A study of the state-of-the-art and potential impact of Phosphorus, Inorganic and Nitrogen Flame Retardants (PIN FRs) on plastic recycling.

February 2023

PNO

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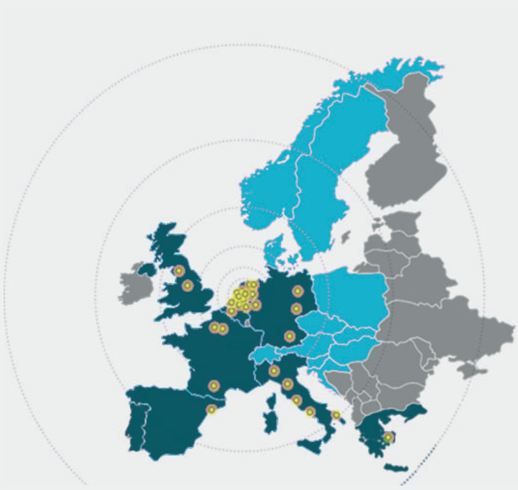

KEY FACTS

35+
Years of activity

9
Countries

2000+
Open Innovation project per year

4.000+
Funded projects and partners in our intelligence tool

70.000+
Qualified RD&I partners

€ 1Bn
Total annual funding won

October 2022

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List of abbreviations and definitions

ABS	Acrylonitrile butadiene styrene
AOH	Aluminium oxide hydroxide
APP	Ammonium polyphosphate
ATH	Aluminium tri-hydroxide
ATO	Antimony trioxide
BDP	Bisphenol A bis(diphenyl phosphate)
BFRs	Brominated flame retardants
CAGR	Compound annual growth rate
CAS	Chemical Abstract Service
CDP	Cresyl diphenyl phosphate
CPA	Circular Plastic Alliance
CPC	Cooperative Patent Classification
CPVC	Chlorinated polyvinyl chloride
CR	Chlorinated rubber
crPET	Chemical recycled PET
CTA	Cellulose triacetate
DEPAL	Diethylphosphinate, aluminium salt
DMPP	Dimethyl propane phosphonate
DPO	Diphenyl (2-ethylhexyl) phosphate
ECHA	European Chemicals Agency
EDAP	Ethylene diamine-o-phosphate
EEE	Electric and electronics equipment
EoL	End of life
EP	Ethylene propylene rubber
EPR	Ethylene propylene rubber

EPS	Expanded polystyrene
EU	European Union
EVA	Ethylene vinyl acetate
FR	Flame retardant
GF	Glass fibre
HIPS	High Impact Polystyrene
HIPS/PPO	High Impact Polystyrene/Poly(p-phenylene oxide)
HTPA	High temperature polyamide (Nylon)
LDPE	Low density polyethylene
LLDPE	Linear low-density polyethylene
MC	Melamine cyanurate
MDH	Magnesium hydroxide
MPP	Melamine polyphosphate
MPyP	Melamine Pyrophosphate
PA	Polyamide
PA6	Polyamide 6 (Nylon)
PA66	Polyamide 6,6
PAPP	Piperazine pyrophosphate
PBT	Polybutylene terephthalate
PC	Polycarbonate
PE	Polyethylene
PET	Polyethylene terephthalate
PF	Phenolic resin
PIN FR	Phosphorous Inorganic Nitrogen flame retardants
PIR	Polyisocyanurate
PMMA	Poly(methyl methacrylate)
PP	Polypropylene
PPE	Polyphenylene ether
PS	Polystyrene
PU	Polyurethane

PUR	Polyurethan
PVA	Polyvinyl alcohol
PVC	Polyvinyl chloride
RDP	Resorcinol diphenyl phosphate
REACH	Registration, evaluation, authorisation and restriction of chemicals
RP	Red phosphorus
r-PUR	Rigid polyurethane
r-PVC	Rigid polyvinyl chloride
SVHC	Substance of very high concern
TCP	Tricresyl phosphate
TCPP	Tris(2-chloropropyl) phosphate
TEP	Triethyl phosphate
TOP	Tris(2-ethylhexyl) phosphate
TPE	Thermoplastic elastomers
TPP	Triphenyl phosphate
TPU	Thermoplastic polyurethane
TRL	Technology readiness level
UP	Unsaturated polyesters
VE	Vinylester
WEEE	Waste electric and electronics equipment
w/o	Without
XLPE	Cross-linked polyethylene

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CONTENT OF THIS REPORT

Several European Commission initiatives aim to achieve the objectives of the EU Green Deal Circular Economy Action Plan, the Sustainable Chemical Strategy, the Sustainable Products Initiative and the Zero Pollution Action Plan: all of them affecting recycling. The sector of Phosphorus, Inorganic and Nitrogen Flame Retardants (PIN FR) producers is increasingly confronted with the requirements raised around plastic recycling within the EU.

This report explores holistically some key topics concerning PIN FRs and their impact on polymers recycling:

- **CHAPTER 1 – PIN FLAME RETARDANTS: MOTIVATION AND MARKET TRENDS:** Why are PIN FRs relevant today and what are the market trends that will make them more relevant in the next five years?
- **CHAPTER 2 – PIN FLAME RETARDANTS FOR DIFFERENT PLASTICS: INDUSTRIAL APPLICATIONS MATRIX;** What are the industrial sectors where PIN FRs are used the most? Are there enough public data to track this information, identifying the presence of PIN FR in plastics for different applications?
- **CHAPTER 3 - PIN FLAME RETARDANTS AND RECYCLING: AN R&D AND TECHNOLOGY OUTLOOK;** What is the R&D around PIN FR focussing on? Is it up to date with the most recent recyclability targets for plastics? Who are the key stakeholders/experts in the value-chain participating in R&D initiatives?
- **CHAPTER 4 - MEASURING PIN FR's IMPACT ON RECYCLING: A RECYCLING MATRIX MODEL;** Can we assess PIN FR's impact on plastics recyclability based on recent R&D cases? What is the best way to correctly collect this information?
- **CHAPTER 5 - WEEE PLASTICS AND PIN FRS: RECYCLING TREATMENT SHOWCASES;** What is the status of R&D for WEEEs streams when PIN FRs are involved?
- **CHAPTER 6 -TALKING TO EXPERTS;** What do polymers recycling experts think and know about PIN FRs impact on recycling and the available technologies?
- Finally, **CHAPTER 7 provides wrap-up conclusions and recommendations.** Notably, the document also includes 4 annexes that elaborate further details on the PIN-FRs Applications (**ANNEX-1**) , and provide methodological details (**ANNEX 2-4**)

EXECUTIVE SUMMARY: KEY RESULTS

PIN FRs: a market opportunity to meet EU targets obligations for plastic recycling

PIN FRs are non-halogenated Phosphorus, Nitrogen and Inorganic mineral-based flame retardants (including PIN fire safety synergists, smoke suppressants and intumescent coatings). They are gaining more and more market shares, becoming a leading segment in the FRs' market. Their worldwide consumption amounted to more than 2.39 million tons in 2019, driven by the increasing trend towards substituting legacy halogenated FRs with more sustainable non-halogenated products.

Several regulations like REACH or RoHS promote PIN FRs as an alternative to BFRs. But, to seize the opportunity, they need to support the upcoming framework of policy measures for plastic recycling.

The shift towards PIN FRs has been supported by increasing regulations limiting the use of hazardous materials such as the REACH[1], the WEEE[2] or the RoHS[3] Directives in Europe, with similar frameworks adopted in other regions. These regulations aim to improve the protection of human health from chemicals, make sure that electronic waste is properly recovered and recycled, and new equipment does not contain problematic substances. This has increased the interest in PIN FRs, which can combine fire safety with better health and environmental profiles.

[1] Directive 1907/2006 on restriction on Chemicals
[2] Directive 2002/96/EC on Waste of Electric and Electronic Equipment
[3] Directive 2002/95/EC on Restriction of certain hazardous Substances in Electric and Electronic Equipment

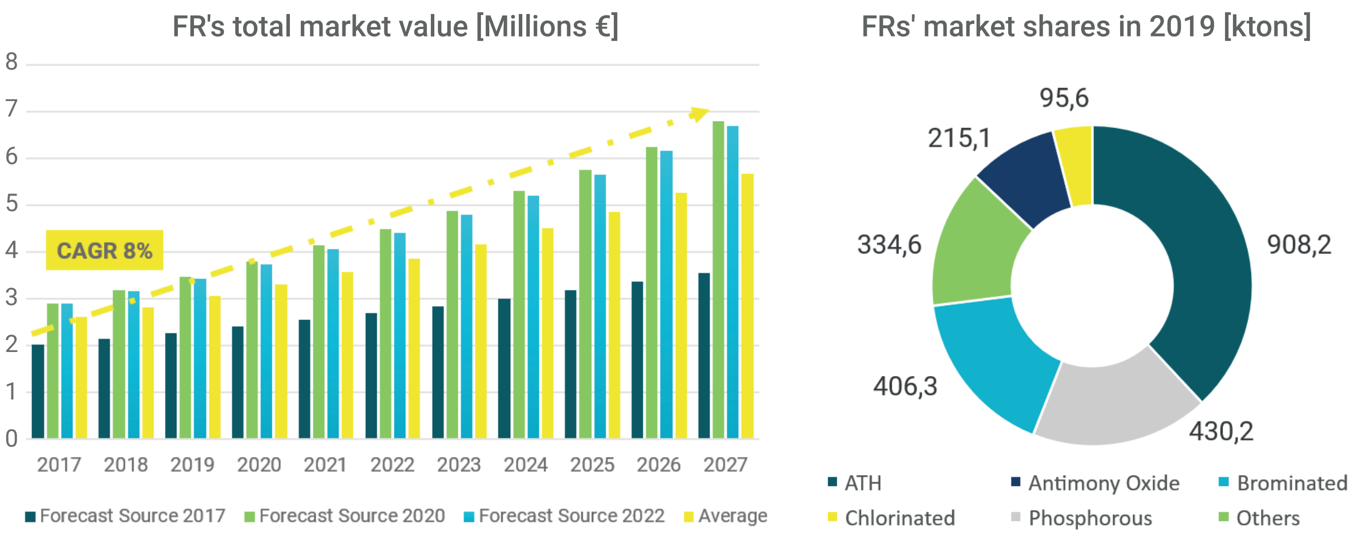


Exhibit 1: Market Forecasts and 2019 shares by FR type

To seize the market opportunity, PIN FRs must support the incoming challenges of plastic recycling.

Regulatory targets of the recently revised waste directives are 10% max landfilling of municipal waste by 2035, 50% recycling of plastic packaging by 2025 and 55% by 2030[1].

The European Commission has set very ambitious circularity objectives for plastics. In 2018 the *EU Strategy for Plastics*[4] in the Circular Economy was issued, establishing the main goals for plastic design, manufacture, use, re-use, and end-of-life management by 2030. Correspondingly, a Circular Plastics Alliance (CPA) was launched: **the target is to bring the EU market for recycled plastics to 10 million tonnes by 2025, which means providing a boost of an additional 3.4 million tonnes of recycled plastics compared to 2020.** An increase in sorting capacity by at least 4.2 million tonnes by 2025 and recycling capacity by at least 3.8 million tonnes, is expected, corresponding to estimated investment needs between € 7.6 billion and € 9.1 billion.

A thorough literature-based analysis in this report confirmed three main major market areas of PIN FR: electric and electronics (E&E) equipment (EEE), building and infrastructure as well as transport (Chapter 2): forty (40) different PIN FR types and more than fifty-four (54) different plastic types or combination of plastics were identified in our analysis. **3 Application Matrices have been built (Chapter 2)**

[1] Directive 1907/2006 on restriction on Chemicals
[4] https://ec.europa.eu/environment/circular-economy/pdf/plastics-strategy-brochure.pdf

EEE
Cables
Connectors and switchers
Electrical components
Engineering plastics for household appliances

Building and Infrastucture
Cable trays, skirting boards
Cables
Castings, Coatings
Façade Decoration
Flooring
Insulation
Laminate structures, Pultruded profiles
Laminates, Panels, Adhesive layers, Tubes/pipes (filament winding)
Pipes
Profiles - window, doors, trim

Transport
Cables
Ceilings, Sidewalls, Panels, Structural Parts
Coatings
Flooring
Insulation
Interior parts
Sealants
Seats

Exhibit 2: Application of PIN FR for the three most important market areas (EEE, Building & Infrastructure, and transport)

Looking at these sectors, in practical terms the "*10 Mt target*" implies an increase in the final recycled outputs by 40% for the automotive sector (part of the transport industry), 15% for the building sector and 52% for the EEE sector, which provide evidence of the efforts and investments needed in a very short amount of time. Furthermore, the CPA has defined a list of 26 recyclable products (by 2025), which includes plastics types related to PIN FRs in the EEE and building sectors.

Sector	Ref. Year	European converters Plastic Demand	Tonnes of Plastic waste collected	Tonnes sorted for recycling	Percentage Sorted for recycling	Tonnes of recyclate produced in the EU	Percentage recyclate production out of sorted for recycling	Legislative Framework	Untapped waste collection & Sorting (Sent to recyclers)	Untapped recycling ouptut	Increase of recyclates output
Automotive	2019	5.100.000	1.500.000	350.000	23%	150.000	43%	ELV Directive	140.000	60.000	40%
Building & Infrastructure	2018	10.137.600	1.746.000	450.000	26%	340.000	76%	Country-based	260.000	50.000	15%
EEE (households only)	2016	1.749.030	752.500	717.589	95%	561.373	78%	EEE Directive	370.000	290.000	52%

Exhibit 3: Overview of waste quantities sorted for recycling today and the untapped potential for recycling to be reached by 2025 as estimated by CPAs industry groups [absolute numbers represent tons/year]

PIN FRs' R&D shares the technological challenges of plastics recycling: emerging technologies are on their way to industrialisation, with still challenges ahead

For this report an R&D trends analysis was completed (Chapter 3[5]). It consists of a technology intelligence looking at a mixed corpus of R&D (public funded) projects, patents and scientific literature (reports, papers) enabling insights into the most relevant research and innovation initiatives related to applications of polymers containing PIN FR, and their recycling technologies, resulting in an overview of the key players and stakeholders. A series of interviews with 8 experts have complemented the data-driven investigation.

There is momentum for PIN FR R&D initiatives focussed on recycling. Yet, they are reasonably a smaller subset compared to brominated or antimony recycling, development and production of PIN FRs or polymers containing PIN FRs, BFRs replacement and development of bio-based plastics containing PIN FRs.

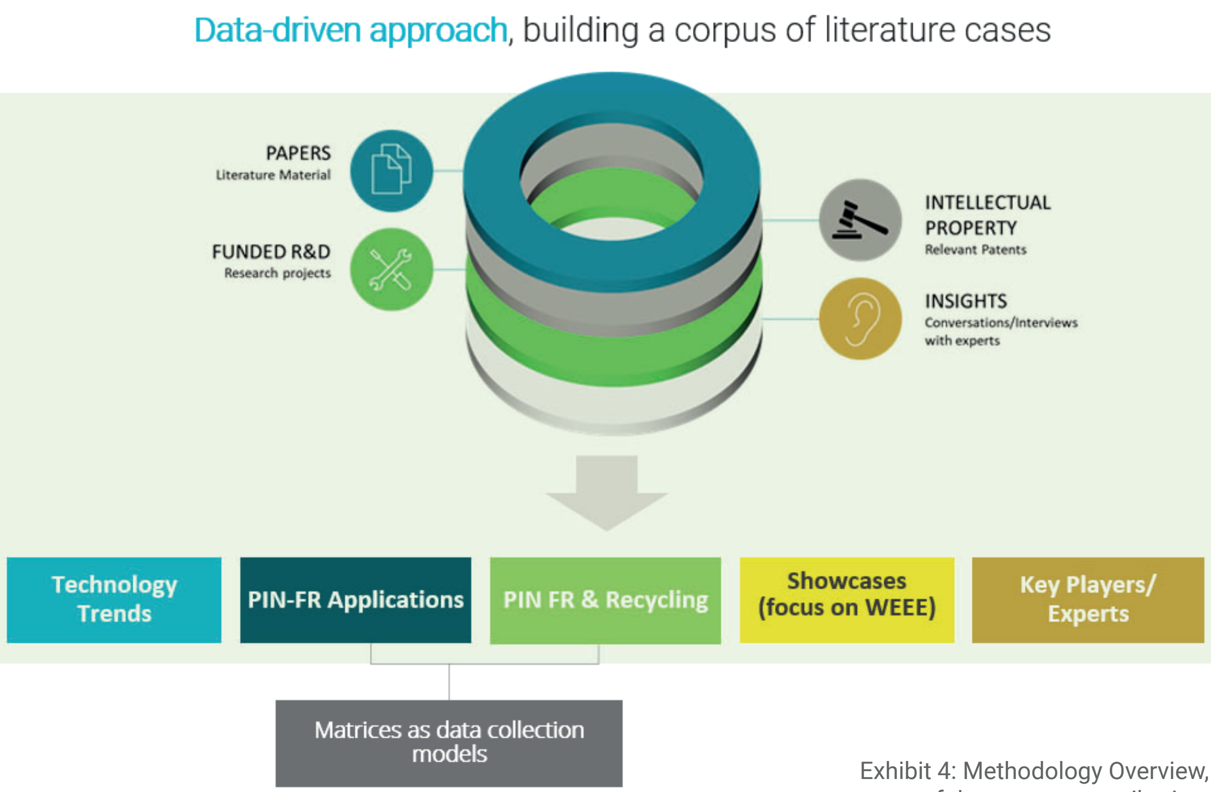


Exhibit 4: Methodology Overview, corpus of documents contribution to the report

[5] For methodology details the reader can refer to ANNEX-3

The **emerging R&D traits** from this analysis confirm that:

- The **primary focus by industry and R&D projects is recycling the polymer, not the PIN FR**, avoiding its downgrading
- **Most R&D cases focus on WEEEs** (especially from cables), which justifies further attention to illustrate relevant cases concentrated on this application (Chapter 5).
- Among PIN FRs, **Phosphorus-based flame retardants are the most considered ones**. Most of them derive from post-consumer plastics from the EEE sector (mainly based on PC/ABS blend or ABS). Specific references to bisphenol A, triphenyl phosphate (TPP), or ammonium polyphosphate (APP) were found, as well as inorganic FR including Aluminium tri-hydroxide (ATH) and magnesium hydroxide (MDH).

3 technologies are clearly represented in current R&D where PIN FR recycling is considered: Mechanical, Chemical and Solvent-Based recycling, while sorting technologies appear as an essential cornerstone to improve the downstream recycling process.

Year	Recycling Type				Waste Source					Flame Retardant Category		
	Mechanical Recycling	Chemical Recycling	Solvent-based Recycling	Waste to Energy by Pyrolysis	WEEE	Transport	Building and Construction	Post consume, not specified	Post consume (Packaging)	Phosphorus based	Inorganic Based	Nitrogen Based
2005	0	1	0	0	0	0	0	1	0	0	1	0
2006	0	0	0	0	0	0	0	0	0	0	0	0
2007	1	0	0	0	0	1	0	0	0	0	1	0
2008	0	0	0	0	0	0	0	0	0	0	0	0
2009	0	0	0	0	0	0	0	0	0	0	0	0
2010	1	0	0	0	0	0	0	1	0	0	1	0
2011	0	0	0	0	0	0	0	0	0	0	0	0
2012	3	2	1	0	3	2	0	0	1	6	2	0
2013	3	1	0	0	4	0	0	0	0	4	0	0
2014	3	0	0	0	2	0	0	1	0	0	1	1
2015	0	1	0	0	0	0	0	0	0	0	0	0
2016	1	0	0	0	0	1	0	0	0	0	1	0
2017	0	1	0	1	0	0	0	1	1	2	0	0
2018	5	0	0	0	3	2	1	2	1	3	3	1
2019	1	0	0	0	0	0	0	0	1	1	0	0
2020	3	0	1	0	2	1	0	1	0	3	0	0
2021	2	1	0	0	0	0	1	0	2	3	0	0
2022	0	0	1	0	1	0	0	0	0	1	0	0
2023	0	0	1	0	0	0	0	1	0	1	0	0

Exhibit 5: PIN FRs Polymers Recycling Heatmap by Combined Results from Literature, Projects, and Patents Analyses

Mechanical Recycling is an established technology with evident limitations. Looking at R&D literature, it has also been the most used and investigated process for the recycling of PIN FRs polymers in the last decade. Different techniques have been used to mechanically separate and recycle polymers containing PIN FRs (e.g., electrical separation combined with the melt-filtering). Achieving good results in this case is usually hindered by technological problems related to the sorting phase. Therefore, a process that combines .different plastic sorting technologies is preferable. After the sorting phase, we found density-based separation to be the most considered technique, upstream of the mechanical recycling of PIN FRs polymers. Looking at more recent techniques, in a recent patent (2020) CO_2 is used as an extraction agent to recover and analyse the flame retardant in a supercritical fluid, therefore presenting CO_2 as a mean to recover pure PIN FRs.

Examples: in Showcase 1 from Chapter 5, KU Leuven and Campine NV,in 2019, assessed the technical feasibility of dismantling-based recycling strategies for the WEEE plastics (LCD TVs back covers). This assessment provided interesting conclusions on the sorting technologies capable of differentiating different plastics containing brominated and phosphorus flame retardants. According to the authors, sorting trials with a LIBS contact measurement showed that this technology can detect plastic types and distinguish between Br and Phosphate Flame Retardants in LCD TV back cover materials during the sorting phase. For the study they considered a Quantum LIBS Scanner developed by Bertin Technologies in Belgium. A recycled PC/ABS with PFR was obtained, suitable for direct re-application in electronic products, having good mechanical, and aesthetical properties as well as recovered flammability.

With investments announced by Plastic Europe increasing from EUR 2.6 billion in 2025 to EUR 7.2 billion in 2030, **Chemical Recycling** appears to be the **complementing technology of the near future**. Accordingly, in this study we have found relevant PIN FR-related chemical recycling process investigations by industry in patents to recover and reuse phosphorus-based flame retardants (especially TPP). Furthermore, interesting studies have been carried out considering the pyrolysis process, where it is possible to recover pure PIN FRs if the temperature range is medium-low, beyond which the PIN FRs start to decompose. On the other hand, much of the literature on chemical recycling is still related to the recovery of BFRs.

Examples: in Showcase 7, six patents have been identified concerning the possibility of separating pure PIN FR from recycled plastics. These patents belong to SABIC (3), Anhui Chaoyue Environmental Protection Technology Company (1), University of Science and Technology of China (1) and Panasonic.

Solvent-based recycling appears to be most highly investigated **emerging technology** in the last years; the process proves to be **an excellent solution to recover pure PIN FR**. Different solvents are considered in the selected literature and patents: MeOH and EG are reported to remove TPP from PS and ABS; DMCHA solvent to remove BDP from PC/ABS; xylene solvent to remove aluminium tri-hydroxide (ATH) from PE and PP.

Examples: in Showcase 3 from Chapter 5, the University of Massachusetts Lowell carried out a study in 2021 on Recycling of Mixed Plastics in Electronic Waste Using Solvent-Based Processing. The feedstock was hand-sorted into the following categories: plastics, printed circuit boards, metals, wires, rubbers, metals, and capacitors. For this study's actual solvent-based recycling process, only the hand-picked polymer fraction of the obtained ESR (i.e., pre-sorted by the E-waste recycler) was used. They showed a significant removal efficiency of Phosporous; and 70% and 94% of PFR (TPP) was removed when using MeOH or EG as the anti-solvent, respectively.

The market of compatibilisers, stabilisers and other recycling additives is certainly a key element to be considered for the future of PIN FR plastics recycling, even if not directly affecting PIN FRs. Additives specifically adapted for recycling can limit the impacts of polymer degradation during re-extrusion, improve reprocessing (lubricants, melt stabilisers) and provide final material stabilisation (including antioxidants, anti-UV, acid-scavengers). They are applied in different plastic types, from low to high end: commodity (PE, PP, PS, PPE), engineering and high-performance plastics (HPA). It is to be noted that Flame Retardants are also mentioned as additives.

Examples: in our selected Showcase 8 (Chapter 5), PIN FRs are added to different polymers (PE, PE/EVA and PC/ABS) during the recycling process, providing final good flame retardancy properties to the recycled plastic (WEEE).

This study acknowledges PIN FRs to be the polymers design choice to support recycling. However, there are evident and reasonable information gaps, compared to BFR. A systematic strategy for comprehensive data and knowledge collection and sharing related to PIN FR is recommended.

What the experts say (Chapter 6): there is a general consensus that the effects of PIN FRs on recycling are not yet investigated enough, neither by industry nor academy. In short, the experts have confirmed that:

- PIN FR should be preferred since the design phase. However, even though they are considered as the cleanest flame retardants, BFR keeps being used because they are cheaper, historically established on the market, and because their FR-performance enables them to fulfill the requirement of many standards. This also justifies much more R&D research available with a focus on BFR.
- The presence of FRs in plastics can affect its use as a recycled material, as well as the approach to recycle it. In this sense the effects of the PIN FRs are still not completely known. In particular, latent acids or latent bases during recycling should be controlled to mitigate quality issues.
- While new technologies, such as chemical recycling, are being industrialised, mechanical recycling is cost and energy efficient, but it cannot deal well with the lack of quality in the input stream, thus, automatic identification using AI, sensor-based enhanced separation and purification are needed.
- There is not a specific regulation for PIN FR and this can create uncertainties for recyclers and contributes to keep the focus on BFR during the material selection. Besides, simple and unique standards for FRs, including screening and testing procedures are needed.

Therefore, a systematic strategy for the collection of relevant data on the characterisation and impact of PIN FR on recycling is required and recommended. An early data-collection approach and tool (the "PIN FR Recycling map") has been realised as part of this study, and used for preliminary qualitative analysis. It is proposed as a basis for further systematic data-collection on PIN FR recycling, **enabling a knowledge-based decision-making**. The *PIN FR Recycling map* allows for:

- Simplifying the collection and organization of information related to plastic recycling and possible PIN FR impacts on it.
- Supporting the understanding of current possibilities related to plastic recycling when PIN FRs are involved.
- Providing an intuitive, easy-to-use instrument based on a common taxonomy, including elements of the EC's Waste Hierarchy.

The resulting map keeps track of the diverse recycling process possibilities with a chosen plastic material substrate, containing a specific PIN FR. It is populated by R&D case studies, each of them describing a unique effort/technique/study to recover a single plastic material based on polymer, mixture, or resin. For each case, the matrix rebuilds the flow across the entire life cycle of a recycled polymer obtained from plastic-containing waste (Exhibit 6) and can therefore provide the information necessary for mapping the possible combinations in terms of waste type, polymer, FR content, treatment process and the fate of both polymers and the contained FRs.

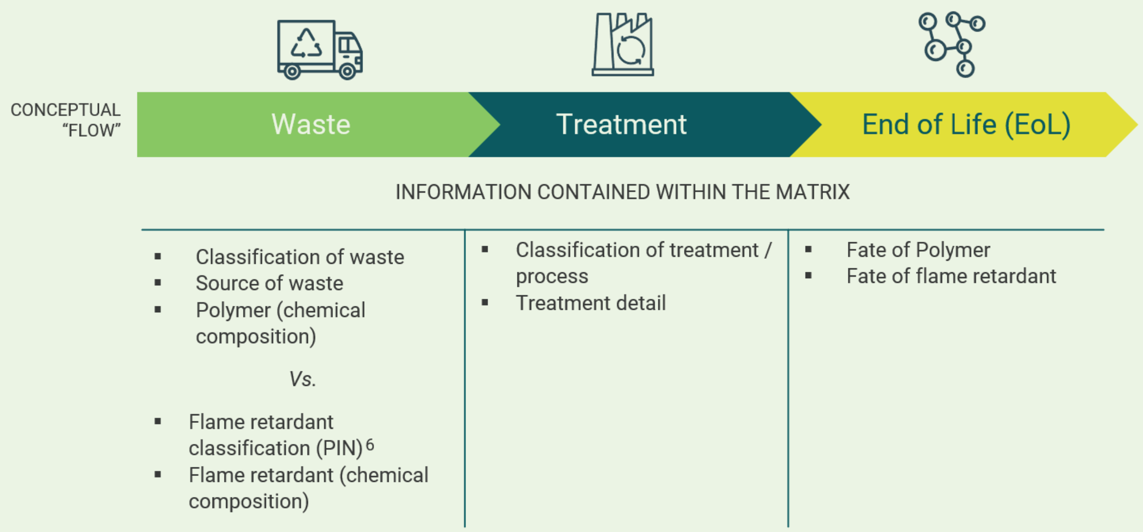


Exhibit 6: Conceptual flow used for the recycling matrix and map design

A preliminary observation of the case-studies in the recycling map tool (Chapter 4) is reported: the amount of data is still limited and doesn't allow for conclusions, but the bird's view of results in the following aims to provide a picture of the available information, and to use it to illustrate how this approach could be replicated with a systematic collection.

The initial dataset includes 30% of case-studies where a full recyclability of polymers is observed. Mechanical recycling still results as the "state of the art" for plastic waste processing. It is mentioned in the vast majority of the cases (50/69) followed with a lower order of magnitude by pyrolysis-based processes (7/69), solvent-based recycling (6/69), chemical recycling – depolymerisation (5/69) and waste-to-energy conversion process (1/69). However, Exhibit 7 and 8, where the available data have been clustered and visualised, also show that chemical recycling and solvent-based recycling have often been used when full recyclability is reported to be the final outcome of the case study. All in all, the available dataset suggests that a more heterogenous presence of FRs within plastic waste can have a larger impact on the final quality of the recycled material output.

[6] The present exercise includes only information concerning FRs. No information are yet available concerning other additives and including synergies, anti drip agents, compatibilizers as well as other agents, etc.

