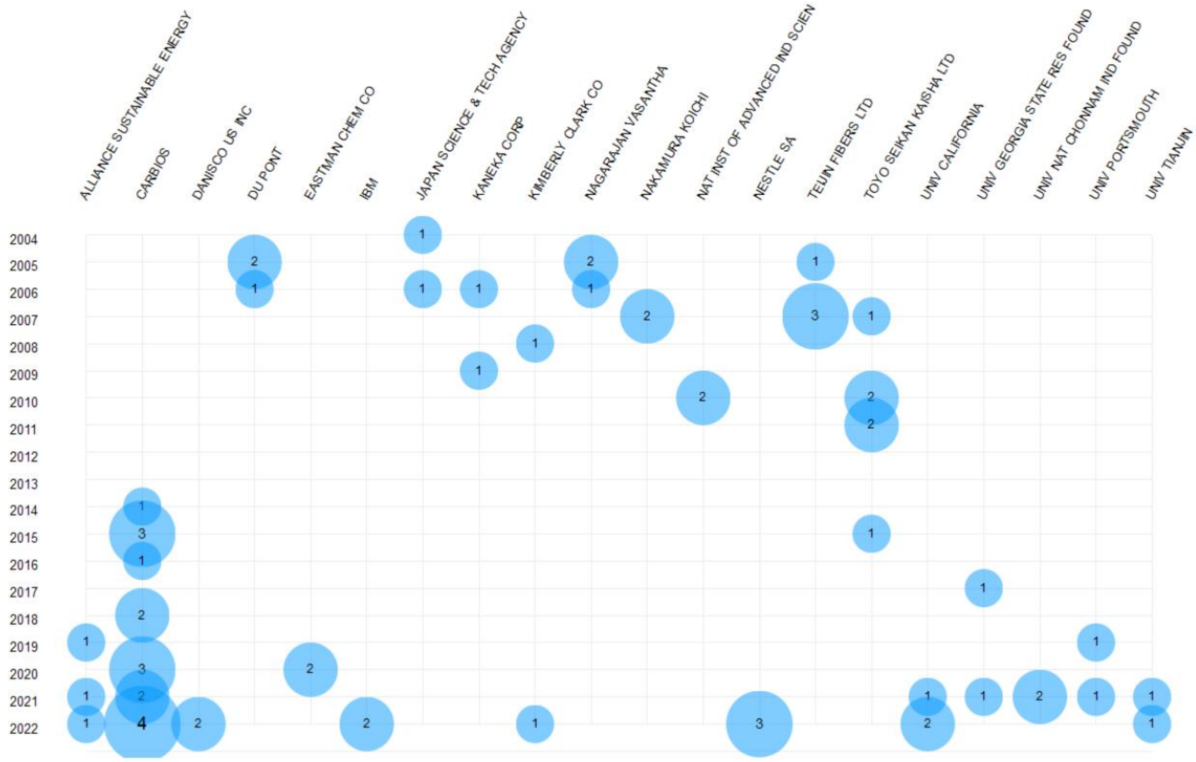

Figure 18: Patent activity of applicants with time (Biological/Enzymatic depolymerization)

In a last step, we looked at valuable patents filed on Biological/Enzymatic depolymerization. Using a patent value analysis, patents are scored based on the size of the patent family and the number of forward patent citations. A high score is obtained if the family is large, which would be an indicator of the market significance of the patent, and if the number of forward citations is large, which is an indicator of the significance of the technology described. From the analysis done, the patents with a high value score as well as patents that have a large number of citations were selected and listed in Table 16 below.

Not surprisingly, the first 4 patents and the last patent listed in this Table are all held by CARBIOS. US2007045456 is the most valuable in this category with a total score of 85. This patent relates to an isolated polypeptide with excellent polyester degradation capability. This polypeptide is said to allow at industrial scale the degradation of polyesters contained in a plastic product. Degradation products (monomers and oligomers) thus obtained can be reused to produce new polyesters both economically and reliably. WO2011039489 is held by CNRS and relates to bacterial strains that can degrade polylactic acid.

Value	Score	Publication Date	Size Family	Size Forward Citations
US2007045456A1	85	01 Mar 2007	48	111
WO2016062695A1	68	28 Apr 2016	14	31
WO2018011281A1	63	18 Jan 2018	12	15
WO2018011284A1	63	18 Jan 2018	12	15
WO2011039489A1	58	07 Apr 2011	11	19
WO2015097104A1	47	02 Jul 2015	2	19

Table 16: Most valuable patents on Biological/Enzymatic depolymerization

6 Conclusion

With the European Green Deal and the ambition of Europe to be a world leader in the circular economy in mind, it is worth looking at the potential that Chemical Recycling (CR) holds. Relocation of essential resources and circular plastic strategies pledge to collect more waste and to recycle it, instead of incineration, export or landfilling. Chemical recycling technologies are on the rise.

This report provides an analysis of the innovative activities in the field of Chemical Recycling by a review of the patent activity. Patent data sets were evaluated using "PatentInspiration" whereby insight was obtained with respect to activity over time, main players and technologies used. Patent value analysis was used to obtain the key innovations in the respective data sets.

A patent data set holding 3,750 patent families relating to CR was studied. The following can be concluded based on our study of the general data set and more focused data sets:

- First patents on CR got published near 2005. In 2007 we see a first peak in patenting activity, with Japan as one of the major filing countries around that time. As of 2018, patent activity in the CR domain is steadily growing worldwide (including in Europe and the US). In 2022 we have already more filings than the year before and it is expected that another 100-200 patent publications will follow before the year's end.
- Overall, the CR patent landscape is scattered. There are over 1,000 applicants (filing entities) of which most have filed 1 patent only. Chinese applicants mainly file utility models (or short patents) and most do not file outside of China. Also most Korean applicants file domestically only.
- The patent landscape is dominated by China, Japan, Europe, the US and Korea. Europe takes the third place. When we look at individual countries (instead of at Europe as a whole), we see that Germany, France and Italy figure in the top 1. Belgium figures in the top 15 of active players.
- Table 3 provides a list of CR patents having Belgian inventors
- The review of patent literature confirms that Chemolysis, Pyrolysis, Catalytic Cracking and Gasification are major CR technologies. Most patents got published on Pyrolysis and Chemolysis than on Catalytic Cracking and Gasification.
- Eastman Chemicals Co [US] is by far the entity that has filed most patents on CR (in the period 2019-2021 mainly). They filed many patents on Chemolysis, Pyrolysis and Gasification in particular. In the area of Biological/Enzymatic depolymerization Carbios [FR] leads by far
- For most valuable patents within major CR technology segments, we refer to 10, 12, 14 and 16. For an overall view on most relevant patents in the sector we refer to Table 7,

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