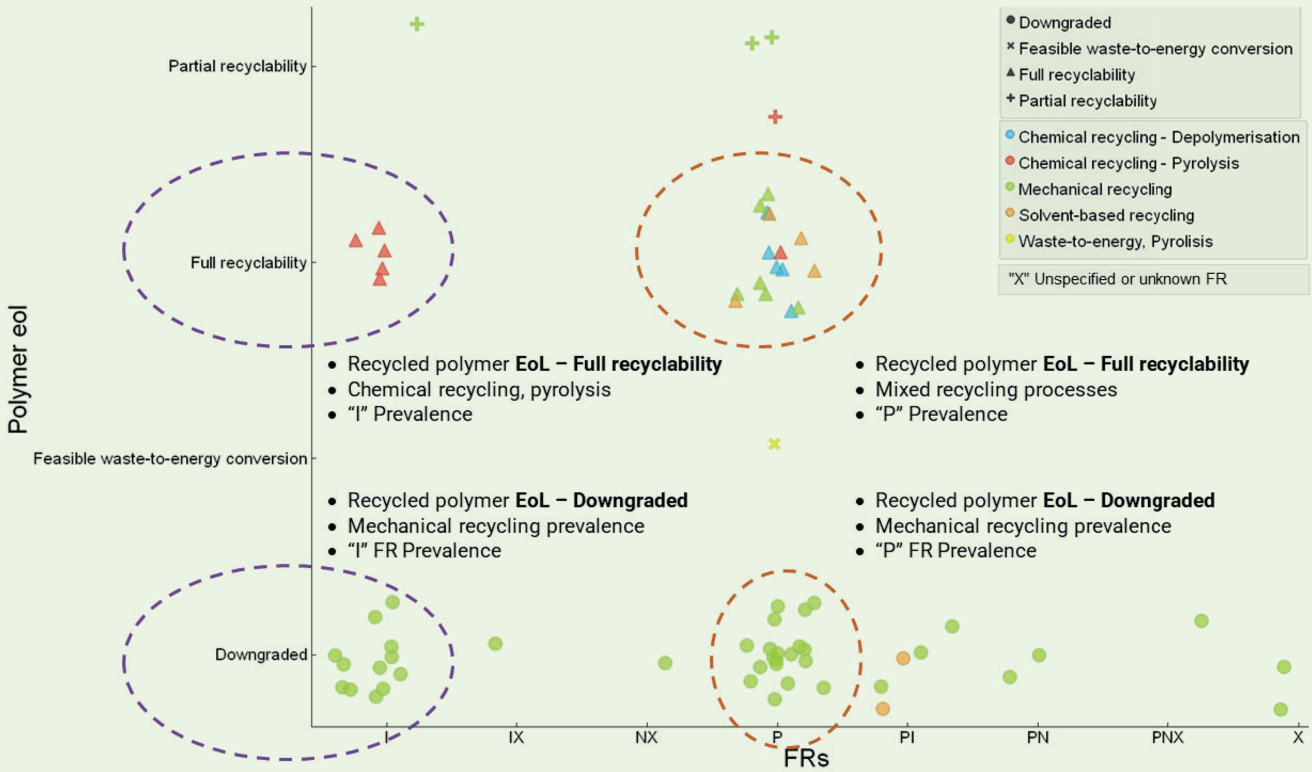


Exhibit 7: Example of data-analysis of PIN FR matrix performed via data mining– PIN FR type Vs recycled polymer fate (EoL) in the available dataset for different recycling processes

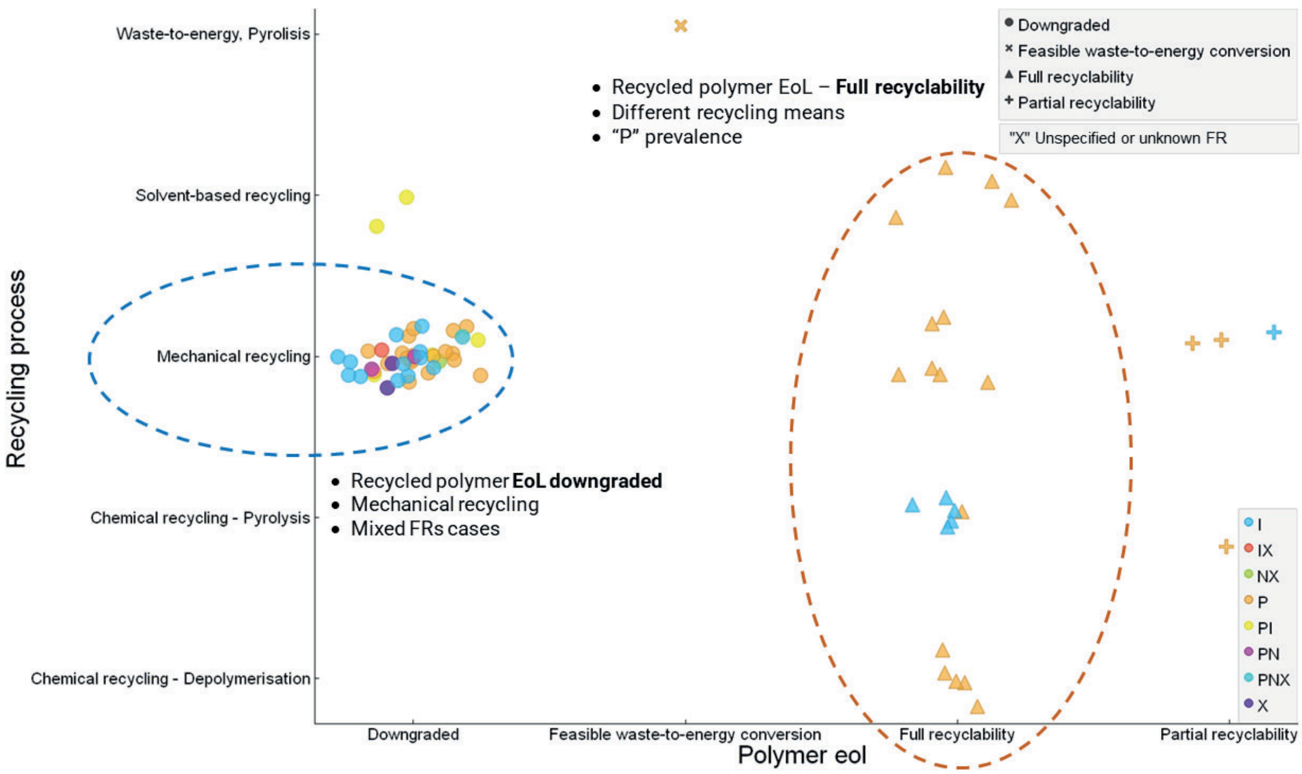


Exhibit 8: Example of data-analysis of PIN FR matrix performed via data mining means – Technology type Vs polymers recycling results

Recommendations

To improve insights, policy and decision-making on PIN FR recycling it is essential **to support a better knowledge-base and information exchange between the different parts of the production and recycling value-chains**. Key recommendations hence include:

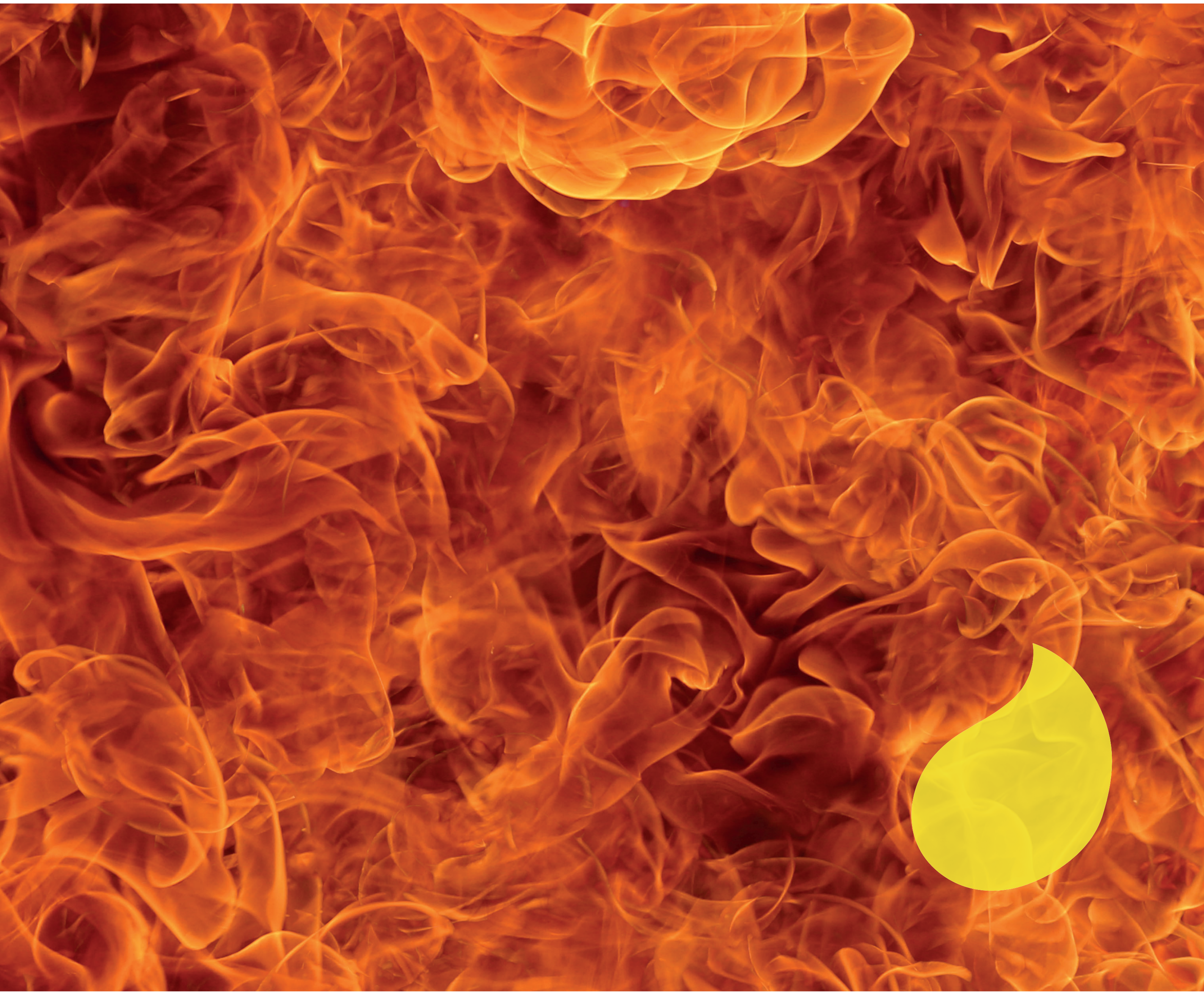
- **Improving the mapping and characterization of PIN FR content.** At the moment, this is still missing since recyclers seem to currently be only characterizing the bromine content. This approach should entail the definition of **specific testing and sampling procedures** to be standardised and the **creation of a centralised database.**
- **Support certainty of regulations about PIN FRs, avoiding redundancies if possible (one substance, one assessment).** This will push the industry and recyclers in particular to characterize PIN FRs on top of BFRs.
- **Fostering "sustainability by design"** schemes to enhance cooperation between producers and recyclers and supporting **FR standardisation** to help the recycling phase. It is to be noted that a similar approach is being followed by the *Circular Plastic Alliance*, which has been working to provide Design-for-recycling guidelines and address CEN and CENELEC standards. A list of 26 products exists with priority, which include the EEE and Building sectors (see Chapter 1).
- **Supporting the private sector in enhancing transparency** – e.g., introducing recycling-oriented marketplaces for the private sector. Besides, some experts suggested **FR content labelling systems and "products passports"** for particular streams (e.g., WEEE).

Developments in FRs recycling are still largely dominated by BFR recycling. This means that there is a need to collect further data and knowledge on the impact of PIN FR on recycling. It is therefore recommended:

- **To take a systematic approach** (e.g., the "PIN FR recycling map") in building a **comprehensive database with PIN FR recycling cases**, to generate comprehensive data and knowledge on the possible relationships between the composition of PIN FR containing waste, recycling technologies, type of polymers, waste collection and separation, in order to support better insights and decision making.
- To invest in increasing the number of **projects to investigate specifically PIN FRs impact on the recyclability of different waste streams**. For example, it is recommended to address acid formation during the recycling process. Projects should include large-scale case-studies involving the whole value-chain. International cooperation is advised, to correctly represent the EU and extra EU value-chain, and also to better consider quality control differences, such as those in labelling and marking procedures.
- **Focus R&D on developing and upscaling novel separation, purification, and sorting technologies to enhance the impact of available treatment**

processes. AI-supported NIR and optical technologies are most promising and should be tested on a suitable scale. Chemical elements, that occur when PIN flame retardants are used could be detected, but the development and testing of specific sorting models would be required. It should be noted that similar technologies are being developed for adjacent recycling streams (e.g., batteries), which could again lead to building R&D synergies in larger projects.

- **To accelerate and upscale testing of solvent-based recycling, with a view to separate PIN FRs, in a way that it is possible to consider its economic sustainability and safety aspects**, given its growing relevance to obtaining highly pure recycled polymers in a cost-effective way



1 PIN FLAME RETARDANTS: MOTIVATIONS AND MARKET TRENDS

Why are PIN FRs relevant today and what are the market trends that will make them more relevant in the next five years?

1.1 What PIN FRs are and why they are relevant

With the technological evolution in the last decades, polymeric materials have rapidly replaced metals and ceramic materials in various applications, due to their remarkable combination of properties like low weight, easy fabrication and low processing temperature (1) (2). **The high flammability of polymers and polymer composites tends to limit their applications though, and more stringent requirements have been introduced for fire safety concerns.**

Reducing the fire hazards accompanied by using polymeric materials can be achieved by incorporating FRs. Their main functions are to provide sufficient time for people to escape by delaying the time of flashover, and reducing smoke (3). The challenge is to develop an FR system that enhances the fire performance of polymeric composites without deteriorating their mechanical and other application-related properties.

Historically, conventional FR formulations were based on halogenated substances, but they were (and are being) banned as they evolve additional toxic gases during combustion (4). This has been providing increasing opportunities for PIN FRs to replace them. PIN FRs emerged as an alternative to Brominated Flame Retardants (BFRs) when these became a topic of environmental concern in the early 1990s, when it was discovered that some BFRs could form halogenated dioxins and furans under severe thermal stress or when they were burnt in accidental fires or uncontrolled combustion (5) Findings in the environment and biota and the suspicion that some FRs bio accumulate in organisms have added to these concerns (6), (7). Meanwhile, the environmental and health properties of BFRs and other types of FRs such as PIN FRs have been studied extensively to determine which FRs could be a valid alternative in terms of efficacy and environmental impact, regardless of their chemistry.

PIN FRs are non-halogenated FRs[7] (HFFR). Nowadays, they are found in a wide range of sectoral applications (see Chapter 2), including Electronic and Electric Equipment, Building & Infrastructure, or Transport. A whole toolbox of (non-halogenated) PIN FR chemistries is now available.

These chemistries are designed to respond to current expectations regarding sustainable fire safety with environmentally friendly FRs (5), (8), (9). They cover a diverse range of chemicals, which are commonly classified as:

- **Phosphorus based FR** include inorganic phosphorus FRs such as red phosphorus and ammonium polyphosphate, or organic phosphorus FRs (organophosphates) such as phosphate esters, phosphonates and metal salts of organic phosphinates. These are characterized by being less impacting than BFR (or completely harmless in some cases), with limited toxic gases evolved during combustion, and can achieve good FR properties with lower loading (weight-% 10 to 20) compared to inorganic-based FR. They are relatively more expensive than other FRs (10) (11). Organophosphorus chemicals are the second most widely used FR (12). They can be used in numerous applications such as textiles, polyurethane (PU) foams, coatings, and rubber (11).
- **Inorganic based FR** include aluminium tri-hydroxide (ATH), magnesium dihydroxide (MDH), aluminium oxide hydroxide – Boehmite (AOH) and calcium/magnesium carbonates. They are characterized by being non-toxic and smoke-reducing, while at the same time being less expensive than organophosphorus FRs. On the other hand, higher amounts are required to be effective (weight-% 30 up to 70). Among inorganic FR, Aluminium hydroxide is the most used FR. It represents almost 40% of FR consumption (13).
- **Nitrogen based FR** include melamine and melamine compounds such as melamine cyanurate, melamine polyphosphate, melamine poly (zinc/ammonium) phosphate, amongst others. They are characterized by low toxicity and low evolution of smoke.
- **Heterogeneus PIN FR**: PIN FR may exist as a combination of various P and N, or P and I based compounds (i.e., Ammonium polyphosphate + Melamine triazine, Melamine triazine + aluminum tri-hydroxide (ATH)) or in other molecules combining P, I, N chemical elements.

[7] As such, PIN FR are a subset of Halogen Free Flame Retardants (HFFR)

1.2 A Market Perspective

PIN FRs are more and more considered the leading segment in the FRs' market. Their worldwide consumption amounted to more than 2.39 million tons in 2019 (12), driven by the increasing trend towards substituting legacy halogenated FRs with more sustainable non-halogenated products.

Figure 1-1 shows the world consumption of different FR types by volume in 2019: aluminium hydroxide is the largest single FR with a 38% share (ca. 900 ktons).
The second place is held by the halogenated FR systems, comprising brominated and chlorinated products which are commonly used together with the synergist antimony trioxide, adding up to 30% (17% brominated, 9% antimony oxides and 4% chlorinated). Organophosphorus and other FRs like e.g., inorganic phosphorus compounds, nitrogen and zinc-based FRs make up the rest at 32% (18% organophosphorus, 14% other FRs).

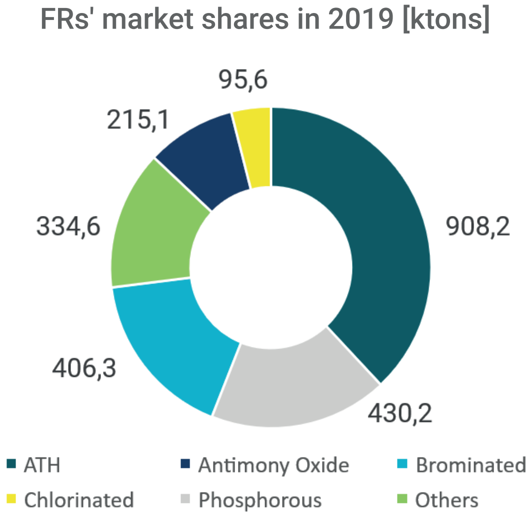


Figure 1-1: World consumption of flame retardants by type, 2019

DRIVERS

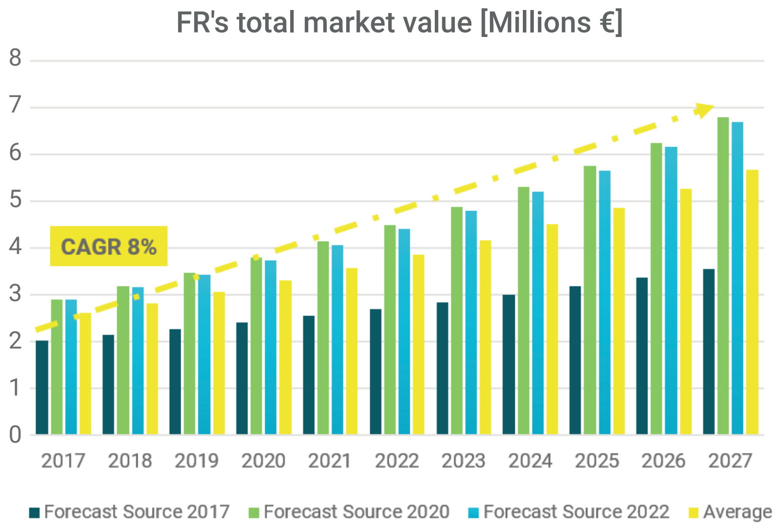
The shift towards PIN FRs has been supported by increasing regulations limiting the use of hazardous materials such as the REACH[8], the WEEE[9] or the RoHS[10] Directives in Europe, with similar frameworks adopted in other regions. These regulations aim to improve the protection of human health from chemicals, make sure that electronic waste is adequately recovered and recycled, and new equipment does not contain problematic substances. This has increased the interest in PIN FRs, which combine fire safety with better health and environmental profiles.

By sector, the building & infrastructure industry has intensified the adoption of PIN FRs for meeting safety standards along with the extensive use of polymers. This is driving the market growth through the **accelerated construction activities being undertaken globally**. The **electronics and transportation industries** are identified as fast-growing acceleration lanes for the market in the next few years.

[8] Directive 1907/2006 on restriction on Chemicals
[9] Directive 2002/96/EC on Waste of Electric and Electronic Equipment
[10] Directive 2002/95/EC on Restriction of certain hazardous Substances in Electric and Electronic Equipment

While aluminium hydroxide accounted for the largest market share by volume, organophosphorous led the market in terms of value in 2019 and is also expected by analysts to register the highest CAGR - Compound Annual Growth Rate – in the next 5 years. Their advantage when used in polymer blends is the lower loading level, making them a preferred choice for different applications (e.g., engineering plastics, polyurethane foams, or textiles). In terms of applications segments, **polyolefins are estimated to be pushed by consumer-centric industries**, due to insulating properties and higher thermal and chemical resistance.

It is worth noting that market analysts' growth projections have been increasing in the last 4 years, leading to more recent forecasts of close to a 6 Billion Euro market value and an 8% CAGR by 2027 (Figure 1-2).



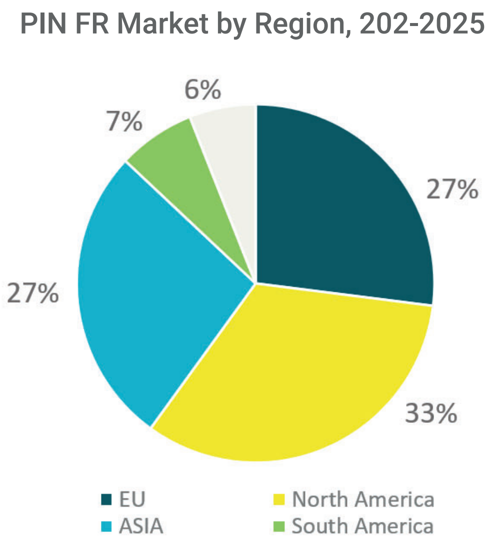


Figure 1-2: Market growth trends as reported by different secondary sources [11](left); Market by region (right)

CHALLENGES

While more stringent regulations pushed the use of PIN FR leading to a better product, **some challenges are still ahead limiting their usage so far. Generally speaking, these include relatively high loading levels, required for some applications and cost**. In fact, PIN FRs should be incorporated at higher loading percentages to be effective, which can alter the mechanical properties (14), (15). In turn, the high loading percentages influence the resin processability. For example, the added particles increase the viscosity or the curing time for the resin, which leads to adjusting the processing conditions (4) (16). All in all, recycling plastics containing PIN FRs is a complex topic that involves an interplay of challenges.

[11] The average market value and growth rate have been evaluated based on 3 different market reports with forecasts from 2017, 2020 and 2022. For a better visualisation, the yearly growth rates of each source have been used to obtain yearly market values over the same timeframe for each forecast.

2 PIN FLAME RETARDANTS FOR DIFFERENT PLASTICS: INDUSTRIAL APPLICATIONS MATRIX

Where are PIN FR used the most?
Are there enough public data to track this information?

2.1 Application areas of PIN flame retardants

A thorough literature-based analysis assessed three main major market areas of PIN FR: electric and electronics (E&E) equipment (EEE), building and infrastructure as well as transport.

EEE	Building and Infrastucture	Transport
Cables	Cable trays, skirting boards	Cables
Connectors and switchers	Cables	Ceilings, Sidewalls, Panels, Structural Parts
Electrical components	Castings, Coatings	Coatings
Engineering plastics for household appliances	Façade Decoration	Flooring
	Flooring	Insulation
	Insulation	Interior parts
	Laminate structures, Pultruded profiles	Sealants
	Laminates, Panels, Adhesive layers, Tubes/pipes (filament winding)	Seats
	Pipes	
	Profiles - window, doors, trim	

Table 2-1: Application of PIN FR for the three most important market areas (EEE, Building and infrastructure and transport)

The main applications that require FRs to be used in polymers for the three major market areas are summarized in Table 1 2. The use of polymers with PIN FR is prominent in the E&E industry, such as in cables, connectors, switchers, electrical components, or engineering plastics for household appliances. Most common examples of such products are televisions, printers, and computer cases. Polymers with PIN FR in the building and infrastructure industry are applied in cable trays, skirting boards, cables, castings, coatings, facade decoration, flooring, insulation, laminate structures, pultruded profiles, laminates, panels, adhesive layers, tubes/pipes (filament winding), pipes, profiles- window, doors and trim. Finally, the use of polymers with PIN FR in the transport industry include cables, ceilings sidewalls, panels, structural parts, coatings, flooring, insulation, interior parts, sealants and seats.

As highlighted in the previous chapter, the consumption of FR has grown substantially in the past 4 years, notably in electronics, and will continue to grow. FRs are mainly consumed by the plastics/resin industry. Textiles and rubber products account for most of the rest. Asia consumed the most significant volume of FRs in 2019 with a %51 share, with China being the largest single consumer at (12) %27.

Forty (40) different PIN FR types and more than fifty-four (54) different plastic types or combination of plastics were identified in our analysis. A classification of the 40 different PIN FR identified is provided in Table 2 2.

It should be noted that this table does not include various other PIN flame retardants, synergists and smoke suppressants (e.g. clays/organo-clays, expanded graphite, metal oxides, specialist minerals) because data were not found concerning their implications for plastics recycling.

Table 2-2: PIN FRs are classified into the three main groups (Phosphorus, Inorganic, Nitrogen) and a combination of different groups (Phosphorus + Nitrogen or Inorganic + Nitrogen).

Phosphorus FR	Aluminium diethylphosphinate (DEPAL)
	Ammonium polyphosphate (APP)
	Bisphenol A Bis(diphenyl phosphate) (BDP)
	Cresyl diphenyl phosphate (CDP)
	Cyclic phosphonate
	Dimethyl propane phosphonate (DMPP)
	Diphenyl 2-ethylhexyl phosphate (DPO)
	6-oxide6H-dibenz(c,E)(1,2)oxaphosphorin (DOPO)
	Ethylene diamine phosphate (EDAP)
	Isopropylated phospate ester
	Piperazine pyrophosphate (PAPP)
	Phosphate esters
	Phosphorus polyol
	Red phosphorus (RP)
	Resorcinol bis(diphenylphosphate) / Resorcinol diphenyl phosphate (RDP)
	Tris(2-ethylhexyl) phosphate (TOP)
	Tricresyl phosphate (TCP)
	Triethyl phosphate (TEP)
	Triphenyl phosphate (TPP)
	tris(2-chloropropyl) phosphate (TCPP) *Not longer rated as PIN since it is a "Clorinated P" FR, (P according to only few references)*
	Metal phosphinate
Inorganic FR	Aluminum oxide-hydroxide (AOH) / Bohemite
	Aluminum tri-hydroxide (ATH)
	Magnesium hydroxide / Magnesium dihydroxide (MDH)
	Zinc borate
	Zinc hydroxy stannates
	Sulphonates
Nitrogen FR	Melamine borate
	Melamine cyanurate (MC)
	Melamine pyrophosphate (MPyP)
	Melamine Polyphosphate
Phosphorus (P) + Nitrogen (N)	Ammonium polyphosphate + Melamine triazine
Inorganic (I) + Nitrogen (N)	Melamine triazine + aluminum tri-hydroxide (ATH)

The information provided in Table 2-1 and Table 2-2 was compiled and organized in 3 Application Matrices, one for each major application area for PIN FRs (EEE, building and infrastructure and transport). Each matrix contains the most used PIN FRs (rows on the left in the matrices), the applications for each market area and the most common plastic types (columns of the matrices) as shown in the below example (Figure 2-1). The number in each cell of the matrix indicates the weight (in weight-%) of PIN FR for each plastic types and application. Simple crosses were used when the weight was not reported in the sources analysed. Thus, the crosses in each cell of the matrix indicates the presence of PIN FR in different polymer types for different applications. Each matrix can be used as a mapping tool to help understanding the use and presence of PIN FRs in different types of plastics for different applications.

The 3 matrices are detailed in Figure 2-2, Figure 2-3 and Figure 2-4, while more exhaustive comments about their contents are included in the Annex-1 of this report. The following are the key take-aways:

- In the E&E industry, polymers with PIN FR are the most prominent used in cables, connectors, switchers, electrical components, and engineering plastics for household appliances.
- In the building and infrastructure industry, polymers with PIN FR are applied in cable trays, skirting boards, cables, castings, coatings, decoration, flooring, insulation, laminate structures, pultruded profiles, laminates, panels, adhesive layers, tubes/pipes (filament winding), profiles-window, doors, and trims.
- The use of polymers with PIN FR in the transport industry includes cables, ceilings, sidewalls, panels, structural parts, coatings, flooring, insulation, interior parts, sealants and seats.
- The most common identified plastic types in this analysis across the three major PIN FR's applications areas are: polyamide (PA), polybutylene terephthalate (PBT), polycarbonates/acrylonitrile butadiene styrene (PC/ABS), polyethylene terephthalate (PET), polypropylene (PP), polyvinyl chloride (PVC), polyethylene (PE), ethylene vinyl acetate (EVA), polyurethane (PU), epoxy resin, vinylester (VE), phenolic resin, thermoplastic polyurethane (TPU). Moreover, recycled PA6/PP has been used with some PIN FR like ammonium polyphosphate (APP), melamine cyanurate (MC) combined with melamine pyrophosphate (MPyP) and melamine polyphosphate (MPP).
- All in all, weight percentages are heterogeneous in ranges and public data about them have proven to be hard to find, with the largest amount of available information concerning Inorganic FRs and cables in particular. While the dosage of inorganic FRs can exceed 70 weight-%, the dosage of phosphorus and nitrogen FRs typically do not exceed 25 weight-%; it can even be lower depending on application case.

Numbers in the Cells represent wt.% values

* combined with P and/or N FR

		Cables											
		CPVC	CR	EVA	LDPE	Polyamides	Polyethylen co-butene	Polyethylene co-octene	PE/EVA	PP	PVC	Rubber	Silicon rubbers
Phosporus FR (P)	APP												
	APP+P-synergist									X			
	BDP												
	DEPAL												
	PAPP									X			
	Phosphate ester												
	Phosphonic acid ester												
	RP												
	RDP												
	TCP											X	
	TPP												
	TCPP*												
	tri (3-hydroxypropyl) phosphine oxide												
	Metal phosphinate												
Inorganic FR (I)	Aluminum trihydroxide (ATH)			50					X				
	AOH			3-15	3-15	X	3-15	3-15		X	ATH 5-15, ATO 1-3	X	X
	ATH (+ ATO)			58-67 OR 20-55*	58-67 OR 20-55*		58-67 OR 20-55*	58-67 OR 20-55*			ATH 5-15, ATO 1-3	X	X
	MDH			< 80	< 80		< 80	< 80	X				X
	Zinc borate	2-5	2-5	3-15	3-15		3-15	3-15			2-5		
	Zinc-Hydroxy-Stannates			3-15	3-15		3-15	3-15					
Nitrogen FR (N)	Alkoxyamine									X			
	MC					X				X			
	Melamine phosphate					X				X			
	Melamine polyphosphate												
Phosphorus (P) + Nitrogen (N)	Substitutedamine phosphatemixture									X			
	Metalphosphinate +N-synergist (e.g. MPyP)												
	Metalphosphinate + MPP												

Figure 2 2: Application matrix for PIN FR for EEE plastics applications

Numbers in the Cells represent wt.% values

* combined with P and/or N FR

EEE – Cables

FR group	FR	CPVC	CR	EVA	LDPE	Polyamides	Polyethylen co-butene	Polyethylene co-octene	PE/EVA	PP	PVC	Rubber	Silicon rubbers	TPU	TPE	XLPE
Phosporus FR (P)	APP															
	APP+P-synergist									X						
	BDP															
	DEPAL															
	PAPP									X						
	Phosphate ester															
	Phosphonic acid ester															10-20
	RP															10-20
	RDP															
	TCP											X				
	TPP															10-20
	TCPP*															10-20
	tri (3-hydroxypropyl) phosphine oxide															10-20
	Metal phosphinate															10-20
Inorganic FR (I)	Aluminum trihydroxide (ATH)			50					X							
	AOH			3-15	3-15	X	3-15	3-15		X	ATH 5-15, ATO 1-3	X	X	3-15	3-15	
	ATH (+ ATO)			58-67 OR 20-55*	58-67 OR 20-55*		58-67 OR 20-55*	58-67 OR 20-55*			ATH 5-15, ATO 1-3	X	X	58-67 OR 20-55*	58-67 OR 20-55*	
	MDH			< 80	< 80		< 80	< 80	X				X	< 80	< 80	
	Zinc borate	2-5	2-5	3-15	3-15		3-15	3-15			2-5			3-15	3-15	
	Zinc-Hydroxy-Stannates			3-15	3-15		3-15	3-15						3-15	3-15	
Nitrogen FR (N)	Alkoxyamine									X						
	MC					X				X				X		
	Melamine phosphate					X				X						
	Melamine polyphosphate															
Phosphorus (P) + Nitrogen (N)	Substitutedamine phosphatemixture									X						
	Metalphosphinate +N-synergist (e.g. MPyP)															
	Metalphosphinate + MPP															

EEE – Connectors and switchers

FR group	FR	HTN	HIPS/PPO	PBT	PBT/GF	PC/ABS	PET	PA6	PA66	PA6/GF30	PA66/GF30
Phosporus FR (P)	APP										
	APP+P-synergist										
	BDP					X	X				
	DEPAL									X	X
	PAPP										
	Phosphate ester					X					
	Phosphonic acid ester										
	RP								X		
	RDP		X			X					
	TCP										
	TPP					X					
	TCPP*										
	tri (3-hydroxypropyl) phosphine oxide										
	Metal phosphinate	X									
Inorganic FR (I)	Aluminum trihydroxide (ATH)										
	AOH			X	X			X	X	X	X
	ATH (+ ATO)										
	MDH										
	Zinc borate										
	Zinc-Hydroxy-Stannates										
Nitrogen FR (N)	Alkoxyamine										
	MC							X			
	Melamine phosphate										
	Melamine polyphosphate								X		
Phosphorus (P) + Nitrogen (N)	Substitutedamine phosphatemixture										
	Metalphosphinate +N-synergist (e.g. MPyP)			X	X		X	X	X		
	Metalphosphinate + MPP			X	X						

EEE – Electrical components

FR group	FR	HTN	PC/ABS	PBT	PET	PA6	PA66	PA6/GF30	PA66/GF30
Phosporus FR (P)	APP								
	APP+P-synergist								
	BDP		1-5						
	DEPAL							X	X
	PAPP								
	Phosphate ester								
	Phosphonic acid ester								
	RP						X		
	RDP		1-5						
	TCP		1-5						
	TPP		1-5						
	TCPP*								
	tri (3-hydroxypropyl) phosphine oxide								
	Metal phosphinate	X							
Inorganic FR (I)	Aluminum trihydroxide (ATH)								
	AOH								
	ATH (+ ATO)								
	MDH								
	Zinc borate								
	Zinc-Hydroxy-Stannates								
Nitrogen FR (N)	Alkoxyamine								
	MC								
	Melamine phosphate					X			
	Melamine polyphosphate						X		
Phosphorus (P) + Nitrogen (N)	Substitutedamine phosphatemixture								
	Metalphosphinate +N-synergist (e.g. MPyP)								
	Metalphosphinate + MPP			X	X	X	X		

EEE – Engineering plastics for household appliances

FR group	FR	HIPS/PPE	PC	PC/ABS	PIR	Polyamides	PU
Phosporus FR (P)	APP						
	APP+P-synergist						
	BDP	8.5-14					
	DEPAL						
	PAPP						
	Phosphate ester			X			
	Phosphonic acid ester						
	RP						
	RDP			8.5-14			
	TCP						
	TPP						
	TCPP*				X		
	tri (3-hydroxypropyl) phosphine oxide						
	Metal phosphinate						
Inorganic FR (I)	Aluminum trihydroxide (ATH)						
	AOH		X	X		X	
	ATH (+ ATO)						
	MDH						
	Zinc borate						
	Zinc-Hydroxy-Stannates						
Nitrogen FR (N)	Alkoxyamine						
	MC						
	Melamine phosphate						
	Melamine polyphosphate						
Phosphorus (P) + Nitrogen (N)	Substitutedamine phosphatemixture						
	Metalphosphinate +N-synergist (e.g. MPyP)						
	Metalphosphinate + MPP						

Figure 2 3: Application matrix for PIN FR for building and infrastructure applications

Numbers in the Cells represent wt.% values

* combined with P and/or N FR
** aluminum diethylphosphinate
*** melamine

		Building and infrastructure																																					
		Cable trays, skirting boards	Cables													Casting, coating and Façades		Flooring								Insulation								Laminate structures, pultruded profiles, adhesive Layers and tubes			Pipes		Profiles- window, doors, trim
		PP	CPVC	CR	EVA	EVA/PE	LDPE	Polyamides	Polyethylene co-butene	Polyethylene co-octene	PP	PVC	Silicone Rubbers	TPE	TPU	PU	PE/EVA	Elastomers	Epoxy Resin	PF	Polyolefins	PVC	UP	Urethane Elastomers	VE	ABS	PC	PF	Polyurethane foam	PIR	PU	r-PUR	PVC	UP	VE	Epoxy Resin	HDPE	PP	r- PVC
Phosporus FR (P)	Aluminium phospinate																							X															
	CDP																					X		X															
	DMPP																													X		X		X	X				
	DPO																					X																	
	APP															X			X					X						X		X		X	X	X			
	DOPO																		X																	X			
	EDAP	X																	X																				
	Isopropylated phospate ester																					X																	
	Phosphate esters																			X						X	X	X	X										
	Phosphorus polyol																															X							
	RP																													X									
	TOP																					X																	
	TCP																		X			X																	
	TEP																													X		X							
	TPP																							X															
	TCPP*																													X	X								
Inorganic FR (I)	AOH	X			3-15		3-15	X	3-15	3-15	X		X	3-15	3-15	X										X	X						X			X	X	X	X
	ATH (+ ATO)	X			58-67 OR 20-55*	tipically 60-70	58-67 OR 20-55*	X	58-67 OR 20-55*	58-67 OR 20-55*	X		X	58-67 OR 20-55*	58-67 OR 20-55*	X	X	X	X		X	X	X		X								X	X	X	X	X	X	X
	MDH	X			< 80	tipically 60-70	< 80	X	< 80	< 80	X		X	< 80	< 80		X	X			X												X				X	X	X
	Zinc borate		2-5	2-5	3-15		3-15		3-15	3-15				3-15	3-15							X											X						X
	Zinc hydroxy stannates				3-15		3-15		3-15	3-15				3-15	3-15							X																	X
Nitrogen FR (N)	Melamine borate																		X	X																			
	MC							X			X												X	X															
	Melamine phosphate	X																					X	X												X			
	Melamine polyphosphate	X						X																X												X			
Phosphorus (P) + Nitrogen (N)	Aluminum diethylphosphinate +Melamine																											10** AND 3***											
	Ammonium polyphosphate + Melamine triazine										20																												
Inorganic (I) + Nitrogen (N)	Melamine triazine + aluminum hydroxide					X																																	
	Melamine triazine + ATH					X																																	

Figure 2 4: Application matrix for PIN FR for transport applications

Numbers in the Cells represent wt.% values

		Transport: Cables						Ceilings Sidewalls, Panels, Structural Parts					Coatings			Flooring						Insulation		Interior parts											Sealants					Seats			
		EVA	LDPE	Polyamides	PVC	Silicone Rubbers	TPE/TPU	Acrylate Resins	Epoxy Resin	Phenolic resin	UP Resin	VE Resin	2K-PU	Acrylates	Epoxy resin	Elastomers	Epoxy Resin	PF	Polyolefins	PVC	Urethane Elastomers	r-PUR	PIR	ABS	Epoxy	PA6	PC	PC/ABS	PE	PET	PU	Recycled PA6/PP	TPU	UP	Acrylics	Elsatomers	Epoxy resin	PU	PVC	PA6	PU	TPU	Recycled PA6/PP
Phosporus FR (P)	APP							X	X		X	X	X	X	X		X				X	X	X									25	X	X	X	X	X	X	X				25
	Aluminium phospinate																				X																						
	BDP																							X				X															
	CDP																			X	X																						
	Cyclic phosponate								X																																		
	DMPP								X		X											X	X											X									
	DOPO								X								X																										
	EDAP										X						X																	X	X	X	X	X	X				
	Isopropylated phospate ester																			X																							
	Phosphate esters									X			X	X	X			X																	X	X	X	X	X				
	Phosphorus polyol																					X																					
	RP																						X												X	X	X	X	X				
	RDP																							X				X															
	TEP																					X	X																				
	TPP																				X			X				X					X										
	TCPP*																																								X	X	
	Metal phosphinate																																X										
Inorganic FR (I)	AOH	X	X			X	X																				X																
	Aluminum trihydroxide (ATH)	X	X		X	X		X	X		X	X	X	X	X	X	X		X	X					X				5-30	X	X		X	X	X	X	X	X	X				
	ATH (+ ATO)																																										
	MDH	X	X			X										X			X																								
	Zinc borate																			X													X		X	X	X	X	X				
	Sulphonates																										X																
Nitrogen FR (N)	Melamine borate									X							X	X																	X	X	X	X	X				
	MC			X																	X					X							X							25			
	MC + MPP																									12.5 respectively						12.5 respectively								12.5 respectively			12.5 respectively
	Melamine phosphate																				X													X	X	X	X	X	X				
	Melamine polyphosphate																				X					25						25		X	X	X	X	X	X	25			25
	Melamine pyrophosphate																																		X	X	X	X	X				

3 PIN FLAME RETARDANTS AND RECYCLING: AN R&D AND TECHNOLOGY OUTLOOK

What does the R&D around PIN FR focus on?
Is it up to date with the recyclability targets for plastics?
Can we identify key stakeholders?

3.1 The EU Circular Economy targets for plastic recycling and why it matters for PIN FRs

"Plastics define the way we live today (17)". If we consider their functionality, it seems impossible to exchange them for a different, more sustainable and climate-friendly material. However, with an **expected doubling of plastics production in the coming 20 years**, it is evident that a linear plastic economy – with plastic waste as an unavoidable destiny – is an option we should discard.

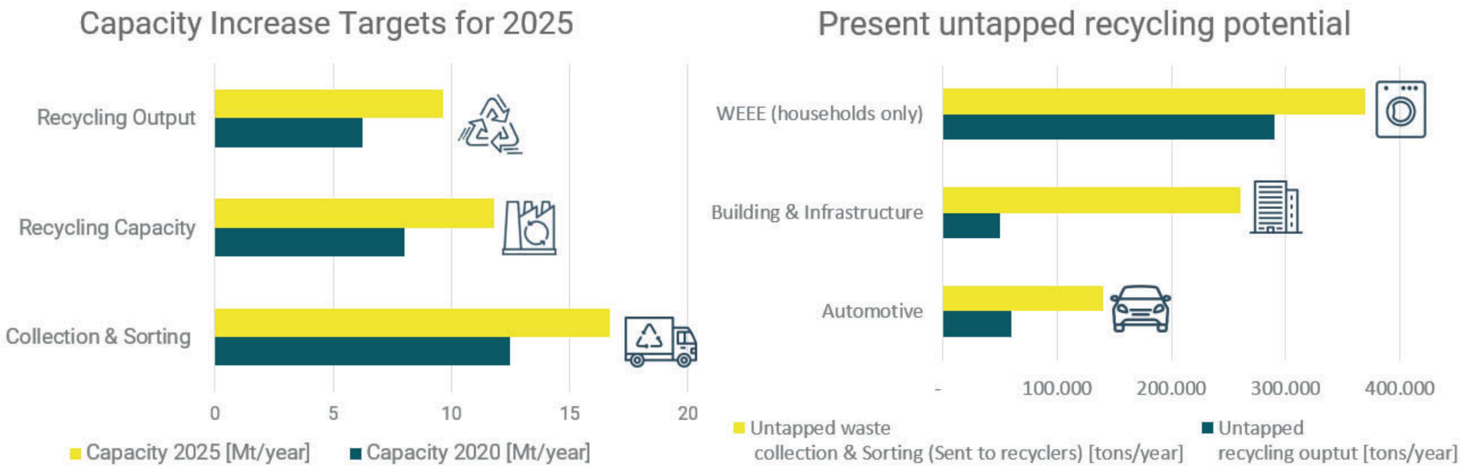


Figure 3-1: EU Plastic recycling targets for 2025 and current untapped potentials as established by the CPA

The European Commission has set very ambitious circularity objectives for plastics. Regulatory targets of the recently revised waste directives are 10% max landfilling of municipal waste by 2035, 50% recycling of plastic packaging by 2025 and 55% by 2030[12].

To address this need, in 2018 the European Commission issued the *EU Strategy for Plastics*[13] in the Circular Economy, establishing the main goals for plastic design, manufacture, use, re-use, and end-of-life management by 2030.

Correspondingly, the Circular Plastics Alliance[14] (CPA) was launched in December 2018 to help the plastics' value-chain meeting their pledged goal[15] to **bring the EU market for recycled plastics to 10 million tonnes by 2025** (hereafter: "the 10 Mt target"), **which means providing a boost of an additional 3.4 million tonnes of recycled plastics compared to 2020.**

To meet such a target, the full potential for greater uptake of recycled plastics will be reached by combining greater recyclability with an efficient separate collection of plastic products and products containing plastics. It has been established that the sector must engage in pursuing (i) increased capacity, (ii) better technologies, (iii) design of more recyclable products and (iv) stimulating demand of recycled products.

Calculations assessed by CPA's industry Working Groups (18) have shown that this implies **an increase in sorting capacity by at least 4.2 million tonnes** by 2025 and recycling capacity by at least 3.8 million tonnes, corresponding to estimated investment needs between € 7.6 billion and € 9.1 billion.

Sector	Ref. Year	European converters Plastic Demand	Tonnes of Plastic waste collected	Tonnes sorted for recycling	Percentage Sorted for recycling	Tonnes of recyclate produced in the EU	Percentage recyclate production out of sorted for recycling	Legislative Framework	Untapped waste collection & Sorting (Sent to recyclers)	Untapped recycling ouptut	Increase of recyclates output
Automotive	2019	5.100.000	1.500.000	350.000	23%	150.000	43%	ELV Directive	140.000	60.000	40%
Building & Infrastructure	2018	10.137.600	1.746.000	450.000	26%	340.000	76%	Country-based	260.000	50.000	15%
EEE (households only)	2016	1.749.030	752.500	717.589	95%	561.373	78%	EEE Directive	370.000	290.000	52%

Figure 3-2: Overview of waste quantities sorted for recycling today and the untapped potential for recycling to be reached by 2025 as estimated by CPAs industry groups [absolute numbers represent tons/year]

[12] Directive (EU) 2019/852 on Packaging and Packaging Waste
[13] https://ec.europa.eu/environment/circular-economy/pdf/plastics-strategy-brochure.pdf
[14] The alliance covers the full plastics value chains and includes over 300 organisations representing industry, academia and public authorities
[15] https://ec.europa.eu/growth/industry/policy/circular-plastics-alliance_en

At large, **the 3 key application industries for PIN FRs, EEE, Building & Infrastructure and Transport (automotive), provide almost 20% of the total plastic waste going to recyclers** (9.4 Mtons /year in 2018)[16].

In practical terms the **"10 Mt target" implies an increase in the final recycled outputs by 40% for the automotive sector (part of the transport industry), 15% for the building sector and 52% for the WEEE sector, which provides evidence of the efforts and investments needed over a very short amount of time.** The recycled plastics are then used in different sectors, with construction being the primary market with a share of close to 50%, while automotive and EEE reach 5% altogether.

The main barriers to more recycled plastics being utilized in European products are derived from a more detailed CPA market sector-based research on legal, economic, and technical barriers to uptake of recycled content. They can be summarised in three main barriers:

A. The **adequate quality of recycled plastics**, adapted to the requirements of the second life application.
B. The **availability and security of supply** of recycled plastics.
C. The **competitiveness (attractiveness/acceptance) of recycled plastics** vs. virgin plastics.

It is thus clear that **any additive in plastic's lifecycle should not interfere with these requirements to support EU recycling targets and leverage the consequent market opportunity: PIN FRs make no exception.**

On the specific products side, **26 key products** have been identified that the CPA has committed to making recyclable (Figure 3-3) by 2025.

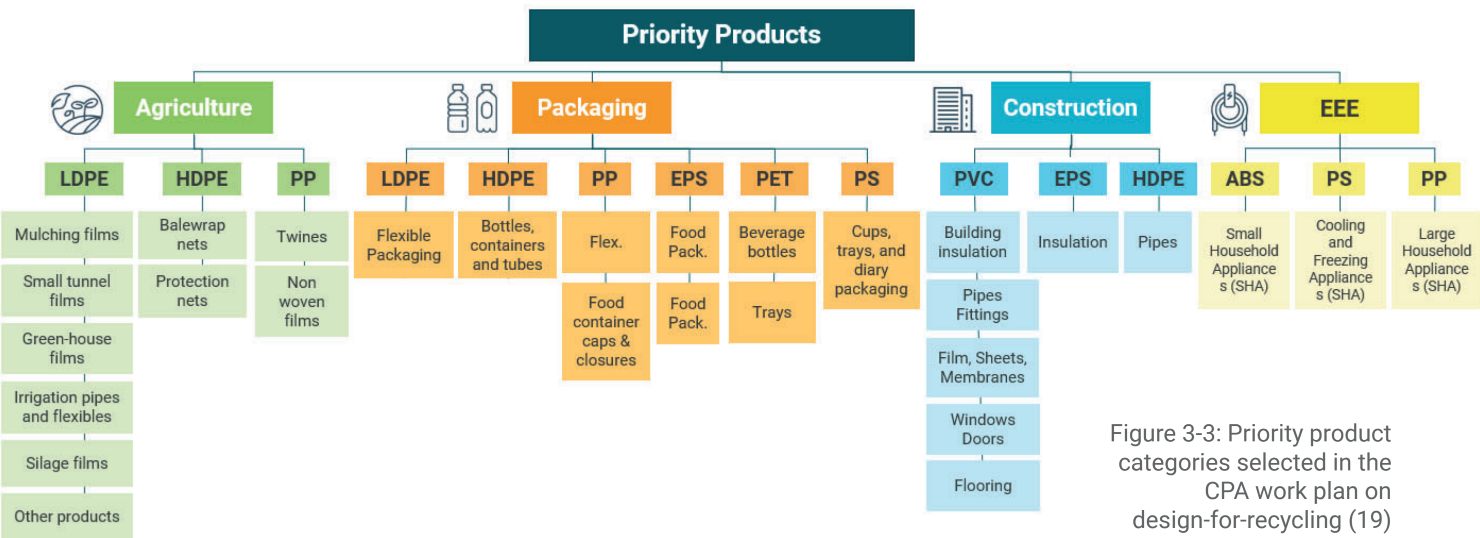


Figure 3-3: Priority product categories selected in the CPA work plan on design-for-recycling (19)

[16] PlasticEurope, https://www.plasticseurope.org/en/resources/publications/1899-circular-economy-plastics-europeanoverview

Construction and EEE are PIN FR-related sectors that will be most likely impacted, addressing products which include different types of polyolefins, that are forecasted to contribute to PIN FRs market growth in the next 5 years (see 1.2).

	Percentages in collected waste			Tons/year converters plastic demand			Tons/year sorted for recycle		
Type of plastic	WEEE	Building and Infrastructure	Automotive	WEEE	Building and Infrastructure	Automotive	WEEE	Building and Construction	Automotive
PE-LD/LLD	0,0%	5,1%	3,0%	-	517.018	153.000	-	22.950	10.500
PE-HD/MD	0,8%	12,8%	7,0%	14.342,0	1.297.613	357.000	5.884,2	57.600	24.500
PP	24,3%	7,4%	26,0%	424.299,8	750.182	1.326.000	174.081,0	33.300	91.000
PS	17,3%	1,7%	2,0%	302.212,0	172.339	102.000	123.991,0	7.650	7.000
EPS	0,0%	8,0%	0,0%	-	811.008	-	-	36.000	-
PVC	1,6%	51,7%	3,0%	28.159,4	5.241.139	153.000	11.553,2	232.650	10.500
ABS,SAN	25,4%	0,0%	4,7%	444.600,7	-	239.700	182.410,0	-	16.450
PMMA	0,0%	0,0%	1,0%	-	-	51.000	-	-	3.500
PA	0,4%	0,0%	11,9%	6.121,6	-	606.900	2.511,6	-	41.650
PCS	0,0%	0,0%	2,0%	-	-	102.000	-	-	7.000
PUR	0,0%	0,0%	15,0%	-	-	765.000	-	-	52.500
PC	2,8%	0,0%	0,0%	48.623,0	-	-	19.949,0	-	-
Other ETP	0,0%	0,0%	21,0%	-	-	1.071.000	-	-	73.500
Other	27,5%	13,4%	4,0%	480.633,4	1.358.438	204.000	197.193,5	60.300	14.000

Figure 3-4: Plastics values in collected waste, converters demand and sorted waste by industry (PNO elaboration of data included in CPA's Working Groups deliverables (20))

The approach suggested and acknowledged by the EC clearly illustrates a double target to improve products and processes. From the products standpoint it entails developing design-for-recycling guidelines as input for some European standards (CEN and CENELEC). However, **new R&D and investments will be needed to find new technology solutions**, especially if design-for-recycling standards should not produce the desired upgrade in recycling yields. It is thus relevant to provide a picture of the current status of plastic recycling R&D when PIN FRs are involved.

3.2 Recycling R&D trends when PIN FRs are concerned

For this report an R&D trends analysis was completed. It consists of a technology intelligence looking at a mixed corpus of R&D (public funded) projects, patents and scientific literature (reports, papers) enabling insights into the most relevant research and **innovation initiatives related to applications of polymers containing PIN FR, and their recycling technologies**. This also allowed for an overview of the key players and stakeholders[17]. A series of 8 interviews with experts have complemented the data-driven investigation. The interviews reports are available in Chapter 6 and have supported the overall conclusions in Chapter 7.

3.2.1 The technologies PIN FR R&D is focussing on

The heatmap below wraps up and relates different information, summarising the content and timeframe of our examined corpus in terms of considered recycling technology, type of flame retardant and waste source categories.

3 technologies are clearly represented: (i) Mechanical recycling, (ii) Chemical recycling and (iii) Solvent-based recycling, while (iv) Sorting technologies appear as an essential cornerstone to improve the downstream recycling process.

- **Mechanical recycling is an established technology. Looking at R&D literature, it has also been the most used and investigated process for the recycling of PIN FRs polymers in the last decade.** Different techniques have been used to mechanically separate and recycle polymers containing PIN FRs (e.g., electrical separation combined with melt-filtering). Achieving good results in this case is hindered by technological problems related to the sorting phase. At the state of the art, there isn't any dedicated technology which can discriminate between BFR and PIN FR containing plastics. Usually, a process that combines different plastic sorting technologies is preferable. Only in recent years LIBS-based sorting tests (Laser-induced breakdown spectroscopy) have shown the capacity to identify different types of plastics and distinguish between BFR and PFR in LCD TV back cover waste. After the sorting phase, we found density-based separation to be the most considered technique, upstream of the mechanical recycling of PIN FRs polymers. Multi-extrusion has been used to recycle polymers containing PIN FRs. This technique can obtain recycled

[17] Details about the methdololgy are reported in the Annex-2 to this report; the corpus of most relevant documents is reported in Annex-3
[18] It is to be noted that the following reference dates have been considered: i) Paper publication date, (ii) Projects start date, (iii) Patents Priority date

compounds (HFFR + Polymers) with excellent mechanical and flammability properties. However, it is not possible to obtain and separate pure HFFR. Looking at novel techniques, in a recent patent (2020) CO2 is used as an extraction agent to recover and analyse the flame retardant in a supercritical fluid, interestingly presenting CO2 as a mean to recover pure PIN FRs (21).

Table 3-1: PIN FRs Polymers Recycling Heatmap of Combined Results from Scientific Literature, Funded Projects and Patents Analyses

Year	Recycling Type				Waste Source					Flame Retardant Category		
	Mechanical Recycling	Chemical Recycling	Solvent-based Recycling	Waste to Energy by Pyrolysis	WEEE	Transport	Building and Construction	Post consume, not specified	Post consume (Packaging)	Phosphorus based	Inorganic Based	Nitrogen Based
2005	0	1	0	0	0	0	0	1	0	0	1	0
2006	0	0	0	0	0	0	0	0	0	0	0	0
2007	1	0	0	0	0	1	0	0	0	0	1	0
2008	0	0	0	0	0	0	0	0	0	0	0	0
2009	0	0	0	0	0	0	0	0	0	0	0	0
2010	1	0	0	0	0	0	0	1	0	0	1	0
2011	0	0	0	0	0	0	0	0	0	0	0	0
2012	3	2	1	0	3	2	0	0	1	6	2	0
2013	3	1	0	0	4	0	0	0	0	4	0	0
2014	3	0	0	0	2	0	0	1	0	0	1	1
2015	0	1	0	0	0	0	0	0	0	0	0	0
2016	1	0	0	0	0	1	0	0	0	0	1	0
2017	0	1	0	1	0	0	0	1	1	2	0	0
2018	5	0	0	0	3	2	1	2	1	3	3	1
2019	1	0	0	0	0	0	0	0	1	1	0	0
2020	3	0	1	0	2	1	0	1	0	3	0	0
2021	2	1	0	0	0	0	1	0	2	3	0	0
2022	0	0	1	0	1	0	0	0	0	1	0	0
2023	0	0	1	0	0	0	0	1	0	1	0	0

- **Chemical recycling is rising to complement other plastic recycling options.** It has been referred to as a "game-changer" by Plastic Europe, which recently announced investments by EUR 2.6 billion in 2025 to EUR 7.2 billion in 2030. With such investments, **looking towards the future, it may become one of the key recycling techniques** combined with (less energy-intensive) mechanical recycling. Accordingly, this study found the most interesting PIN FR-related chemical recycling process investigations by industry in patents (see Figure 3-10), above all to recover and reuse phosphorus-based flame retardants (especially TPP). They claim to allow separating and recovering the flame retardant. In general, it is possible to recover pure PIN-FRs if the temperature range is medium-low, beyond which the PIN-FRs start to decompose. Although chemical recycling can achieve important results separating PIN FRs, no R&D projects (here meaning a project suppored by public funding through grants) were found in our research, that specialised on PIN FRs. A lot of the literature referring to chemical recycling in fact is related to the recovery of BFRs. On the other hand, some interesting studies have been carried out considering the pyrolysis process (22). In Pinfa #136, seven studies are summarised, including (23), which shows that PIN compounds, phosphate esters or nitrogen-mineral (urea-boron) containing polyol, can be used for glycolysis of PUR and then production of a PIN FR recycled PUR.

- **Solvent-based recycling appears as the emerging technology investigated the most in the last years** (see as example Showcase 3 in Chapter 5), also supported by experts in the interviews (Chapter 6), The process proves to be an excellent solution to recover pure PIN FR. Different solvents are considered in the selected literature and patents: MeOH and EG are reported to remove TPP from PS and ABS; DMCHA solvent to remove BDP from PC/ABS; xylene solvent to remove aluminium tri-hydroxide (ATH) from PE and PP

3.2.2 The relevance of additives

Looking at plastic recycling, one cannot discard the relevance of the market of compatibilisers, stabilisers and other recycling additives.

They do not directly concern PIN FRs, but are certainly a key elment to be considered for the future of PIN FR plastics recycling. This research work acknowledges that additives can indeed modify recycling, by ensuring that recompounding is possible for mixtures, which without such additives are not compatible. Different established market research companies agree on its growth with an average CAGR exceeding 5% in the next 5 to 10 years.

Already in 2019, *Compounding World* summarised the offer of specialist additives, designed to address odour problems, improve processing, improve compatibility between different polymers and fillers being remixed, limit colour deterioration and restore properties of the recycled plastic (24). Additives specifically adapted for recycling can hinder polymer degradation during re-extrusion, improve reprocessing (lubricants, melt stabilisers) and provide final material stabilisation (including antioxidants, anti-UV, acid-scavengers). Their use was also confirmed during one of the interviews with the industrial recycler MGG (Chapter 6).
They are applied in different plastic types, from low to high end: commodity (PE, PP, PS,PPE), engineering and high-performance plastics (HPA), with the latter one forecasted by analysts to grow faster. While the construction market growth is pulled by the developing countries, in Europe, the automotive industry is expected to tow the demand, which makes Germany a prominent market.

There are limited established players (mid to large size companies), generating the majority of the revenues: these include BASF, Evonik Industries AG, Clariant AG, The Dow Chemical Company, ExxonMobil or DuPont, to name a few, with specific products offered on the market.

By application, additives includes the key segments impacting PIN FRs, namely automotive, construction and electronics. It is to be noted that Flame Retardants are also mentioned as additives. In this sense, in our selected WEEE Showcase 8 (Chapter 5), PIN FRs are added to different polymers (PE, PE/EVA and PC/ABS) during the recycling process, providing final good flame retardancy properties to the recycled plastic.

3.2.3 What the ID card of PIN FR-related R&D looks like

R&D initiatives with specific focus on recycling and PIN FRs are reasonably a smaller subset compared to other different topics, such as brominated or antimony recycling, development and production of PIN FRs or polymers containing PIN FRs, BFRs replacement and development of bio-based plastics containing PIN FRs. As a mere example, in many of the funded projects that were excluded from a detailed analysis in this research, the flame retardants were brominated materials and in many cases the recycling technique was the Creasolv™ process (25), developed and patented by Fraunhofer (based on solvent-based recycling).

In a glimpse, the recent publication rate of papers, exceeding funded projects and patents, shows how this is a field with still some challenges from the R&D point of view (Figure 3-5).

Regarding maturity, if we consider R&D projects as a display of future improved technologies and methods, most of them ranged between **medium TRLs – Technology Readiness Levels[19] - of ca, 4 to 6.** Such projects (see 10.2) embrace the whole cycle, where plastic waste containing PIN FRs is recycled to obtain a new polymer containing recycled PIN FRs. In these cases - even using mechanical recycling - technical or economical improvements are sought to define better and more viable processes. We also found some more basic-research projects concerning PIN FRs, at still lower TRL.

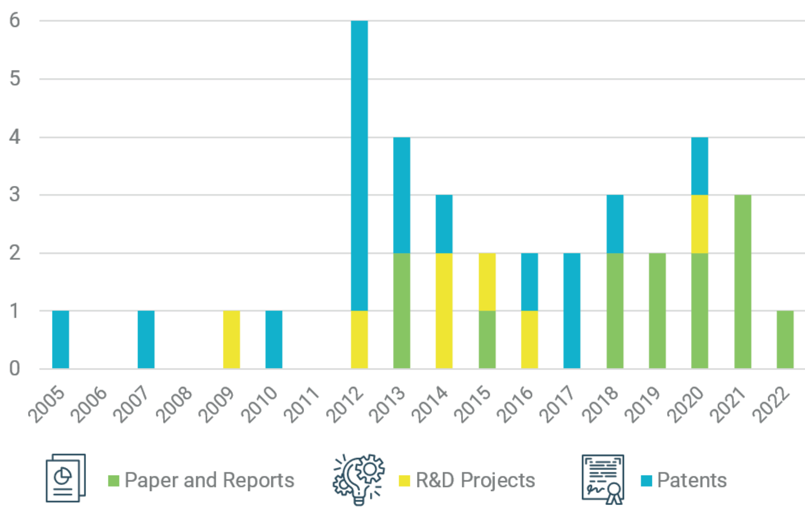


Figure 3-5: Count of most interesting documents in our selected corpus with a focus on PIN FR and recycling

These studies focus on directly testing the recycling possibilities of developed polymers containing PIN FRs as additives. By looking at such different combined results from a varied scoreboard, some trends clearly emerge, and considerations can be made:

- **The primary focus by industry and R&D projects is recycling the polymer, not the PIN FR, avoiding its downgrading**
- **Most cases focus on WEEEs** (especially cables), therefore, further attention has been paid to illustrate relevant cases concentrated on this application. They are reported in Chapter 5.
- **Among PIN FRs, phosphorus-based flame retardants are the most considered ones in the selected cases,** followed by other inorganic FRs. Most of them derive from post-consumer plastics from the EEE sector (mainly based on PC/ABS blend or ABS). We found references to bisphenol A, triphenyl phosphine, aluminium hydroxide, magnesium hydroxide and ammonium polyphosphate, among others. Some illustrative graphics are reported from Figure 3 -6 to Figure 3-8.

The next section, Chapter 4, provides more figures supporting these statements (Section 4.2).

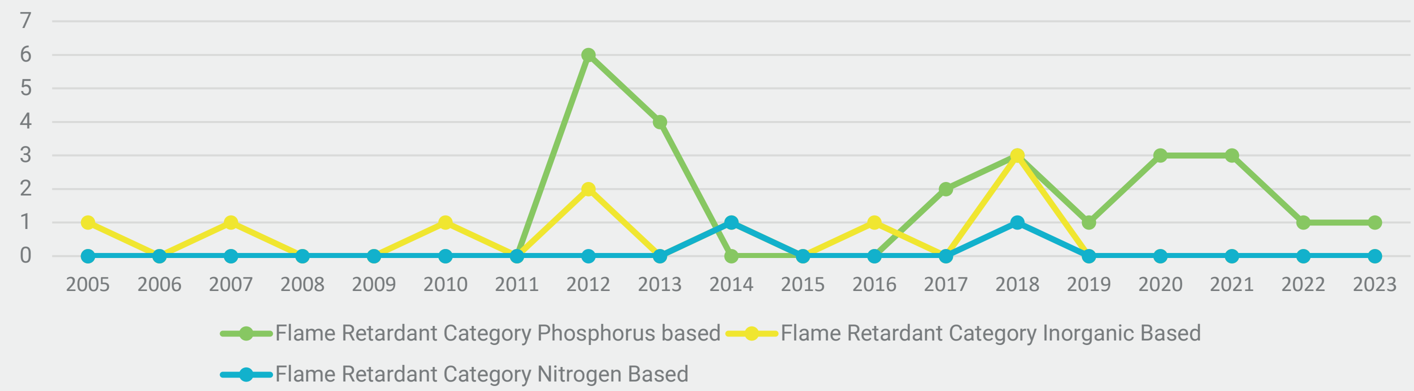


Figure 3-6: FR category represented in the corpus of analysed documents per year of reference

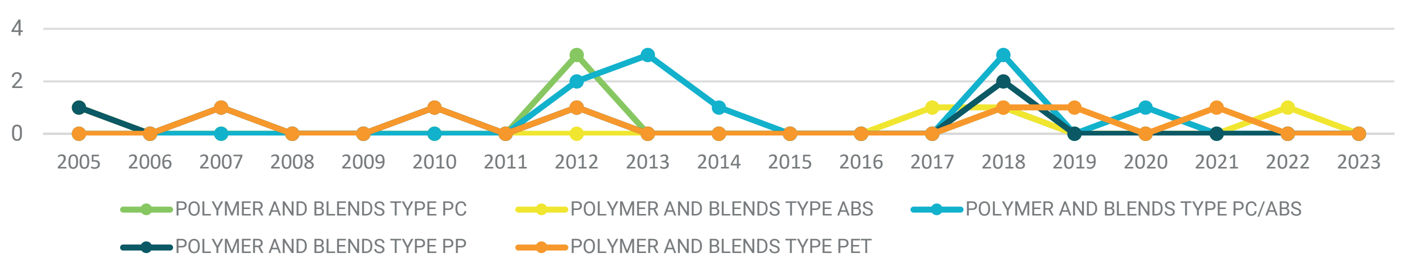


Figure 3-7: Most considered polymers and blends in the corpus of analysed documents per year of reference

[19] https://ec.europa.eu/research/participants/data/ref/h2020/wp/2014_2015/annexes/h2020-wp1415-annex-g-trl_en.pdf

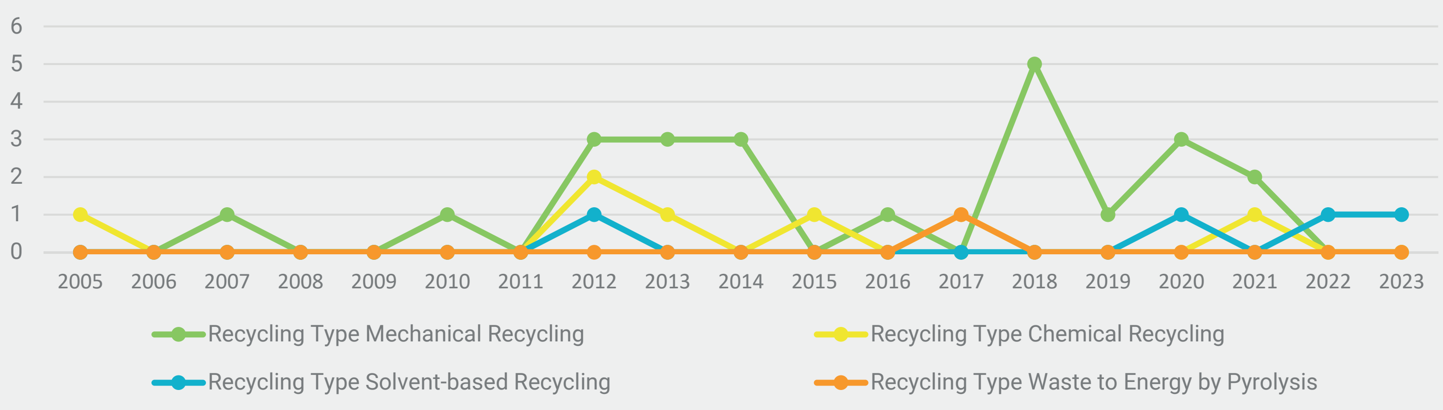


Figure 3-8 PIN FR's recycling methods in the examined corpus per year of reference

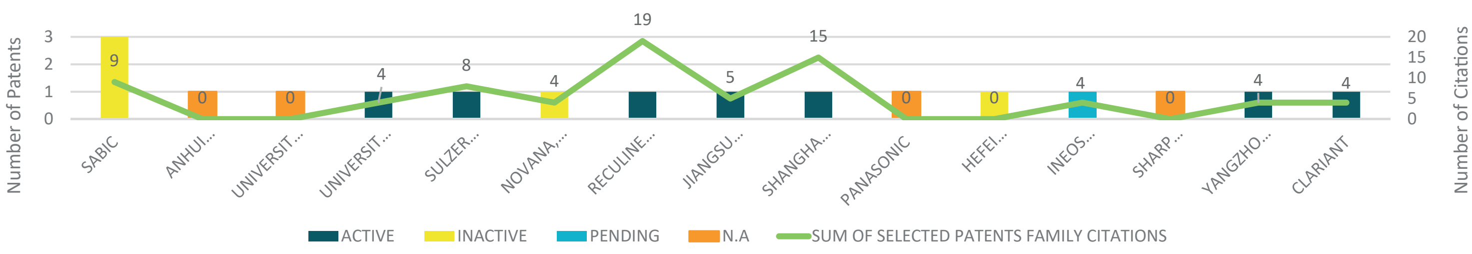


Figure 3-9: Applicants of the selected patents by status and patents family citations

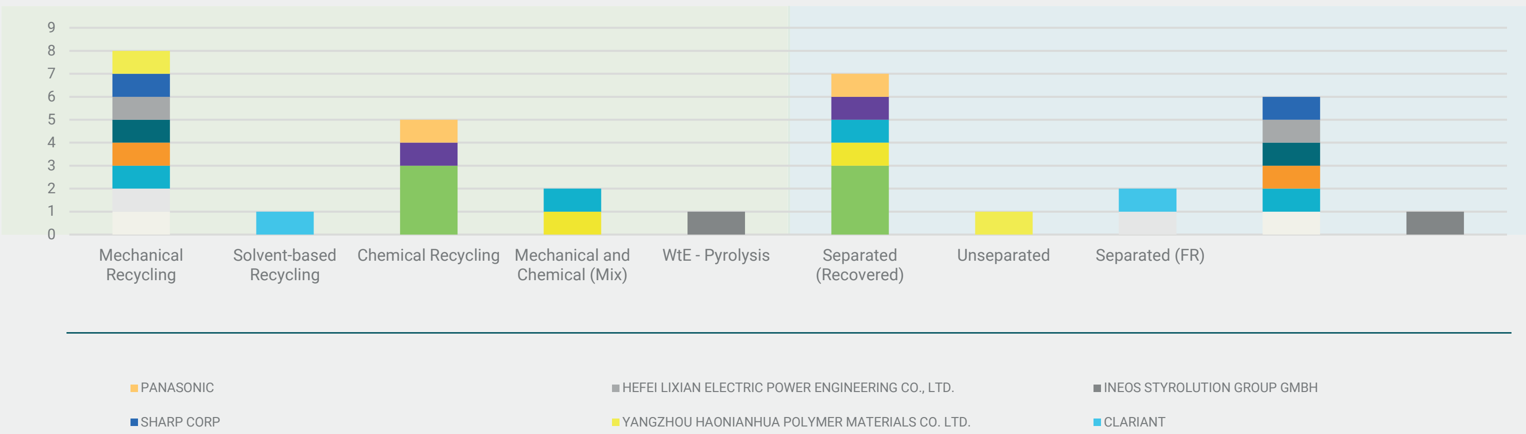


Figure 3-10: Patents applicants by recycling technology (left), process and fate of FR at EoL (right)

3.2.4 Who some key-players are and how they are positioned: a bird's view

The R&D literature study in this report identified a value-chain composed of **7 main categories:**

- **Waste Providers**: companies which provide plastic waste such as WEEE.
- **Sorting and Separation Service Providers**: companies involved in plastic sorting and separation processes.
- **Recycling Companies**: running facilities involved in the recycling process of plastic waste.
- **Recycling Technology Providers**: who develops and provides technologies to recycle polymers containing PIN FRs.
- **Plastics Producers and Compounders**: companies/organisations specialised in the production of different flame retardant-based plastics
- **Flame Retardant Producers**: companies/organisations specialising in the production of halogen-free flame retardants or other chemical specialties.
- **End-Users**: companies using the recycled plastics or the PIN flame retardants.

By type, most of the relevant organisations represent SMEs or large companies but many vital academic stakeholders emerge and position as recycling technology providers or plastic compounders. A rapid bird's view is shown in Figure 3-11: all in all, a final list of players has been highlighted, which was also instrumental in tracking key experts for this report's interviews (Table 3-2).

Figure 3-11: Bird's view of the PIN FR polymers recycling Value Chain emerged from this report

Looking at the geography, some extra-EU companies appear by including patents: they are primarily multinational corporates, whose patents are registered under the Japanese and Chinese patent offices. Indeed, many applicants come from Japan or China. As for highlights and examples, it is worth mentioning that Panasonic patented a heat treatment process to recover pure boehmite from Aluminium Hydroxide, while in the past Sabic patented 2 methanolysis and 1 ammonolysis based process (coupled with use of solvent) to recover pure Bisphenol A. Looking at the citations of the 16 most relevant patents in Figure 3-10 – as an indicator of their relevance in the sector - while SABIC still stands out, other players are also worth mentioning, like Reculiner BVBA and SHANGHAI KUMHO SUNNY PLASTICS CO. LTD.

With a focus on the EU, the analysis has identified the network behind the top 7 R&D analysed funded projects (Figure 3-12), spotting active innovators as key "nodes" with more projects participations: they are Fraunhofer, KU Leuven, Tecnalia, Sitraplas Gmbh and Coolrec BV.

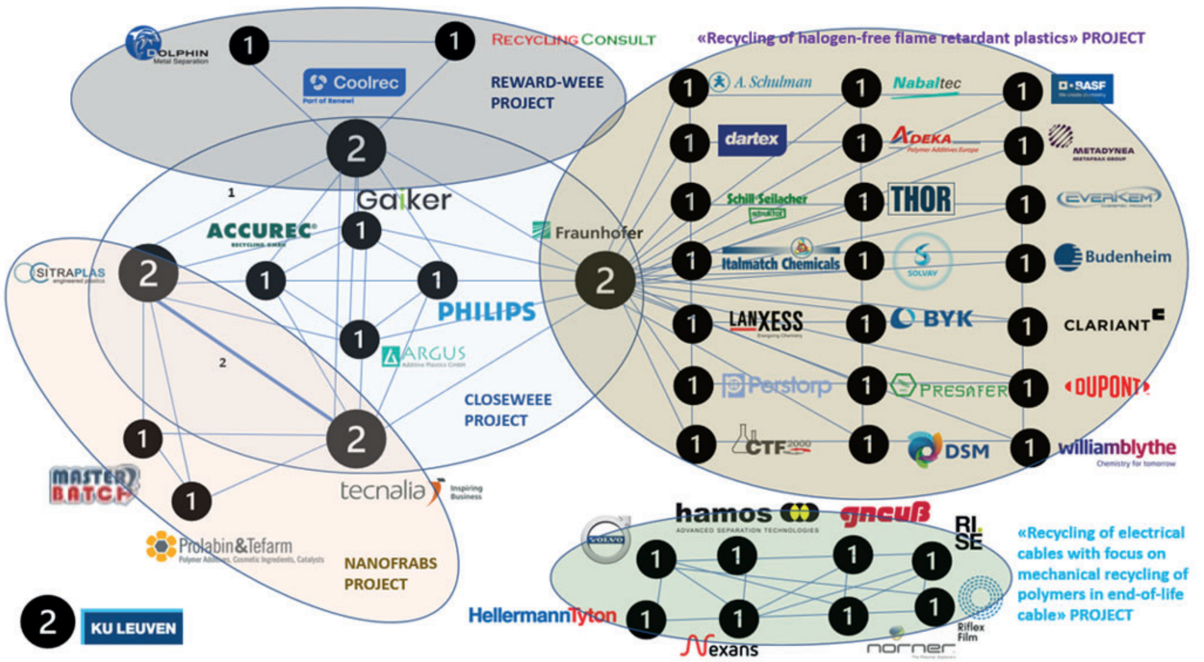


Figure 3-12: Project innovators network (the number in the nodes represent the number of projects participated by the corresponding organisation)

In the field of solvent-based recycling, for example, KU Leuven appears very active, being found both in literature and R&D projects investigations. Of course, PINFA and its members are well represented in literature, with key publications. It is also worth mentioning the University of Cuenca , EMPA, Fraunhofer, ICL-IP Europe, and Recycling Consult BV as noticeable publishers. A list of interesting players connected to the observed R&D literature is reported below in Table 3-2.

Figure 3 -12: Project innovators network (the number in the nodes represent the number of projects participated by the corresponding organisation)

Stakeholders	Why it could be relevant
Fraunhofer	Fraunhofer LBF Institute developed a mechanical process using multiple-Extrusion to recycle several polymers containing phosphorus or inorganic-based flame retardants (25).
gneuß	Gneuss Gmbh provides extrusion and melt-filtering technologies able to recycle PVC containing HFFR (ATH and MDH) without a loss in its mechanical and flame retardancy properties
KU LEUVEN	One of the most representative institutes in Europe studying new methods for PIN FRs recycling, KU LEVEN, is researching and developing methods based on solvent and mechanical recycling. It has also written three papers focused on closed-loop recycling of plastics containing BFR and PIN FR with a specific focus on the issues related to sorting technologies applied to recycle WEEE.
RECYCLING CONSULT	Recycling Consult BV is a spin-off from Delft University of Technology and is specialized in the development of unit processes for waste recycling (above all WEEE). They are part of REWARD-WEEE, an EU project that aims to demonstrate in a prototype facility the generation of recyclable products from WEEE., testing advanced separation and sorting techniques by mechanical (Polymer sensor sorting of brominated and phosphorus flame retardant containing polymers).
tecnalia Inspiring Business	Developing innovative, resource-efficient solutions to complete the loop of post-consumer high-grade plastics, from WEEE to new EEE, through the advanced recovery of valuable plastic streams that do not yet have a recycling system. This includes the subsequent replacement of halogenated flame retardants with halogen-free flame retardants in the new EEE.
INEOS STYROLUTION	INEOS is the leading supplier of styrene monomer, polystyrene, ABS and styrenic specialities. The company is investing in chemical recycling for the depolymerization of polystyrene waste to styrene monomer. INEOS Styrolution, Recycling Technologies and Trinseo have announced that they have reached a significant milestone in their plans to build commercial polystyrene (PS) recycling plants in Europe. *Recycling Technologies* has been selected as the technology partner. The selected patent focuses on a recycling method for styrene containing plastics through pyrolytic depolymerization, removing red phosphorus.
سابك sabic	One of the leading companies in the chemical industry. It has patented chemical recycling methods using ammonolysis and methanolysis for recovering pure bisphenol A and derivates from Polycarbonates and PC/ABS blend.
SULZER	The company is specialized in purification, filtration, and mixing technologies. The Chemtech division is the global market leader in innovative mass transfer, static mixing and polymer solutions for chemicals, petrochemicals, refining and LNG. The selected patent concerns a process to recycle expandable plastic materials by extrusion and Melt-filtering, adding the HFFR during the recycling process to give to the recycled material the proper flame retardancy properties for its reuse.

Panasonic	World leader in Electronic device and expert in plastic moulding and compounding. It patented a method of chemically recycling plastics through heat treatment for recovering pure boehmite from ATH flame retardant.
The University of California 1868	The university has patented a depolymerization process for recovering pure TPP flame retardant from Fiber-reinforced poly(diketoenamine) composite.
SHARP	World leader in Electronic device. It also develops a closed-loop plastic materials recycling technology. It patented a mechanically recycling method to recycle thermoplastic resin and adding phosphorus-based flame retardants during the process to give the recycled materials improved flame retardancy properties.
Gaiker	Within the selected project, Gaiker develops a LIBS (Laser-induced breakdown spectroscopy) prototype to improve the sorting and separation phase of plastics from flame retardants (both BFR and PIN FR). It also has extensive expertise in the recycling of WEEE plastics containing PIN FR.
Campine	Expert in sorting technologies for the recycling of plastics and metals containing PIN FR.
Coolrec Part of Renewi	It owns several recycling plants for WEEE containing PIN FR.
Dolphin Metal Separation	In the REWARD-WEEE project it provides its plant for testing different polymers and metals sorting and separation processes. The technologies tested are: density separation (magfluid), Colour sensor sorting, Dual-energy x-ray sensor sorting for the separation of halogenated polymers, Density separation to remove light plastics from heavy plastics and Near-Infrared sensor sorting for the separation of polymer types. The considered blends are: PC/ABS and HIPS/PPE.
norner	Expert in polymer recycling and provider of additives.
DTU	It carried out a study in 2021 on fast pyrolysis of Polyurethanes and Polyisocyanurate with and without TCPP flame retardant (Note, TCPP is not anymore rated as "PIN" but instead as a "Chlorinated - Phosphorous FR) (26).
Ghent University	It carried out a study in 2021 on fast pyrolysis of Polyurethanes and Polyisocyanurate with and without TCPP flame retardant (Note, TCPP is not anymore rated as "PIN" but instead as a "Chlorinated - Phosphorous FR) (26).
UMass Lowell	Chemical Recycling of Mixed Plastics in Electronic Waste Using Solvent-Based Processing. They considered PIN FR as TPP (27).

4 IMPACT ON RECYCLING: A RECYCLING MATRIX MODEL

Can we assess PIN FR's impact on plastics recyclability based on R&D cases? How to collect this information?

4.1 The Recycling Map and Matrix

To optimize the analysis in the scope of this report, a tool[20] has been developed, which offers an overview of the recycling of plastics containing PIN FR. The resulting Recycling Map keeps track of the diverse recycling process possibilities in relationship of a chosen plastic material substrate, containing a certain PIN FR. For the sake of clarity, in the following, we refer both to the Recycling Map and Matrix.
Their exact distinct definitions are as follows:

- **Map**: a tool that allows the retrieval of relevant information upon given search criteria
- **Matrix**: data structure on which the PIN FR map is built.

The PIN FR Recycling Matrix considers the following flow across the entire life cycle of a recycled polymer (EoL) obtained from plastic-containing waste.

Table 4-1: Conceptual flow used for the recycling matrix and map design

CONCEPTUAL "FLOW"	Waste	Treatment	End of Life (EoL)
	INFORMATION CONTAINED WITHIN THE MATRIX		
	▪ Classification of waste ▪ Source of waste ▪ Polymer (chemical composition) *Vs.* ▪ Flame retardant classification (PIN)[6] ▪ Flame retardant (chemical composition)	▪ Classification of treatment / process ▪ Treatment detail	▪ Fate of Polymer ▪ Fate of flame retardant

[20] The Recycling map is intended as a qualitative tool that can be operated via spreadsheet filters (MS Excel). This allows to simply select single or multiple combinations of given parameters.

The Map provides all the information necessary for mapping the possible combinations in terms of waste type, polymer, FR content, treatment process and fate (EoL) of both polymer and the contained FRs. A detailed description of the Matrix rationale is reported in Annex-4 .
As for the applications mapping in Chapter 2 and Table 2-2, it should be noted that antidrip agents, synergists and smoke suppressants, as well as recycling compatibilising additives have not been included into our analysis.

4.1.1 Applications of the recycling map

The development of this tool and approach was aimed at:

- **Simplifying the collection and organization of information related to plastic recycling and therefore possible PIN FR impacts on it**: the Matrix is a framework that collects and classifies the available knowledge related to the treatment of plastics and resins which are obtained from different waste types, and contain PIN FRs.
- **Supporting the understanding of current possibilities related to the plastic recycling when PIN FR are involved. I**t can be used as an informative tool to understand the end-of-life of the polymers containing PIN FRs and to provide a quick overview of any possible combinations which have already been mentioned in scientific literature, funded projects, or patents.
- **Providing an intuitive, easy-to-use instrument based on a common taxonomy, including elements of the *EC's Waste Hierarchy*.** It can also be used as a decision support tool by industry to quickly understand the optimal combination of PIN FR and polymers, given the knowledge of the state of the art.

4.1.2 "Case" definition

The matrix is composed of cases extracted by the Literature study in Chapter 3: a "case" describes a unique effort/technique/study to recover a single plastic material based on polymer, mixture, or resin.

It should be noted that one article, or reference, may include multiple cases that have been deconstructed and analysed with the purposes of populating the matrix with the highest possible number of inputs. Currently, the Matrix has 69 single cases from 36 references. Each case is classified according to the description in the following section, Section 4.1.3, as shown in **Figure 4-1.**

[21] The present exercise includes only information concerning FRs. No information are yet available concerning other additives and including synergies, anti drip agents, compatibilizers as well as other agents, etc.

Table 4-2: Information sources used for recycling matrix population

Source of information	Nr.	Represented Cases
Relevant Scientific literature issues	13	27
Relevant funded projects	7	15
Relevant Patents	16	27
Total	36	69

The current design allows for the entry of any further relevant data, as well as to providing semi-quantitative information. The semi-qualitative output can then be used to obtain an informative "dashboard" and provide a quick overview of the state of the art concerning plastics containing PIN FR. Part of the information includes references, such as scientifically reviewed papers DOIs, funded project references and patent identifiers. External data sources (e.g., CAS numbers) are also considered.

This approach is open to being extended, especially considering the likely appearance of new technical and scientific know-how in literature, funded projects and patents, which may be capable of replacing or integrating the state of the art.

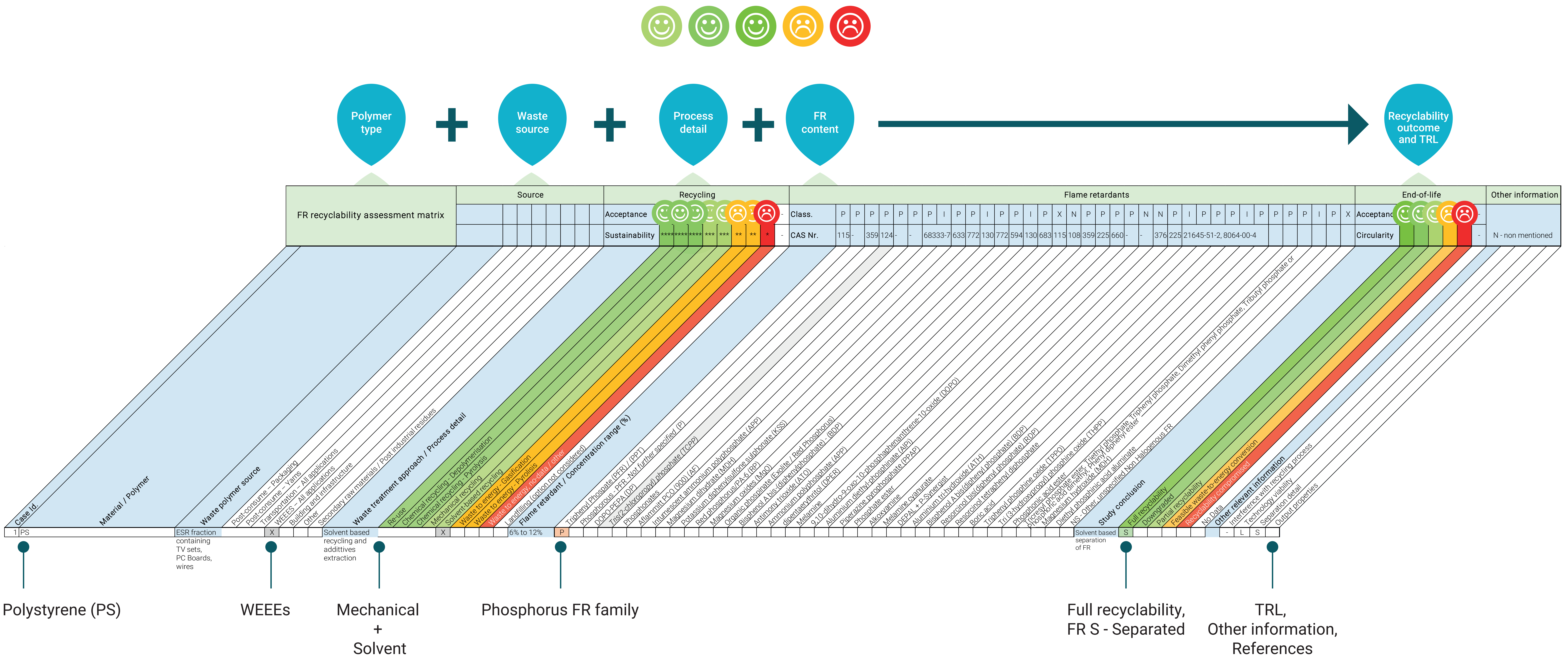
4.1.3 Classification taxonomy

A robust nomenclature helps to prevent any redundancies, or information inconsistencies.

The defined taxonomy (see Annex-4 for details on each of the categories below) allows to limit ambiguity, and cover possible future expansions of the existing knowledge base[22]. The recycling matrix elements have been classified as per the following categories, covering the value-chains of all plastics waste containing PIN FRs:

- **Waste polymer source**
- **Recycling processes**
- **Polymer end of life (EoL)**
- **FR – Flame Retardant end of life (EoL)**

[22] The following classification has been provided to an expert panel in a dedicated session and to verify if the matrix is actually capable of covering all the possibilities sourced from the existing knowledge repositories (literatures, project, patents).



Polymer type
+
Waste source
+
Process detail
+
FR content
Recyclability outcome and TRL
FR recyclability assessment matrix
Source
Recycling
Acceptance
Sustainability
Flame retardants
Class.
CAS Nr.
End-of-life
Acceptance
Circularity
Other information
N - non mentioned
Case Id.
Material / Polymer
Waste polymer source
Waste treatment approach / Process detail
Flame retardant / Concentration range (%)
Study conclusion
Other relevant information
1 PS
ESR fraction containing TV sets, PC Boards, wires
Solvent based recycling and addittives extraction
6% to 12%
P
Solvent based separation of FR
S
Polystyrene (PS)
WEEEs
Mechanical + Solvent
Phosphorus FR family
Full recyclability, FR S - Separated
TRL, Other information, References

4.2 Recycling Matrix current content information summary

A high percentage of the assessed cases (35%) concern WEEE sources (25/69). The most common materials assessed within the WEEE category belong to mixed ESR fractions, which comprise LCD-TV screen parts, and other electronic equipment (boards, cards, wiring, etc.).

This is followed by "other" (21/69), where the nature of collected plastic materials is not specified at all, and then post-consumer packaging (10/69), transportation – general materials, (7/69), other materials of different nature where little details are known to be more precisely classified (6/69).

Concerning polymers, diversity is higher. The most common mentioned cases are PC/ABS blend (11/69) followed by PET (9/11), ABS (5/11), PP (5/11), PC (4/11) and other polymers, or polymer blends (35/11)[23].

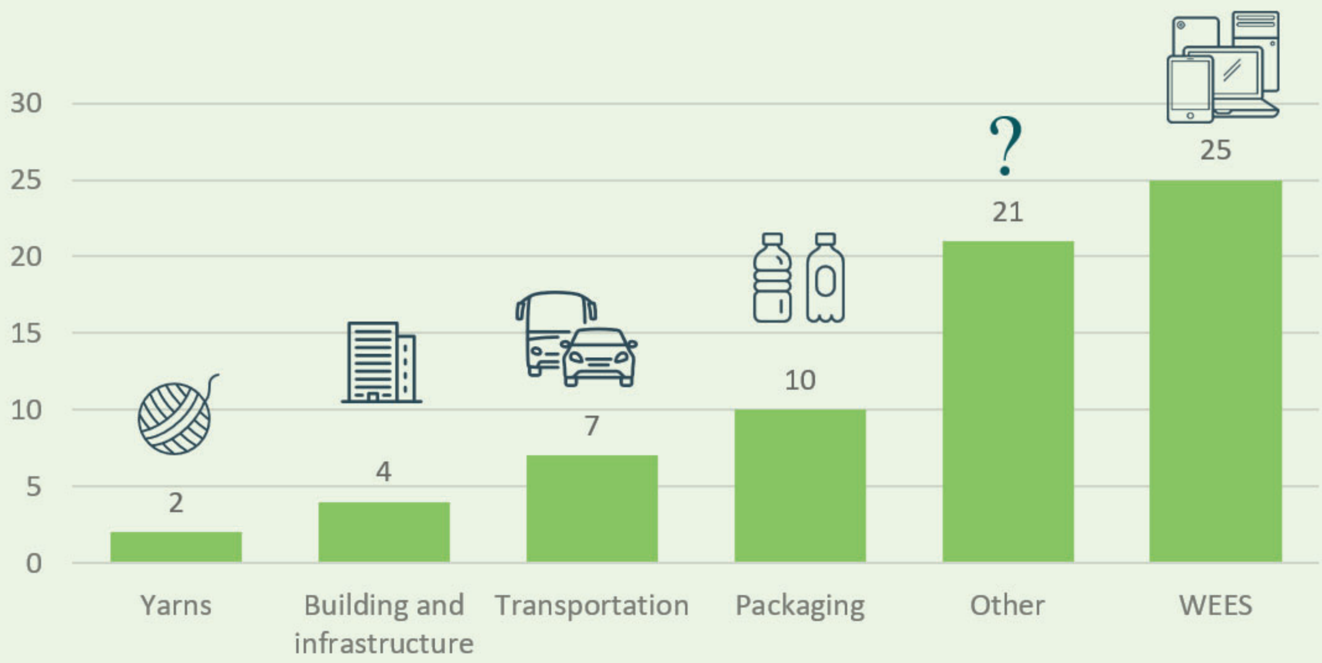


Figure 4-2: Reported cases "source of waste" analysis

[23] or the sake of completeness these include: NS (3/69), HIPS/PPE blend (3/69), PE (2/69), PE/EVA (2/69), CTA/PVA/TPP film (1/69), Vinylogous polyurethane (1/69), PP-flax composite (1/69), LLDPE (1/69), poly (diketoenamine) composite (1/69), EP (epoxy) (2/69), PP/PA blend (1/69), PA-6, PUR (1/69), PA-6.6 (1/69), HIPS-PPE (1/69), polyamide (1/69), EPS (1/69), PP/EPR blend (1/69), EVA. PP/Wood floor (1/69), glass-fiber reinforced polyamide (PA) (1/69), PS (1/69), PE/Tire rubber (1/69), PVC (1/69), HIPS/PPE (1/69), XPLE (1/69), PIR – Polyisocyanurate, PMMA(1/69

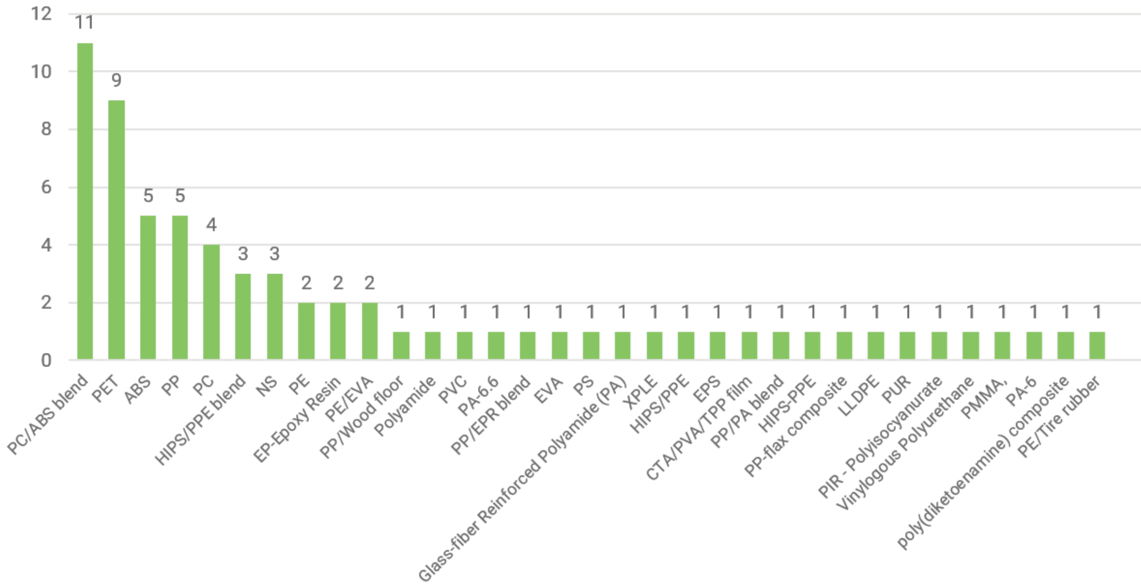


Figure 4-3: Reported cases - plastic composition (polymer, mixtures, resins)

Mechanical recycling is mentioned in the vast majority of the cases (50/69) followed with a lower order of magnitude by pyrolysis-based processes (7/69), solvent-based recycling (6/69), chemical recycling – depolymerisation (5/69) and waste-to-energy conversion process (1/69). It still results as the "state of the art" for plastic waste processing, due to its costs (CAPEX/OPEX) as well as the ease of operations without demanding complex training.

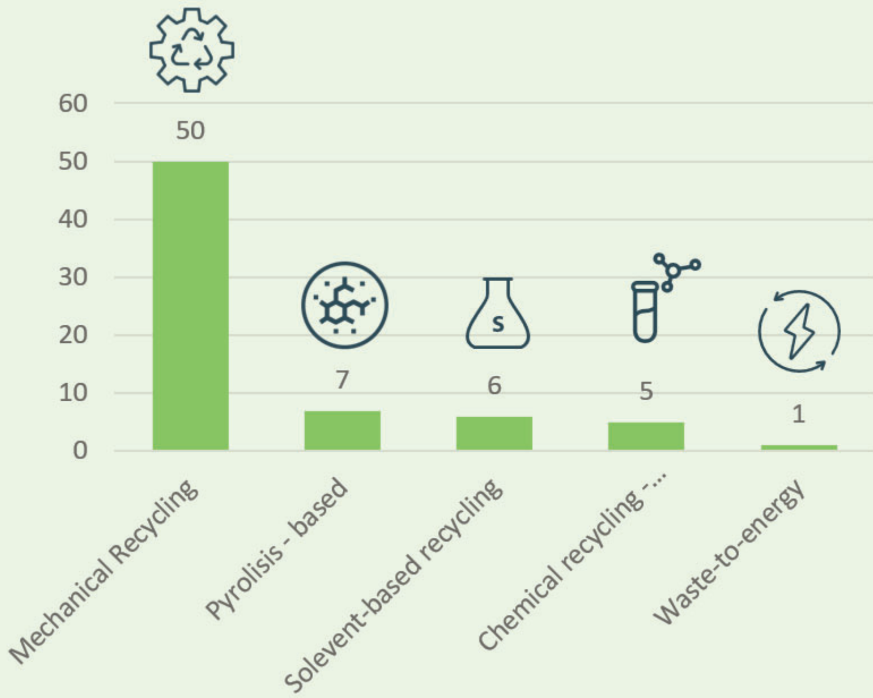


Figure 4-4: Reported cases - Most common plastics recycling processes

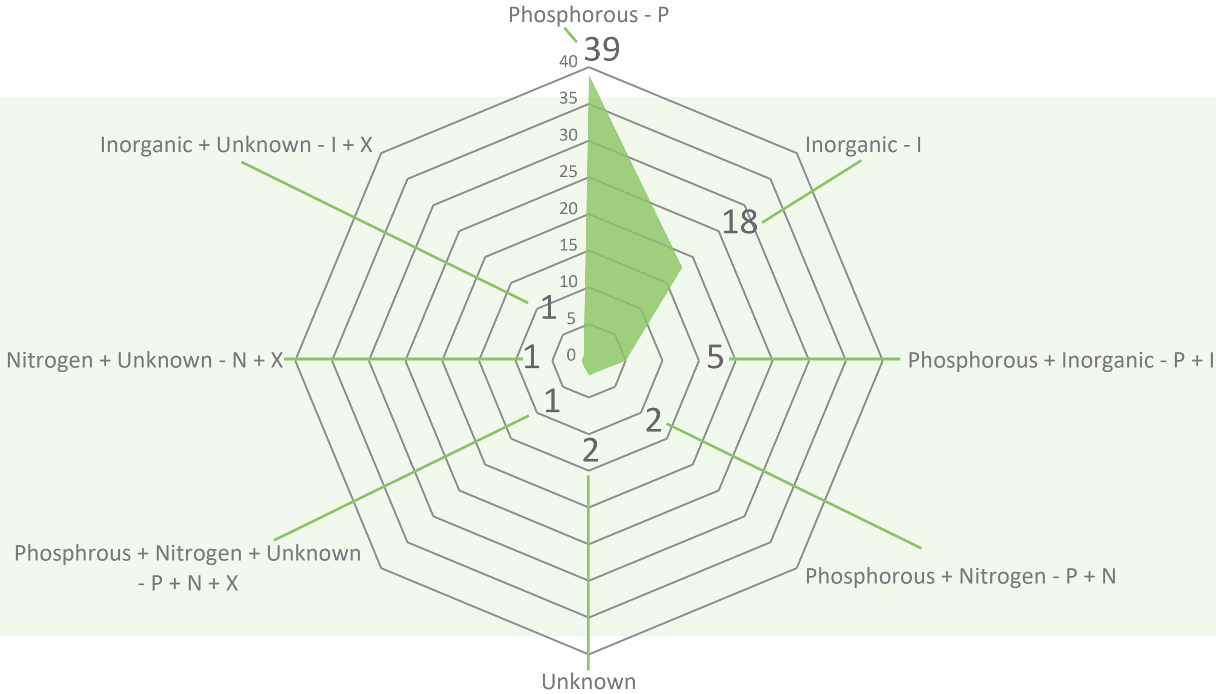


Figure 4-5: Reported Cases: PIN FRs occurance per type

P FRs result as the most common case (39/69) followed by inorganic (18/69), inorganic + P (5/69), N +, N + P (2/69), and unknown formulations either containing nitrogen FR, inorganic FR, or phosphorous FR (5/69).
This also reinforces the **growing relevance of Phosphorous compounds**, as sole FR components, or in mixed formulations used in common plastics.

4.3 PIN FR End-of-Life fate and polymers recycling results

The authors are aware that the 69 cases collected represent a "small-data" set, which needs the availability of a much higher amount of information to identify clear potential patterns and correlations**.**

Thus, the purpose of the following analysis is primarily to provide a picture of the available data and to use them to illustrate how this could be replicated with a systematic collection.

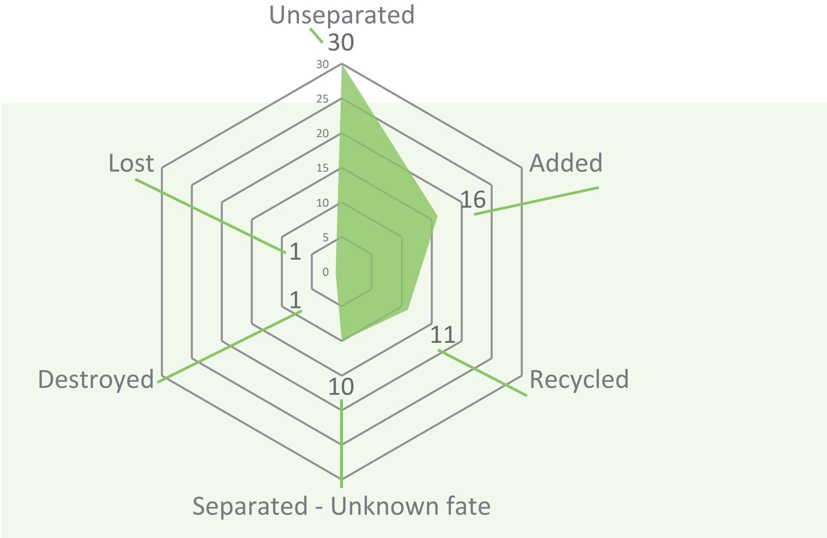


Figure 4-6: PIN FRs Fate (EoL)

Regarding their End-of-Life, in our dataset, PIN FRs are reported to be unseparated in 43% of our cases (30/69)**, to be added as additive post recycling in 23%** (16/69), **recycled in 16%** (11/69) and separated with unknown fate/destroyed/lost in 17% of the cases (12/69).

Specific patterns can then be spotted by breaking down the interdependencies among the different parameters: 30% of the cases reported full recyclability. In comparison, polymers result downgraded in 62% of the cases (it should be noted that no additive has been considered). Within the "full recyclability" EoL cases, the event of unseparated PIN FR is mentioned only 4 times out of 21 (19,5%).

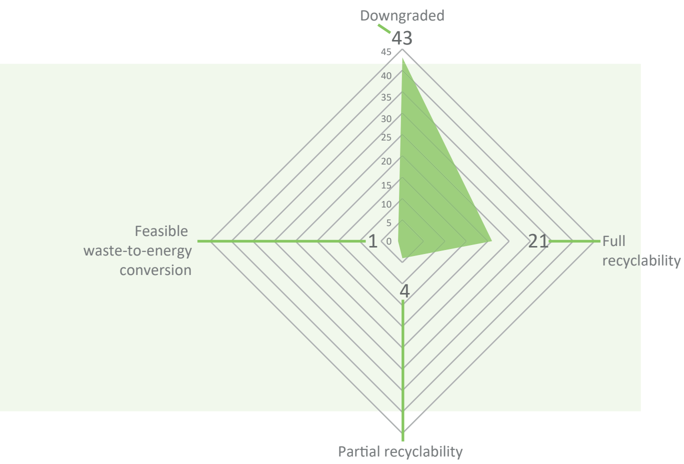


Figure 4-7: Reported cases: Recycling process outcomes (EoL)

Vice versa, when the outcome of the recycling process results in a final "downgrading" in the EoL (43/69 times), the FR results unseparated from the polymer matrix much more, specifically in the ratio of 23/43 (53%), which is significantly higher than the first case.

Looking at the composition, P-based flame retardants are mentioned 16 times out of 21 when full recyclability EoL is the outcome (76%). All in all, the available dataset shows that a mixture, or a more heterogenous presence of FRs within plastic waste, has a larger impact on the final quality of the recycled material output.

Technology-wise, **Figure 4-8 and Figure 4-9 show that chemical recycling and solvent-based recycling have often been used when full recyclability is the final outcome.**

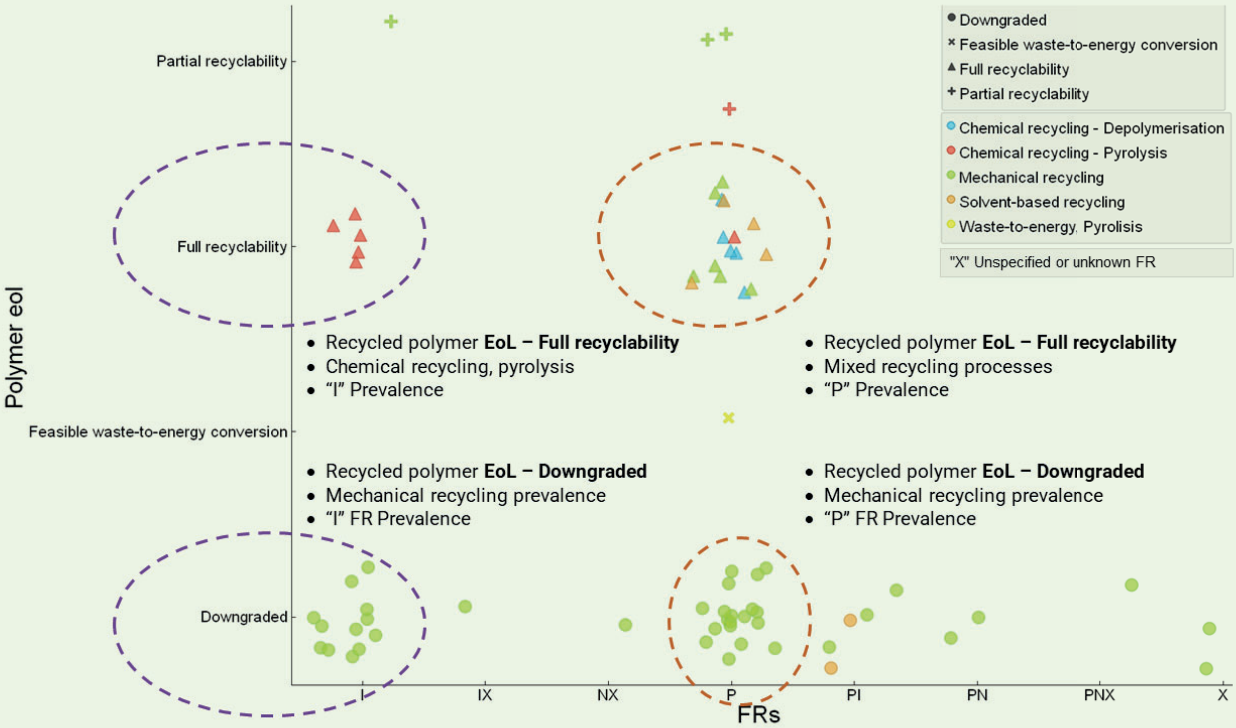


Figure 4-8: Example of data-analysis of PIN FR matrix performed via data mining means – PIN FR type Vs EoL fate in the available dataset for different recycling processes

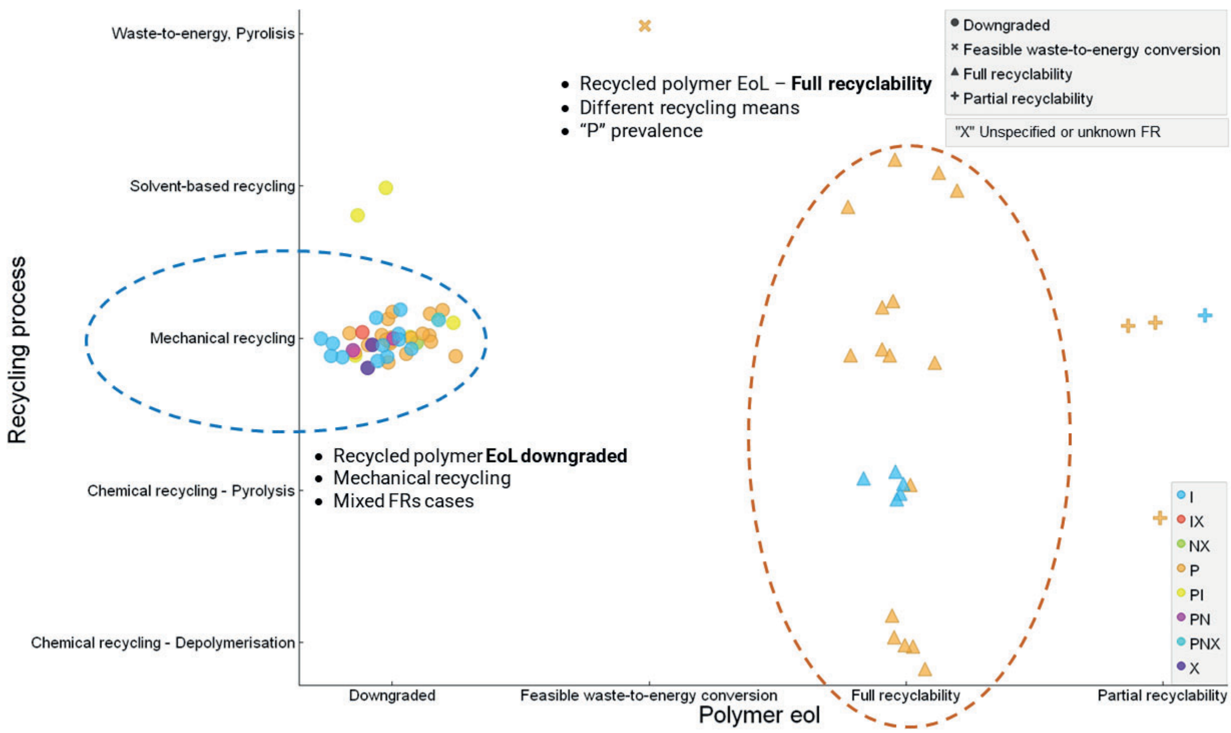


Figure 4-9: Example of data-analysis of PIN FR matrix performed via data mining means – Technology type Vs polymers recycling results

5 WEEE PLASTICS AND PIN FRs: RECYCLING TREATMENT SHOWCASES

What is the status of R&D for WEEEs streams when PIN FRs are involved?

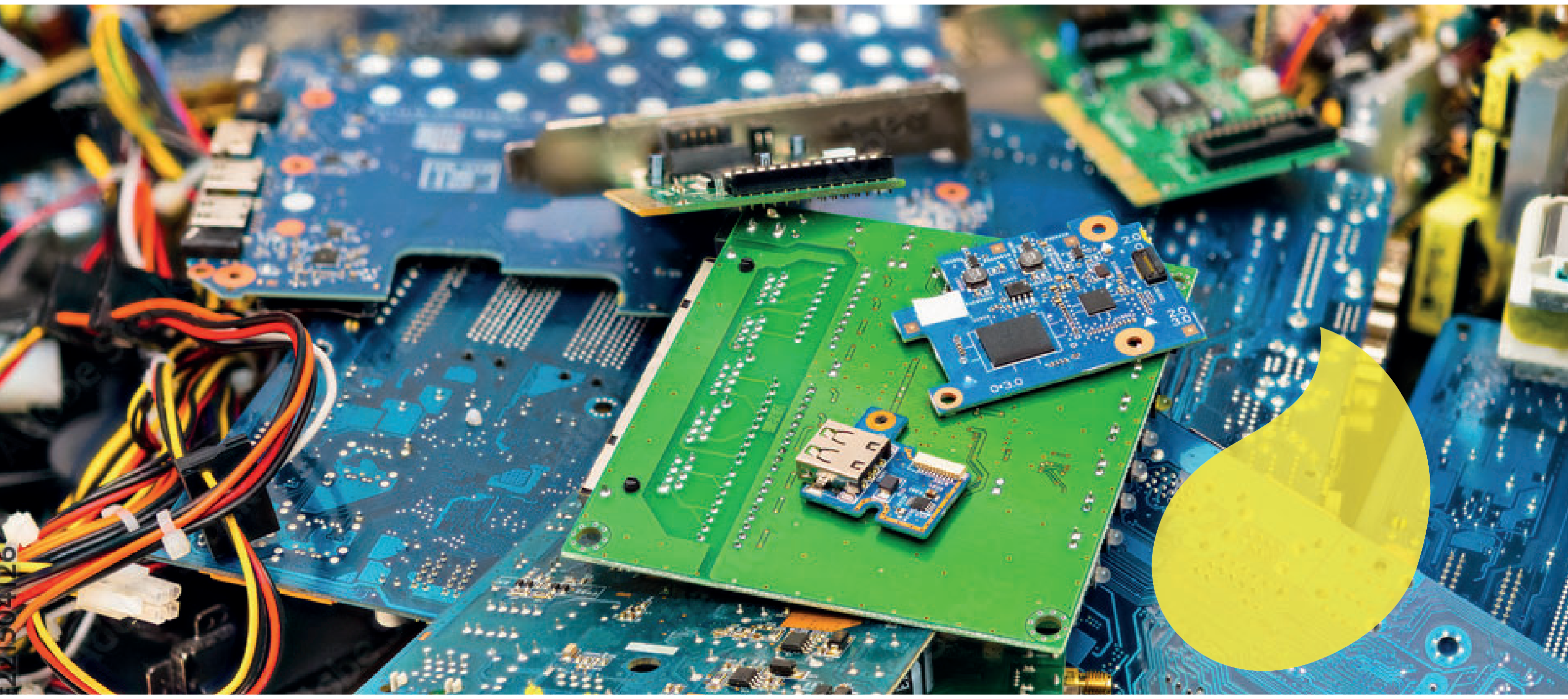
WEEEs emerged as the most represented segment in our literature study. The WEEE recycling value-chain's target is to increase the output by %52 within 2025, confirming the relevance of this sector. Thus, this chapter has a specific focus, illustrating in detail 8 showcases selected and retained during our research to describe approaches to WEEE plastic recycling when PIN FR are involved.

5.1 WEEE, regulations push and PIN FR

Back in 2013, the Electronics Strategy proposed the investment of €100 Bn and set the ambitious goal to create 250.000 jobs and double the creation of European computer chip production by 2020[24].
The European electrics and electronics market reached a global market share of about %14.6 in (28) **2016**. Legislation striving for more energy and fuel efficiency has accelerated the use of plastics in EEE.

Plastics from WEEE such as computers, TV-sets, fridges, and cell phones represent one of the fastest-growing waste streams in the EU, with some 10Mt generated in 2014, and was projected to grow to more than 12 Mt/y by 2020[25]. The electronics sector is one of the most stable consumers of polymers containing FRs, including PIN FRs. To enhance the environmental management of WEEE and to contribute to a circular economy and strengthen resource efficiency, the improvement of collection, treatment, and recycling of electronics at the EoL is essential. Thus the WEEE Directive or Directive 19/2012/EU was adopted. As of 1 January 2015, the WEEE Directive has set the EU targets for the reuse/recycling and the reuse/recovery of WEEE to %80 and %85 respectively[26]. Moreover, the RoHS[27] directive in Europe restricts the use of hazardous substances in EEE to protect the environment and public health. This has increased the interest in PIN FRs in EEE, which combine fire safety with better health and environmental profiles.

On the other hand, dealing with the high rates of FRs, including PIN, is particularly relevant for post-consumers WEEEs. The amount of FRs can typically range between weight-% 6 to (27) %14, with some peak at weight-% 80, for applications such as cables and wires (5) (8) (29).



5.2 WEEE recycling and treatment "as is"

The main steps (excluindg additives/compatibilizers) defining the usual process adopted by plastic recyclers are typically sorting, washing and drying, compounding and finally extrusion (Figure 5-1) (30), progressively turning mixed plastic flakes into plastic granules that can be used to generate new plastic products.

Currently, the average yield of plastic recycling processes from WEEE is around 60% (31). **The main gaps are technical and legislative.** A key issue is that plastic is just one WEEEs component out of different fractions and outputs in their treatment. Furthermore, sorting is usually a weak point in the chain, since other materials are the primary targets, such as rare earths, copper, steel or metals in general. Other technical barriers include the high similarity in density and the great variety of colours in WEEE plastics.

[24] EC, Shaping Europe's digital future, https://ec.europa.eu/info/strategy/priorities-2019-2024/europe-fit-digital-age/shaping-europe-digital-future_en, 2020
[25] EU WEEE waste,https://ec.europa.eu/environment/waste/weee/index_en.htm,
[26] Directive 2012/19/EC onWEEE,https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32012L0019,4.07.2012
[27] Directive 2002/95/EC on Restriction of certain hazardous Substances in Electric and Electronic Equipment

Clearly stated legislative barriers include among others the legal complexity surrounding flame retardant issues and relationships inside the EEE/WEEE value-chain, which are a paramount influencing factor (30). In Europe just %35 *(3.3 million tonnes of 9.5 million tonnes) of EEE discarded in 2012 ended up in official collection and recycling systems (32)*. The rest is traded or dumped out of the recycling channels with low standards. For this reason, **the main target to improve WEEE recycling is acting on the collection and sorting steps with a mixed approach.**

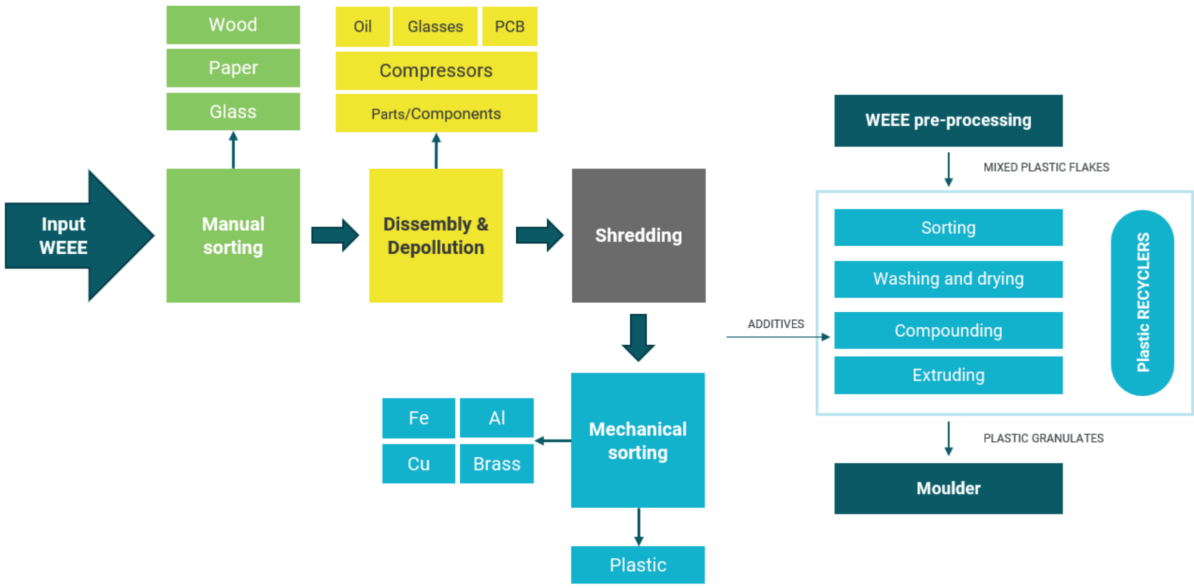


Figure 5-1: WEEE treatment steps (left) and typical WEEE plastics treatment (right) (30)

5.3 WEEE-related R&D showcases of recycling processes and technologies

SC 1: Evolution over time of sorting technologies in aid of mechanical recycling

In 2013, KU Leuven, ICL-IP Europe and Recycling Consult BV studied disassembly-based treatment and size-reduction (shredding) for a sample of 500 LCD TV back covers, with a focus on their separation after granulation, using automated optical sorting and a density-based separation

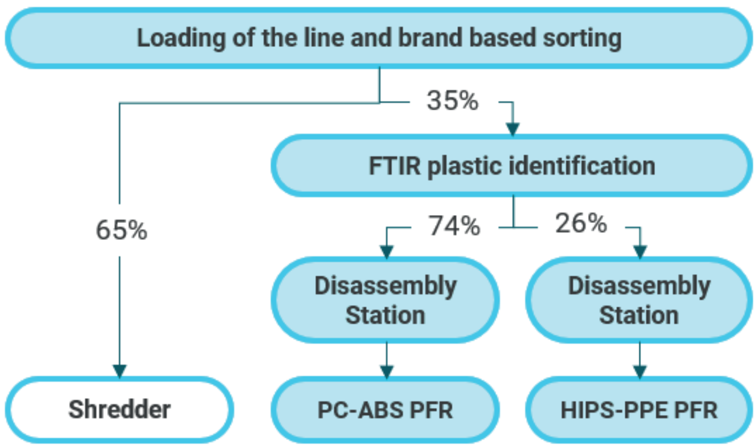


Figure 5-2: Proposed clustering strategy based on FTV brand for PC-ABS and HIBS-PPE with PFR (27)

process. NIR, XRF and FTIR technologies were tested in the study (27). **NIR** was used to identify plastic types, **FTIR** to identify black plastic types, **NIR and XRF** to scan back cover plastic and determine the type and FR presence (Br, P). The study shows that separation between HIPS and PC/ABS is possible with a combination of type/additive and density sorting, however, the shredding of a complete LCD set results in a mixture of plastics with more different types that **needs more processing steps**. The main plastics: HIPS, ABS (w/o Br-FR), PC/ABS and HIPS/PPE (with P-FR) found in the back covers can be recycled if disassembled and processed separately. A year later, the same publishers found that optical sorting processes need to be sequenced prior to density-based separation processes[28].

As a result, the overall recovery for PFR PC/ABS of this process sequence can be augmented from %55 to %71 by using a rotary table, which allows the reject fraction of the XRF sorter to pass once more through this optical sorter. However, many impurities were not separated in the optical separation processes, ending up in the PC/ABS fraction. These **results indicate that improvement in the separation efficiency of optical sorters is required to increase the purity of the FR plastic output fraction, as the recycled polymer resulting from this process requires the addition of virgin material to be reused.**

KU Leuven and Campine NV in 2019 provided a possible solution to the sorting problems previously faced. They assessed the technical feasibility of dismantling-based recycling strategies for the WEEE plastics (LCD TVs back covers) providing interesting conclusions on the sorting technologies able to differentiate different plastics containing brominated and phosphorus flame retardants. **Their assessments on the sorting technologies after LCD TV dismantling are resumed in the table.**

According to the authors, sorting trials with a LIBS contact measurement showed that this technology can detect plastic types and distinguish between Br and Phosphate Flame Retardants in LCD TV back cover materials during the sorting phase. For the study they consider a Quantom LIBS Scanner developed by Bertin Technologies in Belgium. After testing the dismantling-based recycling process (which includes the following steps: manual dismantling of LCD TV back covers, sorting by LIBS, XRF and FTIR, shredding, sink-flotation separation, fine shredding, washing, and drying, extrusion and melt-filtration), a recycled PC/ABS with PFR was obtained, suitable for direct re-application in electronic products, having good mechanical and aesthetic properties as well as recovered flammability.

[28] 1- All ferrous and non-ferrous materials should be separated with a magnet and an eddy-current separator. 2- Subsequently, the white internal and the BrFR containing plastics can be separated with an automated colour or NIR and an XRF sorter. 3-Only afterwards, a sink-float technique can be used to separate the remaining plastics: ABS, PMMA, HIPS, ABS, PFR HIPS/PPE and PFR PC/ABS.

Table 5 1: Overview of the most promising plastics/polymers sorting technologies

Sorting Technology	Available sorting technologies
Fourier Transform InfraRed (FTIR) spectroscopy	It measures the plastics response to infrared light. In contrast to Near InfraRed (NIR), the mid-infrared (MIR) range is used to identify black plastics as well. This technique is commonly used for quality management as it provides a reliable identification of the plastic type (33) (34). The most relevant WEEE plastics can be differentiated with FTIR. On an industrial scale for post-shredder sorting, these techniques are however often considered too slow (35)
Raman spectroscopy	It works with the Raman light scattering effectprinciple and delivers spectra similar to FTIR. This technique is seen as complementary.
X-Ray Transmission (XRT)	It analyses the extent to which X-rays are absorbed by a material and determine the atomic density. XRT can differentiate between FR and non-FR plastics. However, due to overlapping densities no distinction between Br and P-based FRs can be made (22).
X-Ray Fluorescence (XRF)	It uses X-rays to excite the atoms of a material to create a characteristic fluorescence light response to identify the different elements that are present. This technique allows identification of Br and detection limits of 300 ppm have been reported in literature for handheld devices (36)). However, the detection of the specific FR molecule is not possible. The quantification of light elements, such as P is known to be complicated. A testing series for this research in 2015 did not yield results with sufficient accuracy for detecting P in plastics from LCD TVs (22).
Sliding Spark SPectroscopy (SSSP)	It uses a high voltage spark to vaporize a small amount of the plastic surface. The emitted radiation can be measured to differentiate Br and P-based FRs. Determining the plastic type is also possible, but a testing series performed in 2014 delivered unreliable results for the distinction between ABS and HIPS (22).
Laser Induced Breakdown Spectroscopy (LIBS)	Uses a high energetic pulse laser beam to form a plasma and vaporize a small amount of the material surface. Excited atoms emit light and elements present on the surface can be identified. The identification of several plastic types and the detection of Br in plastics have been reported in literature. Sorting trials with a LIBS contact measurement in a nitrogen atmosphere showed that this technology is able to detect both plastic type and distinguish between Br and phosphate FRs in LCD TV back cover material (22).

SC 2: Use of pyrolysis to recycle TPP from LCD panels

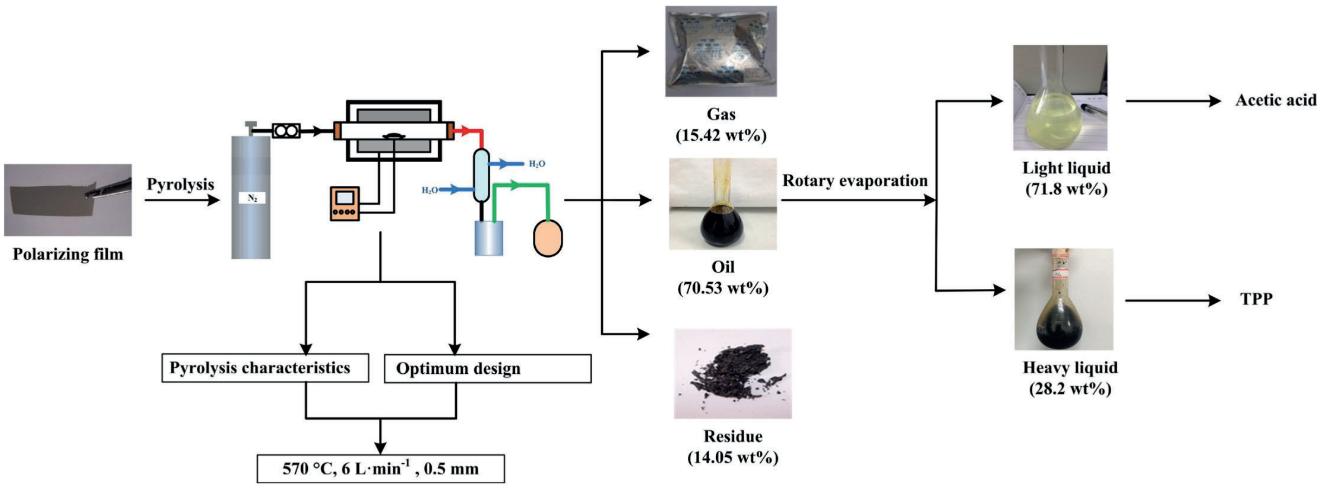


Figure 5-3 - Overview of pyrolysis process for recycling TPP from LCD panels (37)

The Shanghai Jiao Tong University conducted a study in (37) 2015 on recycling of liquid crystal display (LCD) panels waste. A pyrolysis process is used to remove and separate the organic materials and recycle acetic acid and triphenyl phosphate (TPP) by rotary evaporation which allows to reuse TPP in electronics manufacturing industry. The study shows the TPP starts to decompose that within the considered process when the operating temperature exceeds 250°C.

SC 3: Solvent-based recycling to recover TPP from electronic waste

The University of Massachusetts Lowell carried out a study in 2021 on the Recycling of Mixed Plastics in Electronic Waste Using Solvent-Based Processing (27). phosphorus-based flame retardants (PFRs) are relatively polar and are likely to dissolve in polar solvents like MeOH and EG.

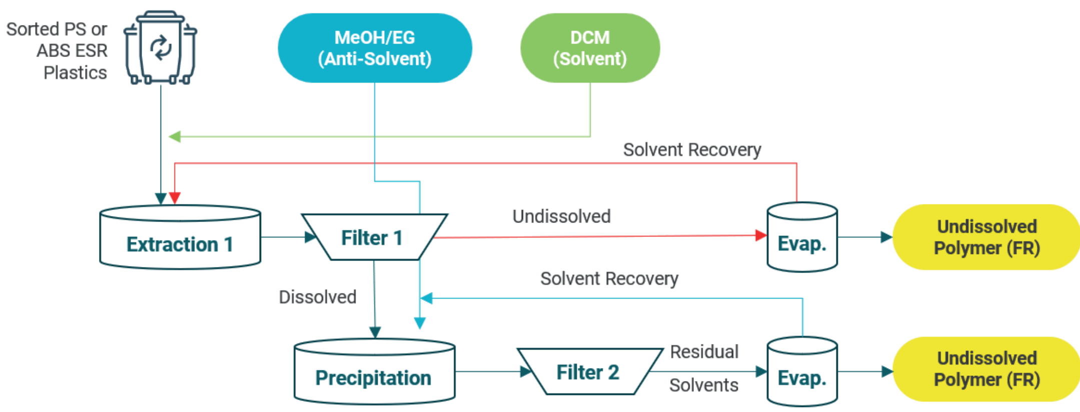


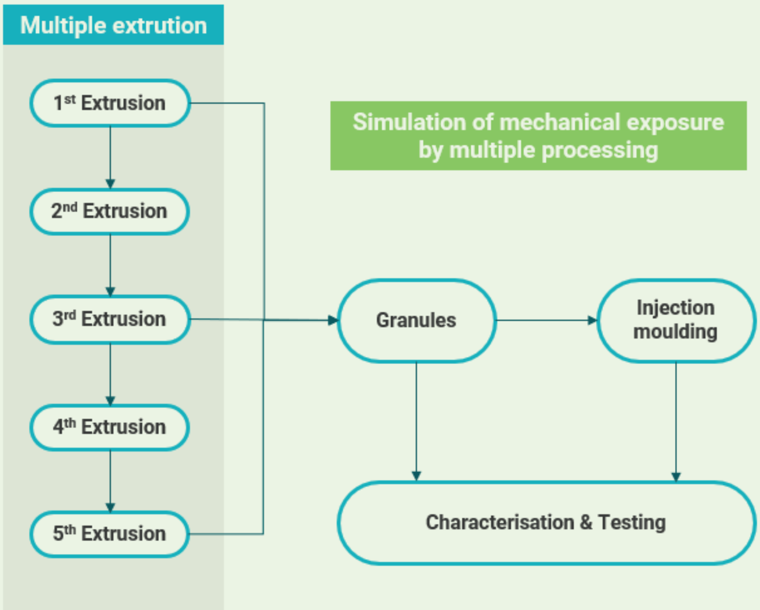
Figure 5-4 - Solvent-based Recycling of TPP from electronic waste (27)

The specific tasks of this study were to: (a) determine the composition of a representative sample for pre-sorted electronic shredder residue (ESR) (provided by Sunnking Inc), (b) identify a list of plastics for recovery using a solvent-based process, (c) scientifically design solvents/anti-solvents to effectively recover the identified plastics using HSP, (d) optimize dissolution conditions for dissolving plastics from the ESR, (e) design an anti-solvent system to recover the polymers and remove FRs from the polymers simultaneously, and (f) evaluate the energy efficiency. The feedstock was hand-sorted into the following categories: plastics, printed circuit boards, metals, wires, rubbers, metals, and capacitors. For the actual solvent-based recycling process in this study, only the hand-picked polymer fraction of the obtained ESR (i.e., pre-sorted by E-waste recycler) was used. The removal efficiency of P was significant—%70 and %94 of PFR (TPP) was removed when using MeOH or EG as the anti-solvent, respectively. This **suggests that phosphorus-based flame retardants (PFRs) are relatively polar and are likely to dissolve in polar solvents like MeOH and EG.**

SC 4: Recycling of halogen-free flame retardant plastics by multiple extrusion

Multiple extrusion is one of the most investigated testing methodologies for verifying the effectiveness of the mechanical recycling for plastics containing PIN FRs. Two of the funded projects selected to be investigated in this work address this process, without starting from plastic wastes but by providing ad hoc PIN FR polymers to be processed for evaluating their recyclability.

The German project "Recycling of halogen-free flame retardant plastics", funded by AiF and coordinated by Fraunhofer-LBF has investigated two recycling processes for 10 different HFFR plastics models, provided ad hoc by the several plastics producers (PINFA members) participating in the project. In the project mechanical recycling is used involving multiple-extrusion (step I) and accelerated oven-ageing (Step II) technologies (provided by Fraunhofer). After the extrusion step, follows the aging of granules and subsequently injection moulding phase or the injection moulding phase and subsequently the aging of the test bars. Finally, the characterisation and testing phase takes place. After this mechanical recycling **it is possible to obtain recycled polymers containing HFFRs. After five extrusion steps, flame retardancy was maintained for nine out of ten formulations.**



CZ	Polymer type /HFFR
1	PP / APP based system
2	PP / piperazine pyrophosphate
3	PP / 0,5 % alkoxyamine
4	PP / 1,0 % alkoxyamine
5	PE / ATH (I)
6	PE / ATH (II)
7	PA66 / GF30 / DEPAL HFFR
8	PA6 / GF30 / DEPAL HFFR
9	PA 6 / MC
10	PC / ABS / phosphate ester

Figure 5-5: 10 Different HFFR Plastics Models Considered in the German Project Funded by AiF

Figure 5-6: Multiple Extrusion with 5 Steps Used in the German Project Funded by AiF

CZ	Polymer type /HFFR	T (Extr.) / °C	Burning behaviour	Mechanical properties		
				Young's modulus	Tensile strength	Elongation at break
1	PP / APP based system	230	V-0	↑	↗	↑
2	PP / piperazine pyrophosphate	200	V-0	–	–	↓
3	PP / 0,5 % alkoxyamine	210	B2		–	–
4	PP / 1,0 % alkoxyamine	210	B2		↘	–
5	PE / ATH (I)	150-160	HB		–	↗
6	PE / ATH (II)	150-160	HB		–	↗
7	PA66 / GF30 / DEPAL HFFR	290	V-0	↘	↓	↘
8	PA6 / GF30 / DEPAL HFFR	270	V-0	↘	↓	↓
9	PA 6 / MC	270	V-0	↗	↗	↓
10	PC / ABS / phosphate ester	260	V-2	–	–	–

% relative change normalised to the respective initial valu

– unchanged
↘ ↗ < 25%
↓ ↑ < 50%
↓ ↑ > 50%

Figure 5-7: HFFR Plastics Recycling Project - Materials properties after Multiple Extrusion

The European funded project NANOFRABS coordinated by Tecnalia investigated a mechanical recycling using re-processing (extrusion – injection) in 4 different cycles. The ABS/HFFR (phosphate or melamine) compound fabricated at **SITRAPLAS** facilities has been reprocessed several times by extrusion and injection cycles to know how many times a material can be reprocessed before losing its applicability in the required application. **Tecnalia** carried out recycling studies collaborating with Sitraplas. **After multiple extrusion, the partners sustain that it is possible to obtain recycled polymers containing HFFRs.** The project results show retention of %85 of mechanical and fire retardancy properties after 3 subsequent extrusion processes.

SC 5: Mechanical Recycling of electrical cables with PVC/HFFR (ATH, MDH)

The Swedish project "Recycling of electrical cables with focus on mechanical recycling of polymers in end-of-life cable" (38), funded by Vinnova and coordinated by Swerea IVF has investigated mechanical recycling at Stena Recycling plant using different technologies at various steps after sorting: Cutting Granulation, Fluid Bed separation, PlastSep process, Drying process, Electrically Separation (KWS and also EKS by Hamos), Melt-filtration (by Gneuss) in order to recycle different metals, plastics and flame retardants from cable scraps.

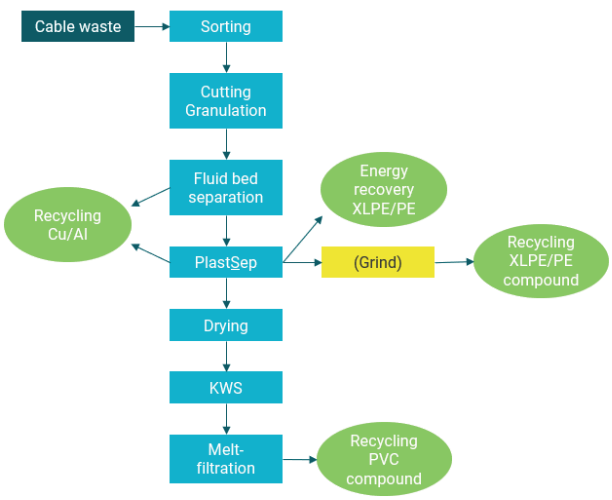


Figure 5-8: Mechanical Recycling Process involved in the Swedish Project

Fluid Bed Separation and "PlastSep" are used to separate plastics compounds from metals. Then, "PlastSep" is also used to separate different cable plastics compounds by gravity (density) separation and isolate the plastics compounds containing HFFR. However, to separate HFFR from PVC the partners tried to proceed through different steps: drying, electrical separation and melt-filtration. As project results, the project reports that after melt-filtration it is possible to obtain PVC with only %20 of HFFR that can be reused for lower-quality applications. Pure HFFR cannot be obtained (PVC fractions are always present, which reduces the mechanical properties of the flame retardant). HFFR and PVC cannot be separated in the PlastSep by gravity (density) separation or in KWS or EKS separator.

The project partners sustain that cable scrap with HFFR and PVC are usually mixed at the cable recycling plant **(often not pre-sorted). It is challenging to avoid mixing PVC and HFFR cable waste. So, sorting cables needs to be improved** to avoid spreading hazardous substances and improve the purity and quality needed to recycle PVC plastics.

SC 6: Sorting Technologies in mechanical recycling of HFFR plastics from WEEE

Two selected European-funded projects investigated the sorting technologies for the mechanical recycling of HFFR plastics from WEEE, CLOSEWEEE and REWARD-WEEE.

- The CLOSEWEEE project had two main goals: **investigate the sorting technologies of BFR plastics** and PFR plastics (PC/ABS/PFR and ABS/PFR) and to recycle BFR plastics by Creasolv (25) process (developed and patented by Fraunhofer) at Coolrec plant. Regarding the first goal different technologies have been studied: a separation process including LIBS technology (provided by Gaiker) for sorting bromine-free plastics, NIR/MIR sorting of plastics by polymer nature, mechanic/pneumatic separation steps, removal of metals, washing and drying steps of the recovered samples. As project result, after the sorting phase the bromine content in all bromine-free sorted fractions is demonstrated to be below the limits established by the directive RoHS.

- **The REWARD-WEEE project had a goal to sort BFR plastics and PC/ABS/PFR HIPS/PPE/PFR from WEEE.** At the Dolphin Metal Separation plant, different polymers and metals sorting and separation processes were demonstrated. Those concerning the separation of polymers were: density separation (magfluid), colour sensor sorting, dual-energy x-ray sensor sorting for the separation of halogenated polymers, Density separation to remove light plastics from heavy plastics, and Near-Infrared sensor sorting for the separation of polymer types. As a project result, **using x-ray sensor sorting a good rate of separation of halogenated plastics from non-halogenated plastics was achieved.**

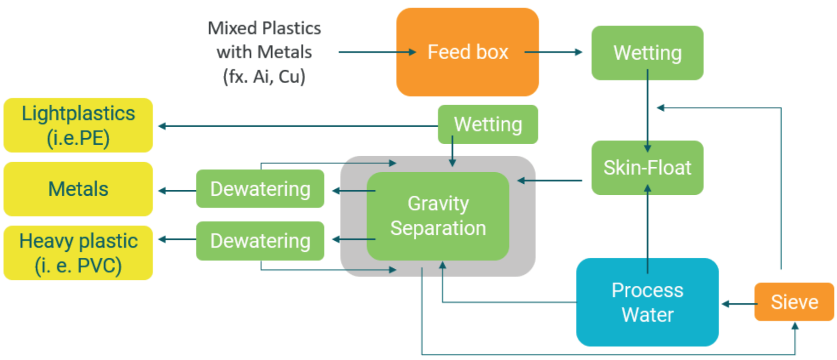


Figure 5-9: PlastSep Process as Intermediate Step

SC 7: Recycling Methods for recovering pure PIN flame retardant from WEEE

- Some selected patents claim to allow to recover pure PIN flame retardant from WEEE. This is the case with 3 of SABIC's patents, 1 patent belongs to ANHUI CHAOYUE ENVIRONMENTAL PROTECTION TECHNOLOGY COMPANY and UNIVERSITY OF SCIENCE AND TECHNOLOGY OF CHINA and 1 to PANASONIC.
- SABIC's patents are centred on the dissolution and separation of the compound and recovery of the bisphenol A from WEEE (PC/ABS containing PFR bisphenol A bis(diphenyl phosphate). Their patents **refer to the chemical recycling technologies by depolymerisation method using alcoholysis or ammonolysis.**
  - **EP2746249:** Heating a polycarbonate-containing composition (comprising PC and PFR) in the presence of alcohol and a catalyst at a temperature of 70 °C to 200°C and a pressure of 50 mbar to 40 bar for a time sufficient to depolymerize the polycarbonate to provide a dihydroxy aromatic compound and a dialkyl carbonate. Given the presence of PFR containing BPA, using a transesterification catalyst with non-neutralizable groups is recommended (the basic catalysts are unable to depolymerize the PC containing this PFR and therefore do not allow recovery of the BPA).
  - **US8846858:** Dissolution of flame retardant through solvents, formation of a filterable suspension of FR and ABS and/or PC and use of a second solvent for new dissolution of the FR in one of the two polymers. Finally, PC and/or ABS are depolymerized by heating with alcohol (methanol) and a transesterification catalyst for recovering the bisphenol A and the dialkyl carbonate.
  - **US9328046:** Non-catalytic method for carrying out ammonolysis of polycarbonate-containing plastic that also contains acrylonitrile-butadiene-styrene (ABS), and/or a phosphorus-containing flame retardant, such as bisphenol A bis(diphenyl phosphate) (BPADP). Surprisingly, a process is provided that allows the separation and recovery of bisphenol A and urea from recycled plastics of low polycarbonate content through selective depolymerization. The ammonia solution is of sufficient strength to selectively sever the ester bond of the polycarbonate and to form the ammonium salt of bisphenol-A (ammonium phenolate) which is soluble in the aqueous phase. A two-phase system is formed, namely a solid phase and a liquid phase of which the liquid phase contains both urea and the ammonium salt of bisphenol-A. Separation of the phases is achieved by filtration.
- The patent of ANHUI CHAOYUE ENVIRONMENTAL PROTECTION TECHNOLOGY COMPANY and UNIVERSITY OF SCIENCE AND TECHNOLOGY OF CHINA (**CN111704632**) claims to allow the recovery of pure triphenyl phosphate from waste circuit boards by mechanical recycling method using

CO_2 to extract HFFR. The method includes the following steps in sequence: 1. Coarse crushing, 2. Fine crushing, 3. Electrostatic separation of the polymers, 4. Extraction of the CO5 ,2. Using CO_2 as an extraction agent to recover FR in the supercritical fluid.

- The PANASONIC's patent (**JP5006523**) claims to achieve a high purity boehmite that can be reused in high-value electronic products. The chemical recycling method regards PP, ABS, Polyamide, PMMA, Epoxy Resin containing IFR (aluminium hydroxide) from WEEE treated at high temperatures. Aluminium hydroxide is used as an inorganic flame retardant in polymers. A heat treatment between 450 – 200°C or 450 – 360°C was implemented, dehydration of the FR and a thermal decomposition of the plastic were considered simultaneously.

SC 8: Recycling Methods for recovering Polymers from WEEE with a flame retardancy property

Some selected patents claim to recover polymers mechanically with a flame retardancy property from WEEE. This is the case of HEFEI LIXIAN ELECTRIC POWER ENGINEERING's (**CN108342011**) and SHARP's (**JP2014214195**) patents. The different halogen-free flame retardants (aluminium and magnesium hydroxide in the case of HEFEI and phosphate-based FR in the case of SHARP) are added to the polymers (PE, PE/EVA in the case of HEFEI and PC/ABS in the case of SHARP) during the recycling process before the extrusion step. The recycled materials have good flame retardant properties.

6 TALKING TO EXPERTS: INTERVIEWS REPORTS

What do polymers recycling experts think and know about PIN FRs impact on recycling and the available technologies?

This section reports detailed interviews with **8 experts from 5 institutions**, selected through the SoA review in the previous chapters. The selected institurions include industry and academia, inside and outside EU. The experts have been asked to answer more generic questions on FRs, describing their own experience, and some specific questions concerning sorting, mechanical recycling, chemical recycling, or solvent-based recycling. The questions were provided through a questionnaire and then face-to-face interview were organised. The reported text is a verified summary of these conversations.

UMASS LOWELL

6.1 University of Massachusetts Lowell (USA)

Wan-Ting (Grace) Chen is an assistant professor in the Department of Plastics Engineering at the University of Massachusetts Lowell (UMass Lowell), where she directs the Plastics & Environment Research Laboratory (PERL). Grace received her B.Sc. in Chemical Engineering from National Taiwan University, M.S. and Ph.D. in Agricultural and Biological Engineering from the University of Illinois at Urbana-Champaign, a postdoctoral training in Chemical Engineering from Purdue University. Her research group has extensive experience in the sustainability area, working with stakeholders including federal/state agencies, industries, and non-profit organizations. To date, her research group has been funded by the REMADE Institute, the U.S. Department of Energy (DOE), the Defense Logistics Agency (DLA), the U.S. Geological Survey (USGS), the U.S. Army Natick Soldier RD&E Center, the United Soybean Board (USB), and several companies.
Prof. Chen's current research focuses on four topics: 1) Advanced recycling of plastic and municipal waste into fuels, polymers, and chemicals, 2) Degradable and biobased polymeric material development, 3) Interfacial phenomena between microplastics and biofilms, and 4) green solvent/chemical design. Her

lab is equipped with reactors for hydrothermal processing of plastic waste and biowaste, as well as characterizations for fuel/polymers/thin films/microplastics. In addition, her lab has a license to specialized software, Hansen Solubility Parameters in Practice (HSPiP), to study polymer dissolution/precipitation behaviour, coating removal, polymer diffusion, and packaging failure mechanisms. The Chen Group's research program contributes to developing new multi-disciplinary materials on the topic of plastic/municipal waste recycling, biodegradable/biobased plastic material, microplastic/biofilm interaction, and sustainability analysis. Prof. Chen is affiliated with American

Evan Yu is a research assistant in the Department of Plastics Engineering at the University of Massachusetts Lowell. Evan has been working with professor Wan-Ting (Grace) Chen since 2019. His research focusses on investigating chemical recycling techniques of polymers including those found in electronic waste (E-waste) and toy waste. Evan has ample experience with the Hansen Solubility Parameters in Practice (HSPiP) software to investigate polymer dissolution and diffusion behaviour. He also has extensive knowledge of various polymer characterization methods including thermal and physical techniques. More recently, Evan has been working on developing a chemical recycling method for Polycarbonate (PC) from E-waste using safer solvents with the additional goal of flame retardant removal. Evan is a member of the Society for Plastics Engineers (SPE) and was the vice president of the UML student chapter of SPE during the academic year fall 2021-spring 2022. In addition, he is a member of the American Society for Chemical Engineers, Tau Beta Pi, and the UMass Lowell honours college. Thus far, Evan has published work about projects relating the removal of acrylic conformal coatings for electronic remanufacturing and the chemical recycling of mixed plastics from E-waste. He is currently working on another publication relating to the more recent research on solvent-based PC recycling.

Experience in the use of FRs

The most common FRs in our experiments are triphenyl phosphate (a PIN FR) and BFR, like triphenyl dibromide, while the most common polymers in EEE are PS, PC and ABS. these are a highly heterogenous mix of plastics sourced from EEE waste feedstocks including televisions, printers, computer cases.
Wire insulation and coded rubber are contained as well, while other polymers are present including HIPS, styrene, blends, epoxy-blends, and PC. The optimal recycling technology is highly dependent on the final use of the recycled plastics. **Regarding the FRs, there is a relevant focus on replacing BFRs due to their well-acknowledged toxicity issues.**
P FRs are then ideal candidates to replacing BFRs, however we still don't know any possible long-term effect.

Solvent-based recycling

Solvent-based recycling is a promising process that can be further improved, optimized and scaled-up.

Although solvent-based recycling offers the possibility of purifying plastics and separating FRs, these tend to accumulate through different mechanical recycling iterations. Solvent separation is therefore suitable for different type of polymers; however, some limitations occur. These primarily concern thermoset polymers, which cannot be recycled due to crosslinks.

The presence of additives in plastics represent indeed an issue to be addressed to obtain an effective recycling, **FRs separation is usually obtained via polar solvents (methanol) while BFR prefers an unpolar solvent (toluene), which implies higher toxicity for the whole process.** BFRs can also benefit from supercritical fluid separation.

Plastics separation processes

Concerning the separation of plastics, the most effective separation technology is still the salty water floating-based separation. This is because different polymers have specific densities that can be separated (Floating Vs. Sinking), resulting highly economically viable and simple. **Water-based separation can be used at large industrial scales and results environmentally friendly. This separation also allows to separate FR containing plastics exactly due to the higher weight.** Water-based separation allows indeed to reduce feedstock heterogeneity and to concentrate feedstock upon polymer type. It is for instance easier to separate high-density polymers (PC) vs. low-density polymers (PS). The amount of salt can also be modulated to enhance the separation. These sorting technologies cannot however tell you the FR content. Some drawbacks also occur with using this technology. For instance, HIPS / PS separation is not feasible[29]. **Concerning more advanced separation means, including optical-based sensors, better technology and investment are still demanded to implement them and it is still complex to transfer at larger industrial scale. Among these, NIR results as the most promising.**

AI can support sensor-based sorting and optical (VIS) and can also support the separation upon external shape, colour and transparency (i.e., bottle). However, optical technologies don't allow to infer the internal contents of materials. Efforts must be then made to improve both HW and SW sides.

Final considerations and conclusions

- **Most promising recycling technology depends on what the end application of the plastics is** (e.g., if we want to be able to still use for example the same cell phone case as the same case).
- **More research [exists] on BFR, as these types of FRs were the first ones developed for EEE** and then other types were developed to replace them because brominated kinds have toxic additives.

[29] This is doable, but has to be done manually by checking the FTIR spectra carefully. HIPS should have the spectra for rubber, which PS may not have. However, today's automatic IR sorting technique cannot achieve this yet.

- **We still don't relly know the long-term effects of the phosphorus Flame retardants 100%** as we do with the brominated content.
- **In conclusion, if you want to design a polymer which is natively recyclable, PIN FRs must be preferred while BFR must be discarded since the polymer design phase.**

6.2 NORNER AS (Norway)

Carlos Barreto-Soler is a Senior Researcher at Norner, with a robust professional and academic background supporting his work in plastics. His expertise is in sustainable composites and circular economy. He has gathered his insight and expertise from about 14 years' experience working at Norner and a solid education comprising a PhD in Polymer Chemistry and Applied Nanotechnology (University of Oslo (NO) 2013) and a Master in Advanced Materials (Chalmers University of Technology (SE) 2008).
Carlos has been delivering and managing projects supporting Norwegian and International industry players through B2B, and Norwegian and EC-funded projects. Carlos contributes to H2020-NONTOX as leading the polymer upgrade of mechanically recycled and purified polymers from WEEE, ELV and C&DW. He is also leader in H2020-REVOLUTION, of the tasks working on the project material development and contributes with his expertise in developing circular thermoplastic composites and compounds.

Experience in the Use of FRs
The most important plastics containing FR (such as BFR or PIN FR) in EEE when it comes to recycling are polycarbonate, ABS, PS/HIPS and polypropylene. These come from a wide range of different waste streams like small domestic appliances and are generally shredder residues. These streams are mostly characterized by Br content; however the characterization of PIN FR is relevant to enable the quality of mechanical recycling.
Even though PIN FR are considered the cleanest flame retardants, they could affect severely the recyclability. It would be indeed important to keep a good mapping of both types of FRs. However, this is still missing since recyclers are currently only characterizing the bromine content.
The target of most of the R&D projects, and the discussion ongoing with technology providers, is then about solvent recycling to purify plastics containing BFR. The solvent recycling is so performed complementarily to separation by density and as a further treatment by optical sorting.

Mechanical recycling and separation

Mechanical recycling is applied with success on open loop WEEE streams while at present most of the ELV plastics end up in incineration

Closed loops of PVC from windows are deployed while closed loops for PVC flooring are under discussion.

Mechanical recycling is still a relevant player in plastics recycling and about %50 of materials can be recycled via mechanical recycling and purification; chemical recycling is not yet a real alternative.

Mechanical recycling is indeed worth for recycling materials falling in the density below 1.1 g/cm3, which comprise a broad variety of plastics. At the same time, **the introduction of optical methods can support addressing issues related to separation of mono-materials.** Materials from open-loop recycling from this density fraction can not, in many instances, exceed 2000ppm bromine and are not considered hazardous when critical chemical substances are below a certain threshold (this is normally controlled by the recyclers).

Typically, materials with a density above 1.1 g/cm3 will require optical sorting to separate the polymer type and solvent recycling to purify from contaminants that could be hazardous or that will hamper mechanical recycling into products in one or another way. These optical methods are slowly approaching the market.

In the fraction 1.1-1.0 g/cm3 styrenics are recovered. This fraction includes mainly HIPS/PS and ABS and is separated into HIPS/PS and ABS by electrostatic separation. Furthermore, below 1.0g/cm^3 polyolefins are recovered as PE/PP mixture, contaminated with foams and wood, which is further enriched towards a higher content of PP by optical separation

In all streams Br content is typically controlled, however the content of non-bromine-based flame retardants, which could be latent acids or latent bases, should be controlled to mitigate quality issues during the subsequent upgrading stages.

Surely, this kind of recycled plastics cannot be used for food-grade (food contact) applications.

Solvent-based recycling

Solvent recycling should be categorized under the “mechanical recycling” group for the sake of clarity.

This is also a mean for decontaminating plastics, - for instance, a process like CreaSolv® (25) which recover styrenics (PS/HIPS, ABS), PC and polyolefins.

Solvent-based processes are indeed suitable for recycling “heavier fraction” of plastics due to the higher contamination in it but could well be used in lighter fractions that could require decontamination.

Solvent-based recycling allows easier decontamination and also results effective for plastics blends (PCs, polyolefins, styrenics, etc.) and highly contaminated plastics.

The aim of the purification process has been then to recover plastics, and not to recover directly flame retardants.

HFFR (halogen-free flame retardants) cannot be recovered as “pure”

substances, and their recycling seems not even economically feasible. FR containing stream from solvent separation usually contains both BFR and HFFR residues; recovered FRs are therefore usually discarded (incinerated).
Solvent based recycling is also particularly worth for EEE, and even including cables, which usually result as the most challenging material due to the higher number of additive substances contained.
The best upstream separation technology to be coupled with solvent recycling is optical and includes NIRs. There is however a problem concerning "black" materials that should be addressed.

Final considerations and conclusions

- **Mechanical recycling (including Solvent recycling) could be an alternative to recover about %50 of WEEE and ELV.** Mechanical recycling by density separation in open-loop recycling can recover materials up to a density of 1,1 g/cm3 (HIPS/PS, ABS, PP/PE) with Br content below 2000 ppm. Further optical sorting and electrostatic sorting can result in good quality HIPS/PS, ABS, and PP/PE streams. This density fraction can take advantage of solvent recycling if contamination with specific substances is an issue
- **Materials with $d>1,1g/cm^3$ rely on optical sorting (with the limitation of black colour) and on solvent recycling to remove specific substances (e.g., Br flame retardants and halogen-free FRs)**
- **In all streams of mechanical recycling Br content is typically controlled, however the content of non-bromine-based flame retardant that could be latent acids or latent bases should be controlled to mitigate quality issues** during the subsequent upgrading stages as polymers prone to hydrolysis can be degraded during typical temperatures for thermoplastic processing.

Gaiker
MEMBER OF
BASQUE RESEARCH
& TECHNOLOGY ALLIANCE

6.3 GAIKER TECHNOLOGY CENTER (Spain)

Jesus Ballestero Maestu*, with a degree in Chemical Sciences obtained in 1989, has more than 30 years of experience in flame-retardant plastics and composites design, development, and fire characterization. Nowadays* ***responsible for the reaction to fire laboratory and projects regarding flame retardancy at the GAIKER TECHNOLOGY CENTRE****. He is also representative of EGOLF (European Group Of Laboratories of Fire)*

Sixto Arnaiz, PhD (Chemical Engineering), MSc (Industrial Chemistry) has been **contracted researcher at GAIKER** since 1998 and currently **is responsible for the Recycling and Circular Economy Team**. He has been involved in R&D and innovation projects related to waste management (packaging waste, end-of-life vehicles, waste electric and electronic equipment, construction and demolition waste, etc.) covering the following recycling and recovery technologies: (1) development and application of advanced separation and automatic identification and sorting operations for waste streams treatment, fractions concentration (plastics, metals, etc.) and material loop closing, (2) Application of chemical processes for the feedstock recycling of plastic wastes, (3) Technical and sustainability assessment of materials, products, operations and processes.

Experience in the Use of FRs

PIN is the family of FRs currently most investigated in R&D projects. Through these projects, GAIKER has learned that the most common PIN FRs contained in EEE are ammonium polyphosphates, melamine polyphosphates, melamine derivates, phosphinates, phosphonates and phosphates, etc.

Nevertheless, the focus of R&D projects, related to the recycling of FR plastics from WEEEs management, revolves almost exclusively around BFRs.

GAIKER has used PIN FRs to improve fire behaviour in plastics for building (mainly for wall/ceiling/floor coverings and cables) and transportation, including railway and buses (mainly for wall/ceiling/floor coverings, seat shells, cab housings and external walls).

The most common plastics which contain PIN as flame retardant are: (i) thermosetting resins such as polyester*, phenolic, furan, epoxy, acrylic and polyurethanes, (ii) thermoplastic matrixes*, such as *polyethylene*, and *polyamide*, (iii) *gel coats* and (iv) rubbers. For the specific case of EEE, GAIKER retrieved BFRs from housing and plastics parts of devices like televisions, screens or IT&T equipment, other components, including, printed circuit boards and spare elements such as jackets or insulators in electrical cables.

Separation and sorting technologies are mainly focused on BFR plastics (e.g., density separation, sensor-based sorting using spectroscopies to recognise materials or elements - i.e., LIBS to identify Br).

For very demanding applications, such as large surfaces and internal walls in railway or building sectors, an amount between 20-30 % of PIN FRs content in the final composite is typically necessary to achieve the strict requirements demanded.

The presence of FR in plastic will indeed affect its use as a recycled material and the approach to recycling it.

Plastics separation and Sorting

Recycling of FRs plastics has focused on solvents to dissolve polymers. Although, for separating additives from plastics containing BFR, the CreaSolv®

Process has been developed precisely for this purpose and it is a very well-known example. **There isn't**, however, a**ny specific regulation yet to recycle plastics containing PIN FR.**
Sink-float is a typical industrial practice in plastic recycling, it offers a compromise between the investment and the capability of separating BFR and non BFR-containing plastics. **Nevertheless, sensor-based technologies are the natural candidates to improve the separation process, however their uptake demands investments.**
At the state of the art, there isn't any dedicated technology which can discriminate between BFR and PIN FR containing plastics. The separation of brominated plastic is demanded due to legal requirements on the restrictions of hazardous substances during recycling. However, **FR-based sorting is not practiced.** It can be theoretically possible to predict the amount of FR just by classifying the source of plastics (WEEE), or more precisely parts or components contained and only if there is a system which is well trained against a complete set of reference samples.
Chemical elements, that occur when PIN flame retardants are used could be detected using the same approach, but the development and testing of specific sorting models would be required.
Separation technologies such as shredding, screening or even automatic identification and sorting of undesired material fractions, are necessary to condition the waste input in physical format or in composition.
Separation technologies such as filtering, rinsing or crystallization are necessary to purify products. Another possible solution to mitigate the sorting difficulties is to standardize FRs

Mechanical recycling
Mechanical recycling offers both advantages and disadvantages. Advantages include: (i) well stablished method and simple operations, (ii) it requires moderate investments, and it can be easily scaled-up, (iii) it produces marketable mechanically recycled plastics when dealing with known/controlled waste streams.
Disadvantages (non-exhaustive) of mechanical recycling are conversely: (i) **they cannot deal with very mixed, aged or waste plastics including additivities and strongly rely on the quality of the input waste stream,** (ii) they can produce cross contaminations, (iii) the quality of the mechanically recycled plastics is always lower than the virgin plastics. This process is suitable for both closed loop and alternative loop.

Chemical recycling
Concerning instead the chemical recycling processes, different chemical recycling technologies are suitable for different plastic waste feedstocks and including: (i) thermochemical technologies, such as *pyrolysis* and *gasification*; these are suitable for mixed plastic wastes which are turned, respectively, into "*pyroil*" and "*syngas*"; (ii) Solvolysis, these processes are more specific and can be applied to specific plastic wastes as polyesters or polyurethanes. Economic

barriers are, however, associated with the required investment linked to the scale economy of the process. **Thermochemical recycling processes can be applied to virtually any type of plastic, but they are not selective towards single organic compounds but organic fractions.**

It seems however that the presence of any chemical substance in the reaction media could either interfere with the reaction or mix with the reaction product (FRs). The presence of traces of certain undesired substances, or chemical elements, can restrict the final use of a chemically recycled polymer and the final users will not accept those materials for high-end applications (including FRs).

Finally, Hydrolysis and pyrolysis cannot be compared. Pyrolysis is a general-purpose route - polymers and organics are transformed into gases, “char” (a carbonaceous solid) and “pyroil”. Solvolysis is conversely a very specific route in which only some selected polymers are transformed in a specific monomer or chemical.

Solvent-recycling specific questions

Solvent recycling can be performed if a solvent is found which is highly selective for a given polymer.

Nevertheless, **the economy of the process besides sustainability and safety issues related with solvent uses need to be considered.** Primary feedstocks include polyolefins (using a mixture of xylenes) or styrenics plastics (using the specific mix of solvents developed for the CreaSolv® Process) (25).

Final considerations and conclusions

- **The focus of R&D when it comes to recycling of plastics, especially from WEEE, is mainly on BFR**
- **There is not a specific regulation to recycle PIN FR**, but their presence affects the material to recycle
- **Separation, purification, and automatic identification are needed; FR standardisation can also be useful**
- **Mechanical recycling is cost-effective, but cannot deal with the absence of quality of the input stream, chemical recycling is affected by any chemical substance in the reaction media, while solvent-based recycling is effective, but economics and safety needs to be considered**

6.4 FRAUNHOFER LBF (Germany)

Dr. Elke Metzsch-Zilligen heads the Additivation and Durability (AD) department at the Fraunhofer Institute for Structural Durability and System Reliability LBF in Darmstadt. The department is part of the Plastics Business Unit, which is headed by Prof. Dr. Rudolf Pfaendner. Dr. Metzsch-Zilligen has already been working at the institute, which was run as the German Plastics Institute (DKI) before its integration into the Fraunhofer-Gesellschaft, since 2011. Prior to that, she worked in the food industry for several years.
She studied chemistry at the University of Cologne, receiving her doctorate in physical chemistry in 2006. Her research focused on the structure elucidation and thermal degradation behaviour of high-temperature homo- and copolyamides using direct inlet mass spectrometry.
Her work in the AD department focuses on both the synthesis of new additives and the selection and evaluation of optimized additive systems for thermoplastics tailored to specific applications. In recent years, ecological issues have also increasingly come to the fore: For example, the increasing replacement of virgin materials by recyclates places special demands on additivation, as these end-of-life materials may exhibit processing- and use-related damage. This requires adapted post-additivation in order to be able to keep the plastics further in the material cycle. The AD department in the Fraunhofer "Circular Plastics Economy" cluster and the Fraunhofer "Waste4Future" lighthouse project are working intensively on this topic.

Dr. Michael Großhauser has been a scientist in the Additivation and Durability department at the Fraunhofer Institute for Structural Durability and System Reliability LBF in Darmstadt since 2017. The department is part of the Plastics division, which is headed by Prof. Dr. Rudolf Pfaendner. The plastic division was run as the German Plastics Institute before its integration into the Fraunhofer-Gesellschaft in 2011. Prior to 2017, he worked in the ionic liquid producing industry.
He studied chemistry at the Ruprecht-Karls-University of Heidelberg, receiving his doctorate in inorganic chemistry in 2015. The focus of his research was the synthesis and physical and spectroscopic characterization of single molecular magnets.
His work in the Additiviation and Durability department focusses on both the

synthesis of new additives and the selection and evaluation of optimized additive systems for thermoplastics, tailored to specific applications. In recent years, on the one hand new regulations caused the need for reliability, and on the other hand new sustainable ecological issues have also increasingly come to the fore: E.g., the increasing replacement of virgin materials by recyclates places particular demands on additivation, as these end-of-life materials may show damage due to processing and use. This requires adapted post-additivation to be able to continue keeping the plastics in the material cycle.

Experience in the use of FRs

The most important PIN FR are: aluminium hydroxide, magnesium hydroxide, red phosphorus, melamine cyanurate, melamine polyphosphate, DEPAL. These FRs are present in cables, TV screens or some parts of lamps, and electronic housings.

Concerning recycling, **mechanical recycling, combined with solvent-based recycling, appears to be the most promising technology for recycling plastics containing PIN FRs** from an economic point of view.

Before operating mechanical recycling, a special type of sorting like sensor-based sorting is also needed.

In Europe, about 70% of products containing FR are halogen-free FR due to regulations and ecotoxicological aspects. Contrary in Asia, where the most flame retardant products are BFR. Regulations widely differ across countries.

Sorting and separation

The correct selection of polymer by type is the most relevant goal of an effective plastic waste sorting technology. The main criteria to select the optimal sorting technology is exactly what you want to obtain at the end of the process. If you want, for instance, to separate the flame retardant contained in the polymer you need a solvent-based process, or chemical recycling. A solvent-based process is precisely what you need to do this job. **A sorting step is always necessary, independently of the recycling technology you're going to adopt.** Concerning the chemical recycling, instead, it is a highly specific recycling technology, and it allows to recycle only a small number of materials – mainly poly-condensates or polyamides. Possibly polyolefins can also be recycled via chemical recycling (pyrolysis). **You should anyway always consider that you get monomers from the chemical recycling process.**

Some technology also allows separation upon flame retardant type, including PIN FR.

Mechanical recycling and separation

Mechanical recycling is the easiest way to recycle plastic materials, especially if you want to newly melt them. **Solvent-based recycling is conversely more energy-intense, and surely you need to use a solvent.** Furthermore, you need to circularize the solvent within the process, which can be a very energy-intense process.

There is an impressive amount of electronic wastes (WEEEs), and they are the

largest group to be recycled. EEEs are not made of a single part, but appliances (washing machine, television screen or computers) are composed of multiple components; there is also a country-related variability due to regulations (i.e., USA, Japan, etc.).
Mechanical recycling doesn't however allow for recovery of FRs. In post-consumer material it is quite challenging to perform closed-loop recycling. Usually, you have open loops due to material degradation. Materials are instead currently downcycled a lot whenever the obtained material is not burnt

Final considerations and conclusions

- **Coupling with other technologies**, including sensors and AI may result helpful in improving the sorting effect
- "**Design for recycling**" is the answer to optimize recycling processes. "Design for recycling" (sustainability by design) allows indeed to perform a more efficient recycling process. Tracers may be introduced to support the separation process.
- **Information will be vital for sustaining recycling** – especially to train a dedicated artificial intelligence
- Artificial-driven system would indeed allow improving recyclability, **availability of standardized data, and transparency, therefore supporting the introduction of a recycling-oriented marketplace for private sector** with all necessary information to stakeholders.
- **In Germany, for instance, many companies and institutions are trying to define the standards concerning recycling and related information.** Ideally a "product passport" could be a powerful tool for supporting a circular product lifecycle. Nevertheless, you cannot recycle without the proper information.

6.5 MGG Polymers GmbH (Germany)

Before moving to the recycling business, Mr. Slijkhuis has been working for a global EEE manufacturing company – Flex Electronics (USA) in the role of operational field and supply chain maintenance manager. **Over the years, Mr. Slijkhuis acquired a remarkable experience in the procurement of raw materials for electronics and dealing then with their supply for the manufacturing of new EEEs goods.**
In the early 2000s, he moved to MGG, leading polymer recycling company which operates joint-ventured facilities located in China, UK, California, and Austria focusing on the recycling of electronic wastes (WEES).
Since 2007, he has been involved in discussions with institutional stakeholders and concerning exactly the role of FRs in plastic recycling also attending to

dedicated institutional working groups which included REACH and focusing on the related regulatory and authorization procedures.
His expertise concerns in detail the handling of WEEEs containing brominated flame retardants for electronic applications, also having knowledge about the emergence of PINs FR as alternative in industry. Currently, he works as external consultant at MGG.

General experience and considerations about the use of FRs
The final goal of recycling industry is to obtain recycled plastic from WEEEs at the same quality of new materials, for allowing its use in the manufacturing of newly made EEEs goods. This is important since many electronic applications need FRs. So, we, as recyclers, are also interested in creating the optimal recipes for flame-retarded plastics. The FRs are relevant considering the need ofmanufacturing recycled EEEs products capable of offering the same flame retardancy features.
Legal tresholds are already in place for BFR and not yet for other types, even if PIN FRs first initiatives are in place. At the same time, despite former restrictions on certain brominated flame retardant substances, the BFRs which industry uses today are perfectly allowed (and known) substances. There are however ongoing issues related with BFRs which are also driven especially by the regulators.
As far as PIN FRs are concerned, at the moment we don't perform any separation of PIN FRs, moreover we don't do any analyses and we do not have any knowledge about the separation of PIN flame retardants.
Considering instead the use of PIN FRs, there are two main concerns: one is related to phosphoric FRs (P) and the creation of phosphoric acid in combination with heat and moisture. The second concern is regulatory, meaning that BFRs are well known in terms of legislation and how to deal with it, while **less knowledge exists on PIN FRs regulatory future, which may generate worries about possible changes.** For instance, if we move away from BFRs, and one PIN FR suddenly becomes restricted, its separation from the waste stream can generate new unknowns.

Separation and mechanical recycling
Our company is capable to cover the entire value chain from downstream the waste collection, processing a 50kton/year of polymers. MGG focuses on the mechanical separation, before performing an extrusion step and the introduction of additives. Our company uses an extruder setup which allows to add up **to 7 additives in only one step of extrusion.** The separation process also foresees a step of metal and battery separation, which are routed to smelters. We just need to analyze the plastic flakes before being extruded to verify if thesecomply with the EU legislation. Following, we introduce additives to confer the functionalities as it is required by clients, thus ensuring certain physical features including, strengths, surface features, impact resistance, UV resistance, etc.

Products are made in alignment with the costumers needs which are predominately high-tech.
The most fundamental plastics recycled from the WEEEs streams PP, HIPS, ABS, and since not too long PC-ABS. With the exception of the latter, the former three can be then easily separated based on density, allowing to identify the BFR containing plastics exactly based on their higher relative weight. Flame retardants are characterized by a high density and one of the fundamental steps in the recycling process today is that most FRs are separated by a density separation. We can imagine a different relative weight for PIN-FRs, but an impact on weight nonetheless.
As MGG, it is not hugely important which FR we use yet, but it may become. As recyclers, we still don't know so much about the PIN FR. This is an actual issue for the industry considering that, for instance, the Stockholm Convention, or other regulators, can decide to suddenly restrict some substances. This can also happen in the European legislation level or in other regulatory bodies.

Other recycling technologies
From an energy point of view, the best method to maximize the sustainability is the mechanical recycling. This preserves indeed all the original energy stored in the polymer chain. Basically, we just separate the plastic polymer and recreate a polymer maximizing the energy efficiency.
However, considering that is impossible to recycle all plastics, chemical recycling methods which are being developed now are an actual alternative, despite the physical recycling (based upon density) is still the most efficient and sustainable way for recycling plastics. Technologies like chemical recycling in fact are based on wrecking the chains: this allows to break the polymer chain, obtaining monomers that can be newly used for synthetizing a polymer. From an environmental sustainability point of view, these technologies will be more impacting since further energy is needed to recreate the original polymer.

Sustainability by design
Sustainability by design is already happening (to support). For instance, the **European Project PolyCE** is a highly successful Horizon 2020 funded project dedicated to the sustainability by design of plastic polymers. This project, is exactly looking at providing the most convenient solutions for designing something which can be easily recycled afterlife, and, at the same time, what can the recycling deliver to help designers creating new products.

Sorting
We are going **from mainly physical simple density sorting, to more high-tech separation** processes which include sensor based techniques (without entering into much details). Things are happening fast.

Marking/labelling
The marking of the plastics is **not relevant for mechanical recycling** for two

reasons: **marking is often not correct.** Unfortunately, a marking-based approach is not completely reliable, especially for product sourced in far east (i.e., manufacturers of cheap electronic goods). For instance, these **products may either not have the FR correctly declared on the labels or conversely, they can contain some flame retardant even if this it is not actually required in the product.** For these reasons, the labeling of plastics is not a reliable separation aid especially for basing a mechanical recycling process. Only in markets (not EU) when manual separation takes place people might look at them.
Secondly, recycling starts with shredding, which makes hard to find the marking on plastic flakes.

Conclusions
Concerning the FRs, for recycling company like MGG is a priority to duly observe the chemical legislation. A single substance is now subjected either to POP (ECHA), REACH (ECHA) and moreover to the Stockholm and Basel Conventions as well as specific industrial standards. All regulators can indeed produce different set of rules for a certain substance, and this implies that a same substance can be subjected up to 3 different regulations. Today we are facing discussions around thresholds that come back any 2 years because new rules are always changing. For creating a circular economy this should be simplified.
The industry needs one substance and one assessment.
We therefore need to know in advance if in the future any of these substances will be restricted; this is a key factor for the recycling industry.

If any PIN FR would become a restricted substance, it is extremely important for the recycling industry to have one simple screening measurement method to see what the amount of PIN is. As a consequence, a proper analytic method capable of detecting the correct amount of N and P must be provided, and without implying complex and time-consuming methodology like GC-MS (Gas Chromatography/Mass Spectrometry) on a daily bases by a laboratory.

7 CONCLUSIONS

Key outcomes
PIN FR and plastic recycling
Evidenced Gaps and recommendations

7.1 Key outcomes of this work

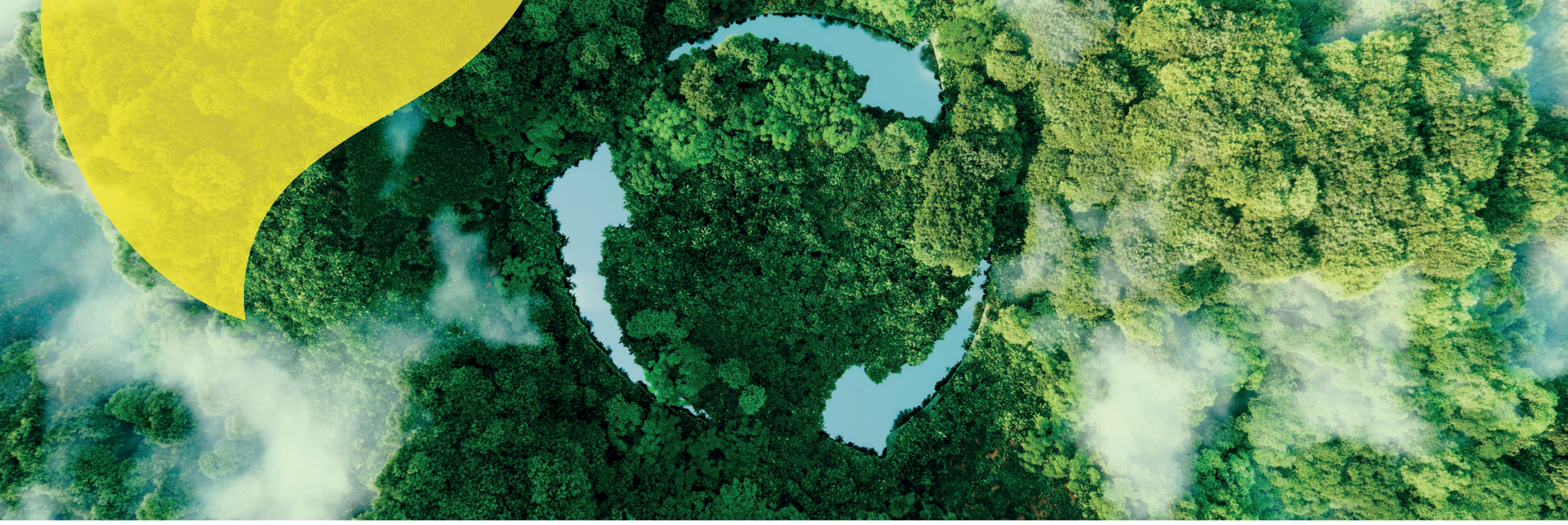
The current technology state-of-play, the emerging processes for plastic recycling and uses of PIN FRs have been mapped: *a comprehensive literature review and technology outlook have been elaborated to populate* **novel PIN FR Applications and Recycling Matrices**, but more data are needed. *A focussed analysis on WEEEs recycling has been added to the picture.*

PIN FR are nowadays applied in a wide range of sectors, including EEE, building and infrastructure, or transport. 40 different types of PIN FRs and 54 different plastic types or combination of plastics have been identified and represented in the form of 3 *Application Matrices* in this study.

PIN FR plastics mostly consists of PBT, PC/ABS, PET, PA6, PA66, PA6/GF30, PA66/GF30, PP, PVC, PU, epoxy resin, VE, phenolic resin, PU, and TPU. Moreover, recycled PA6/PP has been used as sealants or interior parts in the transport sector with some PIN FRs like APP, MC combined with MPP. The most common PIN FR used by industry is aluminium hydroxide followed by phosphorous based FR molecules.

By extracting 69 "cases" from a shortlist of 7 relevant R&D funded projects, 16 patents and 24 scientific papers/reports focussed on PIN FR plastic recycling, a framework relating polymers, FRs, recycling processes and outcomes and readiness of the technology has been created, as an original PIN FR *Recycling Matrix*, which is replicable, scalable, and expandable.

Pulled by a growing market and a legislative push for circularity and resource efficiency, WEEEs have been established as the most represented waste source category in the analysed literature. This can be attributed to the accelerated spread of electronics in every market, and linked to the relatively high content of FRs. Therefore, the efforts that must be committed for effective recycling are increased, justifying additional R&D investments and defining the most significant focus area for the PIN FRs. A set of *8 Showcases* of PIN FR recycling in the WEEEs sector have been selected looking at the shortlisted R&D literature.



7.2 PIN FRs and recycling
7.2.1 Key Findings

Despite a growing market where PIN FRs are taking the largest share among FRs, there is an evident and reasonable information gap of knowledge and specific regulations comparing PIN FR to BFR.

The present study offers a qualitative overview of the current recycling trends and policy targets related to PIN FRs. General trends have been spotted, supporting a better understanding of the priorities and the R&D efforts by both industry (products designers and recyclers) and academia.
Market, technology and legislation perspectives have been taken into account and the key findings are reported in the following.

What the market looks like - by reviewing different market analyses developed in the last 5 years, it is evident that analysts agree that PIN FRs will have a relevant opportunity to increase their business.

- ✓ PIN FR are gaining more and more market shares, becoming a leading segment, with a growth by %8 CAGR.
- ✓ In order to fully seize the business opportunity, the PIN FRs industry can take advantage of the incoming challenges for plastic recycling set by the EU Strategy for Plastics: providing an additional boost of 3.4 million tonnes of recycled plastics in 2025, compared to 2020. Looking at the key sectors of WEEE, Building & Construction and Transport, in practical terms this target means increasing the final recycled outputs by %40 for the automotive (prominent part of the transport industry), %15 for the building and %52 for the WEEE sector.
- ✓ The market of recycling additives, compatibilisers, and other stabilisers is growing with +%5 CAGR and is certainly a key element to be considered for the future of PIN FR plastics recycling, since additives specifically adapted for recycling can limit the impacts of polymer degradation during re-extrusion. Flame Retardants can also be applied as additives during recycling.

What the experts say - there is a general consensus that the effects of PIN FRs on recycling are not yet investigated enough, neither by industry nor academy. In short, the experts have confirmed that:

- PIN FR should be preferred since the design phase. However, even though they are considered as the cleanest flame retardants, BFR keeps being used because they are cheaper, historically established on the market, and because their FR-performance enables them to fulfill the requirement of many standards. This also justifies much more R&D research available with a focus on BFR.
- The presence of FRs in plastics can affect its use as a recycled material, as well as the approach to recycle it. In this sense the effects of the PIN FRs are still not completely known. In particular, latent acids or latent bases during recycling should be controlled to mitigate quality issues.
- While new technologies, such as chemical recycling, are being industrialised, mechanical recycling is cost and energy efficient, but it cannot deal well with the lack of quality in the input stream, thus, automatic identification using AI, sensor-based enhanced separation and purification are needed.
- There is not a specific regulation for PIN FR and this can create uncertainties for recyclers and contributes to keep the focus on BFR during the material selection. Besides, simple and unique standards for FRs, including screening and testing procedures are needed.

What R&D literature data suggests - the data-driven approach defined in this study can support and initiate an analysis. Through a preliminary observation of the case-studies in the PIN FR Recycling Matrix we have provided a bird's view to illustrate how this approach could be replicated with a systematic data collection. However, it must be stressed that to reach clear and reliable conclusions, expanding the database is necessary .

- We found that the most frequent appearances are related to P based FRs, while the most investigated polymers are PC/ABS, PC, ABS, PP and PET. The initial dataset includes %30 of case-studies where a full recyclability of polymers is observed.
- Mechanical recycling still results as the "state of the art" for plastic waste processing, but the available data also suggest that chemical recycling and solvent-based recycling have been used in most of the cases when full recyclability is reported to be the final outcome.
- The available dataset shows that there seems to be a correlation between the final quality of the recycled products and the capability of separating the flame retardant from the waste polymer feedstock during the recycling process.
- Even the composition of the PIN FR can be correlated to the Polymer EoL: a more heterogenous presence of FRs within plastic waste can affect the final quality of the recycled material output.

7.2.2 Recycling technologies overview

The general perspective defined through the interviews and the technology readiness of the techniques analysed in the R&D outlook, highlights that PIN FRs cannot, in general, be recovered as "pure" substances and that their separation is not yet developed at an industrial scale:

The most recurring recycling process described in literature is mechanical recycling, (it should be noted that additives were not considered) yet a number of reported cases in our study are unsuccessful in recycling the polymer without downgrading it. On the other hand, chemical recycling processes (e.g., based on the de-polymerisation of plastic polymers) can achieve significant results in separating PIN FRs. They are well represented in relevant patents and quite promising in obtaining virgin-like plastic materials from highly pure wastes (i.e., crPET), which can offer higher quality performances.

The solvent-based recycling process, instead, has been increasingly investigated especially in recent years and typically appears in patents. The interviewed experts also confirmed its potential as a true emerging solution.

The availability of cost-effective sorting technologies is an enabler for more efficient recycling of plastics, improving the overall output quality and thus determining relevant benefits in terms of economics and resource efficiency. Separating and sorting the different types of plastic upstream is also a way to increase the capacity of upcycling the post-consumer materials, thus directing the materials towards separated recycling processes, optimized according to their different plastic compositions (polymers, additives, blends, etc.).

This is also true when PIN FRs are involved. Especially at higher percentages, separating PIN FRs from plastics represents an aspect to be duly addressed during the recycling phase. With this specific purpose, different sorting technologies have been developed to effectively separate the various plastic types according to their compositions. Research has been performed mainly concerning LCD screen covers and also other electronic residues, including cases, cables, boards (39) etc.

More advanced separation technologies require sensor-based sorting and Optical (VIS), where AI can provide strong support. However, while sensors are slowly approaching the market, more complete and smart sorting solutions are still complex to transfer at a larger industrial scale. Among these, NIR results are the most promising and can be considered the reference choice to be coupled with mechanical and solvent-based recycling. **However, at the state of the art, there is hardly any dedicated technology for FR-based sorting.** It could be theoretically possible to predict the amount of FR by classifying the source of plastics (WEEE), with a smart system which is well trained against a complete set of reference samples.

7.2.3 Legislations overview

The legislative landscape can, on the one hand, push the use of PIN FRs indirectly, on the other hand, there is not any specific regulation yet to recycle plastics containing PIN FR.

During our interviews with industry representatives (Norner and MGG recycling, Chapter 6), they acknowledged that the latter point can create uncertainty and keep the attention on BFRs, since most recyclers are currently only characterizing the bromine content. However, there are key legislative drivers and trends pushing PIN FR, specifically, the requirements for safer products (**need for FRs**), the progressive ban of Halogenated FRs (**need to replace them with HFFRs**) and circularity (**need for recyclability and reuse**) – emerge from the following EU directives:

- ***The EU Strategy for Plastics***: establishing the main goals for plastic design, manufacture, use, re-use, and end-of-life management by 2030. The target is to bring the EU market for recycled plastics to 10 million tonnes by 2025.
- ***Directive 96/2002/EC on Waste of Electric and Electronic Equipment***: plastics containing brominated flame retardants need to be removed
- ***Directive 95/2002/EC on Restriction of certain Hazardous Substances in Electric and Electronic Equipment (RoHS)***: the available evidence indicates that measures on the collection, treatment, recycling and disposal of waste EEE are necessary to reduce the waste management problems associated with the heavy metals and flame retardants concerned

- ***According to the Commission Staff Working Document evaluation of the End-of-life Vehicles (ELV) Directive (53/2000/EC)*** in 2021, the most important POP-related issue for the treatment of ELVs relates to the presence and disposal of the flame retardant decabromodiphenyl ether (DecaBDE) and other POP-BDE in shredder residue, which needs to be taken into account in recycling. The disposal and recovery of waste containing such POPs is regulated through the POPs Regulation[31] (Article 7)
- ***Directive (EU) 852/2019 on Packaging and Packaging Waste:*** %10 max landfilling of municipal waste by %50 ,2035 recycling of plastic packaging by 2025 and %55 by 2030

All in all, as already pointed out, contrary to Brominated FR, WEEE and EoL vehicles directives do not present regulations for the case of PIN FR but can indirectly foster their adoption as possible replacements, provided that reliable recycling processes are available along with regulatory certainties. Investing (by recyclers and technology providers) and funding (by policy makers) are needed to ensure that the promising technologies reach the required capacity in a short timeframe.

7.1 Sustainability by Design strategies

A relevant contribution to support the recycling of plastic materials, from a sustainable life cycle perspective, is the adoption of sustainability-by-design principles, which implies a stronger link between designers and recyclers.

The observance of these principles would ease many recycling processes, including the most innovative solvent-based recycling procedures, by acting both on the polymer formulation and on the choice of additives, including Flame Retardants substances.
Any choice of plastic additives should, therefore, by design, consider the possible recycling steps and significantly ease the resistance of polymers toward the solvents commonly used in the recycling process (i.e., by reducing cross-links, especially in high resistance material applications, when possible).
In this perspective, Bromine-based FRs also show an intrinsic complexity in separating them from a polymer substrate using the known separation technologies or including the most innovative new solvent-based separation means. Solvent separation of BFRs demands more complex efforts, including the use of supercritical fluids or unpopular solvents like toluene.
Conversely, **P based FRs, which are usually introduced in lower amounts, can be more easily separated using polar solvents.**

[31] Persistent Organic Pollutants - Regulation (EU) 2019/1021 of the European Parliament and of the Council of 20 June 2019 on persistent organic pollutants (OJ L 169, 25.6.2019, p. 45–77)

Another strategy to foster plastic recycling and reduce waste could be attention to **minimising blends, or mixtures, of different polymers, which cannot be treated within the same recycling process.** Otherwise, specific processes are required to allow the effective recycling of polymer blends (i.e., the mixture with some rubbers is indeed associated with a very scarce recycling capability).
Reducing the use of thermoset polymers by industry would also support a more effective circularity in the plastics industry, given the impossibility of recycling them through the existing solvent-based recycling processes.
Overall, **wide adoption of sustainability by design principles can broaden the range of plastic substrates suitable for solvent-based recycling** with relevant outcomes in terms of circular economy development.

7.2 Gaps and Recommendations

To improve insights, policy and decision-making on PIN FR recycling it is essential **to support a better knowledge-base and information exchange between the different parts of the production and recycling value-chains.** Key recommendations hence include:

- **Improving the mapping and characterization of PIN FR content.** At the moment, this is still missing since recyclers seem to currently be only characterizing the bromine content. This approach should entail the definition of **specific testing and sampling procedures** to be standardised and the **creation of a centralised database**.
- **Support certainty of regulations about PIN FRs, avoiding redundancies if possible (one substance, one assessment).** This will push the industry and recyclers in particular to characterize PIN FRs on top of BFRs.
- **Fostering "sustainability by design"** schemes to enhance cooperation between producers and recyclers and supporting **FR standardisation** to help the recycling phase. It is to be noted that a similar approach is being followed by the Circular Plastic Alliance, which has been working to provide Design-for-recycling guidelines and address CEN and CENELEC standards. A list of 26 products exists with priority, which include the EEE and Building sectors (see Chapter 1).
- **Supporting the private sector in enhancing transparency** – e.g., introducing recycling-oriented marketplaces for the private sector. Besides, some experts suggested **FR content labelling systems and "products passports"** for particular streams (e.g., WEEE).

Research and development in FRs recycling are still largely dominated by BFR recycling. This means that there is a need to collect further data and knowledge on the impact of PIN FR on recycling. It is therefore recommended:

- **To take a systematic approach** (e.g., the "PIN FR recycling map") in building a **comprehensive database with PIN FR recycling cases**, to generate comprehensive data and knowledge on the possible relationships between the composition of PIN FR containing waste, recycling technologies, type of polymers, waste collection and separation, in order to support better insights and decision making.

- To invest in increasing the number of projects to investigate specifically PIN FRs impact on the recyclability of different waste streams. For example, it is recommended to address acid formation during the recycling process. Projects should include large-scale case-studies involving the whole value-chain. International cooperation is advised, to correctly represent the EU and extra EU value-chain, and also to better consider quality control differences, such as those in labelling and marking procedures.
- **Focus R&D on developing and upscaling novel separation, purification, and sorting technologies to enhance the impact of available treatment processes.** AI-supported NIR and optical technologies are most promising and should be tested on a suitable scale. Chemical elements, that occur when PIN flame retardants are used could be detected, but the development and testing of specific sorting models would be required. It should be noted that similar technologies are being developed for adjacent recycling streams (e.g., batteries), which could again lead to building R&D synergies in larger projects.
- **To accelerate and upscale testing of solvent-based recycling, with a view to separate PIN FRs, in a way that it is possible to consider its economic sustainability and safety aspects,** given its growing relevance to obtaining highly pure recycled polymers in a cost-effective way

8 ANNEX-1: DETAILED PIN FR APPLICATIONS (A COMMENTARY TO THE APPLICATION MATRICES)

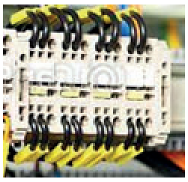
8.1 PIN flame retardants in EEE plastics

The use of polymers with PIN FR in the E&E industry is the most prominent one such as in cables, connectors, and switchers, electrical components, or engineering plastics for household appliances. The application matrix for PIN FR in the E&E industry is shown in Figure 2-2

*For **cables** used in the E&E industry, the most important plastics formulated with PIN FRs are: poly-ethyl-co-vinyl acetate (EVA), low density polyethylene (LDPE), polyamides (PA), polyethylene co-butene, polyethylene co-octene, polyethylene (PE)/EVA, PP, PVC, chlorinated polyvinyl chloride (CPVC), rubber, CR (chlorinated rubber), silicon rubbers, thermoplastic polyurethane (TPU), thermoplastic elastomer (TPE) and cross-linked polyethylene (XLPE) (5).*

Polymers such as CPVC and CR are formulated with zinc borate. The sources analysed reported a weight of zinc borate in these polymer types between weight-%2-5 (5) (40) EVA is formulated with numerous inorganic FR, including, AOH (weight-% 3-15 (5) (40) (38), ATH (weight-% 58-67 or weight-% 20-55 combined with P and/or N FR) (40) (41) MDH (up to weight-% 80) (40) zinc borate (weight-% 3-15) (40) (42) zinc hydroxy stannate (weight-% 3-15) (5) (40) (42). LDPE, polyethylene co-butene, polyethylene co-octene, TPU, TPE are formulated with AOH, ATH, zinc borate and zinc hydroxy stannates. The same weight (in weight-%) have been reported as for the case of EVA. Polyamides are formulated with inorganic FRs like, and with nitrogen FRs such as MC or melamine phosphate (5) (40). The combination of PE with EVA (PE/EVA) is formulated with inorganic FRs like ATH and MDH. PP is formulated with various types of FRs, including phosphorous FRs (alkoxyamine, APP with P-synergist, or PAPP), inorganic FRs like AOH, or nitrogen FRs such as MC, melamine phosphate, or substituted amine phosphate mixture. PVC is formulated with inorganic FRs such as ATH, the combination of ATH and ATO, or zinc borate. The sources analysed reported a weight of ATH between weight-% 5-15 and between weight-% 1-3 for ATO and the weight of zinc borate in PVC between weight-% 2-5 (5) (40). Rubber is formulated with phosphorus FRs such as TCP (5), or AOH, and inorganic FRs like ATH (40). Similarly, silicon rubbers are formulated with inorganic FRs like AOH, ATH and MDH. XLPE is formulated with phosphorus FRs such as Phosphonic acid ester, TPP, or tri (3-hydroxypropyl) phosphine oxide and their weights vary between weight-% 10-20.

*For **connectors and switchers** used in the E&E industry, the most important plastics formulated with PIN FRs are high temperature polyamides (PAHT), High impact polystyrene/poly(p-phenylene oxide) (HIPS/PPO), PBT, PBT/GF, PC/ABS, PET, PA6, PA66, PA6/GF30 and PA66/GF30 (40).*



PAHT is formulated with P FRs such as metal phosphinate. HIPS/PPO is formulated with phosphorus FR like RDP. PBT and PBT/GF are formulated with inorganic FRs such as AOH, and P FRs like metal phosphinate combined with N-Synergist (e.g MPyP). A blend of PC/ABS is formulated with various phosphorus FRs, including BDP, phosphate ester, RDP, or TPP. PET is formulated with BDP and metal phosphinate combined with N-Synergist (e.g MPyP). Polyamides such as PA6 and PA66 as well as their combinations with GF are formulated with AOH, ATH. In addition, PA6 is formulated with MC and metal phosphinate combined with N-Synergist (e.g MPyP), whereas PA66 is formulated with RP, melamine polyphosphate and metal phosphinate combined with N-Synergist (e.g MPyP). DEPAL along with AOH were found in some PA6 and PA66 combined with GF.

*For **electrical components** used in the E&E industry, the most important plastics formulated with PIN FRs are PC/ABS, PAHT, PBT, PET, PA6, PA66, PA6/GF30 and PA66/GF30 (40). The type of plastics and PIN FRs for electrical components do not differ significantly with the ones used for connectors and switchers (5).*

PAHT is formulated with P and N FRs such as metal phosphinate and melamine phosphinate. A blend of PC/ABS is formulated with various phosporus FRs, including BDP, RDP, TCP or TPP (between weights-% 1-5). PBT and PET are formulated with metal phosphinate combined with melamine polyphosphate. PA6 is formulated with MC and Metal phosphinate combined with melamine polyphosphate. PA66 is formulated with red phosphorous (RP), melamine polyphosphate and metal phosphinate combined with melamine polyphosphate. The combination of PA6 and PA66 with GF are formulated with DEPAL.

*For **engineering plastics** for household appliances used in the E&E industry, the most important plastics formulated with PIN FRs are HIPS/PPE, PC, PC/ABS, Polyisocyanurate (PIR), Polyamides, Polyurethanes (PU).*

HIPS/PPE is formulated with the phosphorus FR BDP (between weight-% 8.5-14) (22). PC and polyamides are formulated with AOH. PIR and PU are formulated with TCPP (Note, TCPP is not anymore rated as "PIN" but instead as a "Chlorinated - Phosphorous FR) (26). PC/ABS is formulated with phosphate ester, RDP (weight-% 8.5-14) (22) and AOH

8.2 PIN flame retardants in building and infrastructure

Polymers with PIN FR in the building and infrastructure industry are applied in cable trays, skirting boards, cables, castings, coatings, decorations, flooring, insulation, laminate structures, pultruded profiles, laminates, panels, adhesive layers, tubes/pipes (filament winding), profiles- window, doors, trim. The application matrix for PIN FR in the building and infrastructure industry is shown in Figure 2-3

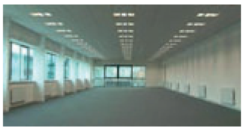
Cable trays and skirting boards *used in the building and infrastructure sector contain plastic such as PP. Similarly to cables in the EEE sector, the most prominent plastics formulated with PIN FRs for cables in the building and infrastructure sector are EVA, EVA/PE, LDPE, polyamides, polyethylene co-butene, polyethylene co-octene, PP, PVC, CPVC, CR, silicon rubbers, TPU, TPE.*

PP used for cable trays and skirting boards is formulated with a various combination of PIN FRs, including phosphorus FR such as ethylene diamine phosphate (EDAP), inorganic FRs such as AOH, ATH, MDH, or nitrogen FRs like melamine phosphate or melamine polyphosphate (8). For the case of cables, polymers such as CPVC and CR are formulated with zinc borate. The sources analysed reported a weight of zinc borate in these polymer types between weight-% 2-5 (42). The polymers EVA, LDPE, Polyethylene co-butene, polyethylene co-octene, TPU and TPE are formulated with numerous inorganic FR, including AOH (weight- %3-15) (42), ATH (up to weight- %80 or weight-% 20-55 combined with P and/or N FR) (42), MDH (up to weight- %80) (42), zinc borate or zinc-hydroxy-stannates (weight-% 3-15). The combination of PE with EVA (PE/EVA) is formulated with inorganic FRs like ATH and MDH (typically weight-% 60–70) (8), and with the combination of inorganic and nitrogen FRs such as melamine triazine with ATH (41). Polyamides are formulated with inorganic FRs like AOH, MDH, and with nitrogen FRs such as MC or melamine phosphate (40). PP is formulated with various types of FRs, including inorganic FRs like AOH, ATH, MDH, nitrogen FRs such as MC, or the combination of inorganic and nitrogen FRs like ammonium polyphosphate with melamine triazine (weight-% 20) (43). PVC is formulated with inorganic FRs such as ATH. Silicon rubbers are formulated with inorganic FRs like AOH, ATH and MDH.

*For **castings, coatings or elements for the façade decoration** used in the building and infrastructure industry, the most important plastics formulated with PIN FRs are PU and PE/EVA (8) (40).*

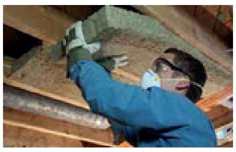
PU used for castings and coatings is formulated with inorganic FRs including AOH, ATH and APP, whereas the combination of PE with EVA (PE/EVA) for I decoration is formulated with inorganic FRs like ATH and MDH.

*The most important plastics formulated with PIN FRs used for **flooring in building and infrastructure** are elastomers, epoxy resin, phenolic resin (PF), polyolefins, PVC, Unsaturated Polyesters (UP), Urethane Elastomers and VE (8) (40)*



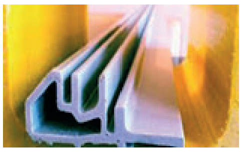
Elastomers are formulated with inorganic FRs such as ATH and MDH. Epoxy resin is formulated with phosphorus FRs (including DOPO, EDAP, TCP), inorganic FRs (ATH, APP), or nitrogen FR (MPP, melamine borate). Phenolic resin (PF) is formulated with phosphorus FRs such as phosphate esters and nitrogen FR like melamine borate. Polyofelins are formulated with ATH or MDH. PVC is formulated with a wide number of FRs, including phosphorus FRs like CDP, DPO, isopropylated phosphate Ester, TOP, TCP, and inorganic FRs such as ATH, zinc borate or zinc hydroxy stannates. UP is formulated with ATH, or nitrogen FRs such as MC or melamine phosphate. Urethane elastomers are formulated with phosphorus FRs (aluminium phosphinate, CDP, TPP), inorganic FRs like ATH and APP, or nitrogen FRs (MC, melamine phosphate, melamine polyphosphate). VE is filled with ATH.

*For **insulation in building and infrastructure**, the most important plastics filled with PIN FRs are ABS, PC, PF, Polyurethane foam, PIR, PU, r-PUR, PVC (8) (40)*



ABS and PC are formulated with phosphate esters and AOH. PF and polyurethane foam are filled with phosphate esters. Moreover, the use of aluminium diethylphosphinate (weight-% 10) with melamine (weight-% 3) has been reported as a PIN FR filler for PF (44). PIR is formulated with phosphorus FRs (DMPP, RP, TEP, TCPP, and inorganic FR (APP). PU is formulated with TCPP (Note, TCPP is not anymore rated as "PIN" but instead as a "Chlorinated - Phosphorous FR), whereas R-PUR is formulated with DMPP, phosphorus polyol, TEP, or inorganic FRs such as APP. PVC is formulated with various inorganic FRs, including AOH, ATH, MDH, or zinc borate.

The most important plastics formulated with PIN FRs used for ***laminate structures, pultruded profiles, panels, adhesive layers****, tubes/pipes (filament winding), and profiles for windows, doors and trim in the building and infrastructure sector are UP, VE, epoxy resin, HDPE, PP and r-PVC (8).*



Unsaturated polyester resins (UP) and vinyl ester (VE) for laminate structures, pultruded profiles are formulated with DMPP, and inorganic FRs such as ATH and APP. Epoxy resin for panels, adhesive layers, tubes/pipes (filament winding), is formulated with phosphorus FRs (DOPO), inorganic FRs (AOH, ATH, APP), and nitrogen FRs (melamine phosphate, melamine polyphosphate). HDPE and PP for pipes are formulated with AOH, ATH and MDH. R-PVC for profiles for window, doors, trim is formulated with AOH, ATH, MDH, zinc borate and zinc hydroxy stannates.

8.3 PIN flame retardants in transport

The use of polymers with PIN FR in the transport industry include cables, ceilings sidewalls, panels, structural parts, coatings, flooring, insulation, interior parts, sealants, or seats. The application matrix for PIN FR in the transport industry is shown in Figure 2-4.

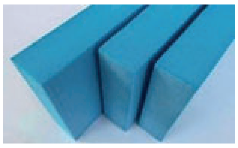
For ***cables in the transport sector****, the most prominent plastics formulated with PIN FRs are EVA, LDPE, polyamides, PVC, Silicon rubbers, TPE/TPU (9) (40)*

For the case of cables, polymers such as EVA and LDPE are formulated with inorganic FRs (AOH, ATH, MDH). Polyamides are formulated with nitrogen FRs such as MC. PVC is formulated with inorganic FRs such as ATH. Silicon rubbers are formulated with inorganic FRs like AOH, ATH and MDH. TPE/TPU is formulated with AOH.

The most important plastics formulated with PIN FRs used ***in ceilings, sidewalks, panels, structural parts and coatings in the transport sector*** *are acrylate resins, epoxy resin, PF, UP resin, VE resin, 2K-PU, acrylates and epoxy resin (9) (40)*

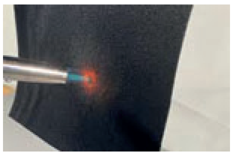
Acrylate and epoxy resins are formulated with ATH and APP. Moreover, epoxy resin is also formulated with cyclic phosphonate, DMPP and DOPO. PF is formulated with phosphate esters and melamine borate. UP resin is formulated with DMPP, EDAP, ATH and APP. VE resin is formulated with ATH and APP. Plastics used for coatings in the transport sector such as 2K-PU, acrylates and epoxy resin are formulated with phosphate esters, ATH and APP.

*For flooring and **insulation elements in the transport sector**, the most important plastics are elastomers, epoxy resin, PF resin, polyolefins, PVC, urethane elastomers, r-PUR and PIR (9) (40)*



Elastomers and polyolefins are formulated with ATH and MDH. Epoxy resin is formulated with ATH, APP and melamine borate. PF resin is formulated with phosphate esters and melamine borate. PVC is formulated with CDP, isopropylated phosphate ester, ATH and zinc borate. Urethane elastomers are formulated withvaluminium phospinate, CDP, TPP, APP, MC, melamine phosphate and melamine polyphosphate. R-PUR and PIR are used for insulation elements in transport. R-PUR is formulated with DMPP, phosphorus polyol, TEP, and APP, while PIR is formulated with DMPP, RP, TEP, APP.

*The most important plastics formulated with PIN FRs used in **interior parts in the transport sector** are ABS, epoxy, PA6, PC, PC/ABS, PE, PET, PUR, recycled PA6/PP, TPU, UP (9) (40)*



ABS is formulated with BDP, RDP and TPP. Epoxy is formulated with ATH. PA6 is formulated with MC, MC combined with MPP (weight-% 12.5 respectively) and melamine polyphosphate (weight-% 25) (45). PC is formulated with phosphorus FRs such as sulphonates or inorganic FRs like AOH. PC/ABS is formulated with BDP, RDP, and TPP. PE and PU are formulated with ATH (weight-% 5-30). Various PIN FRs have been tested with TPU, including phosphorus FRs (TPP, metal phosphinate), inorganic FRs (ATH, AOH, APP, zinc borate), and nitrogen FRs (MC). Unsaturated polyester resin – (UPR) have been formulated with a wide variety of PIN FR (DMPP, EDAP, ATH, APP, melamine phosphate, melamine polyphosphate).

*The most important plastics formulated with PIN FRs used as **sealants in the transport sector** are acrylics, elastomers, epoxy resin, PU and PVC (9) (40)*

These plastic types used as sealants are formulated with a wide range of PIN FRs including phosphorus FRs (EDAP, phosphate esters, RDP), inorganic FRs (ATH, APP, zinc borate) and nitrogen FRs (melamine borate, melamine phosphate, melamine polyphosphate, melamine pyrophosphate).

*Finally, the most important plastics formulated with PIN FRs for **seat elements** in the transport sector are PA6, PU and TPU.*

PU and TPU are formulated with the phosphorus FR TCPP, while PA6 is formulated with nitrogen FRs such as MC (weight-% 25) (Note, TCPP is not

anymore rated as "PIN" but instead as a "Chlorinated - Phosphorous FR), MC combined with MPP (weight-% 12.5 respectively) and melamine polyphosphate (weight-% 25) (45). Moreover, recycled PA6/PP has been used as sealants and interior parts in the transport sector with some PIN FR like APP (weight-% 25), MC combined with MPP (weight-% 12.5 respectively) and melamine polyphosphate (weight-% 25) (45).

9 ANNEX-2: DATA -DRIVEN APPROACH AND INTELLIGENCE DETAILS

9.1 General Approach

A four-step data-driven approach (illustrated in Figure 9-1 and Figure 9-2) was defined, with the purpose of identifying current and future recycling technologies, with the additional scope to support the construction of both a PIN FR Application Matrix (Chapter 2) and a PIN FR Recycling Matrix (Chapter 4), and to identify key technology showcases (Chapter 5) and players. The examined corpus of information for the impact assessment consisted of a mixed scoreboard of 184 R&D papers/reports, 172 funded projects and 735 patents, further scrutinized in a funnel, to shortlist the most relevant ones.

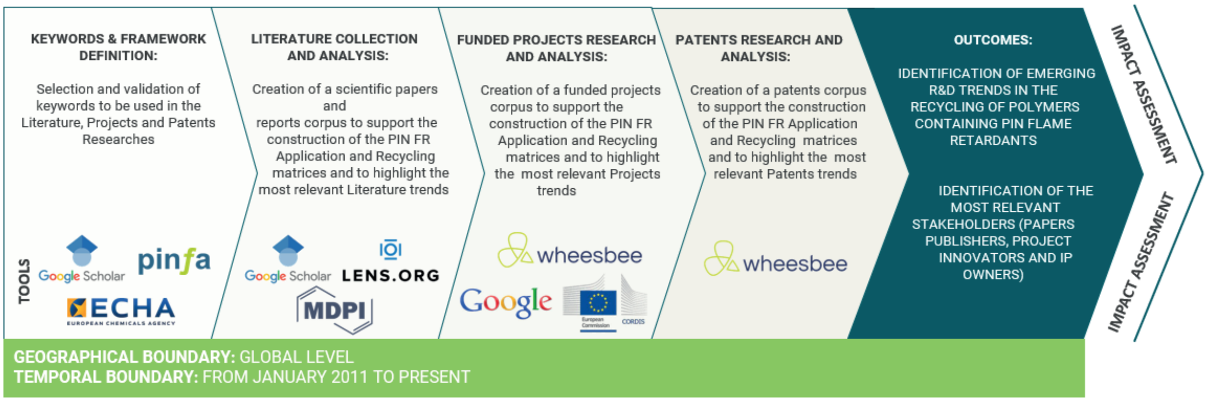
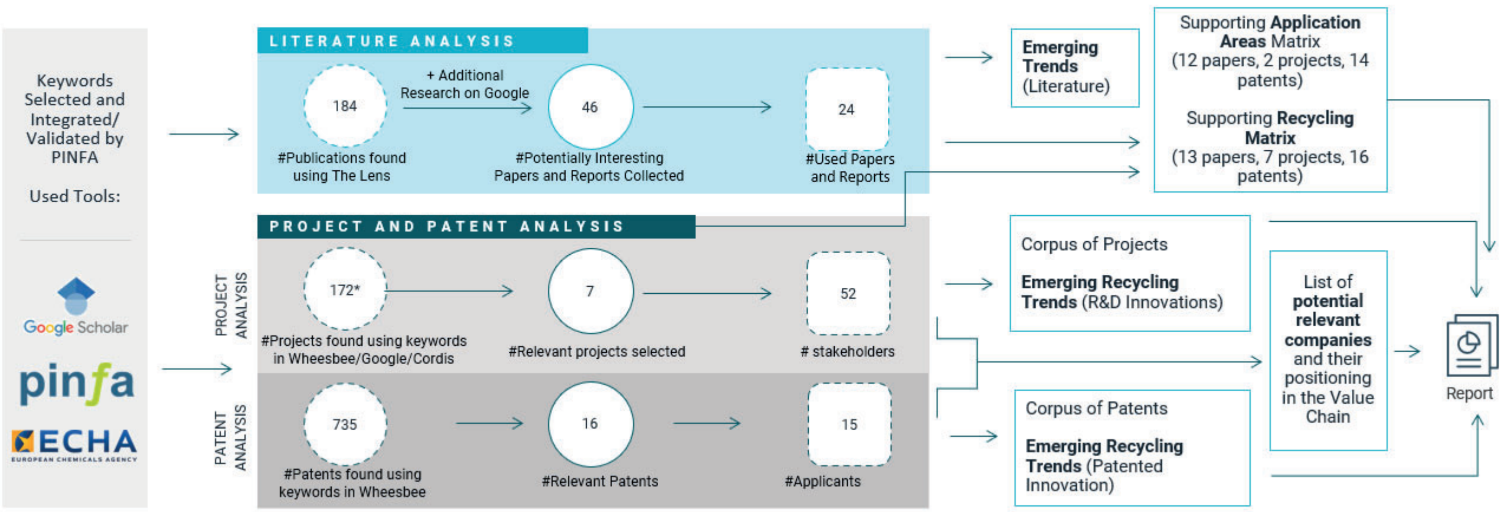


Figure 9-1: Impact Study Methodology Overview-A four Step data-driven Approach

In our data-driven approach, multiple public and private data sources were used. For instance, during the scientific literature analysis Google Scholar, MDPI and The Lens databases were employed.

Geographically, **R&D initiatives from EU and extra EU countries were explored.** The R&D projects that were explored have been financed by the European Union or by other national grantor bodies since 2011 (in the specific UK, Germany, Norway, Finland, Netherlands, Portugal, Spain, Switzerland, France, Italy, Belgium, USA, Australia, Canada). Correspondingly, the largest funding programmes like Horizon 2020 (or FP7 before it) and LIFE (L'INSTRUMENT FINANCIER POUR L'ENVIRONNEMENT) were scrutinised, whilst other national programmes schemes such as InnovateUK, EPSRC, BMVI and BMVi, Business Finland, Research Council of Norway, Open Coesione, FWO, SBIR (US) have been included as well.

On the patents side, this study had **access to approximately 100 million patents from a variety of international databases**, including US, Europe, Japan and China, along other national patent offices from 1985 to date.



* Some doubly counted (emerged from different queries)

Figure 9-2: Methodology Overview, corpus of documents contribution to the report

9.2 Analysed corpus of documents for the creation of the study database

Looking at the analysed corpus of documents, this study has thus considered three categories of stakeholders. The most significant among these players have been identified and classified in the different sectors of the value-chain.

Publishers: the authors of the scientific papers, articles and technical reports in the field of recycling of polymers containing PIN FR.

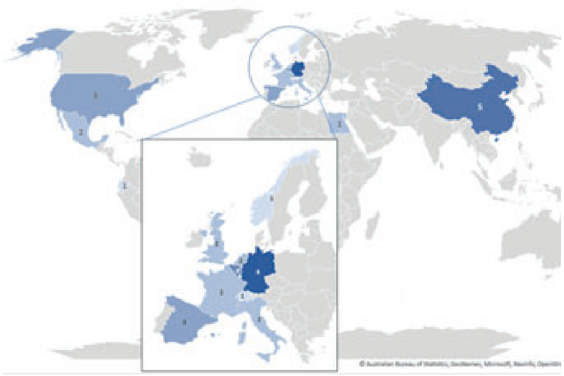


Figure 9-3: Papers publishers by country

Figure 9-4 - Literature Topics Heatmap

184 publications from 2011 to present have been found (Lens, Google Scholar, Wheesbee). The complete set is distributed uniformly in the considered period and is in large part composed of journal articles. 46 of them have been selected as potentially relevant for PIN FR recycling. 24 out of the 46 have been finally retained and defined as most significant for the purpose to support the construction of the PIN FR Application and Recycling Matrices.

More precisely, 12 papers have supported the PIN FR Application Matrix and 13 papers have supported the PIN FR Recycling Matrix (2 publications have been used for both matrices). Furthermore, one paper has been used exclusively to get more insights into the sorting technologies.

Innovators: the participants in relevant funded projects with a focus on PIN FRs and plastic recycling.

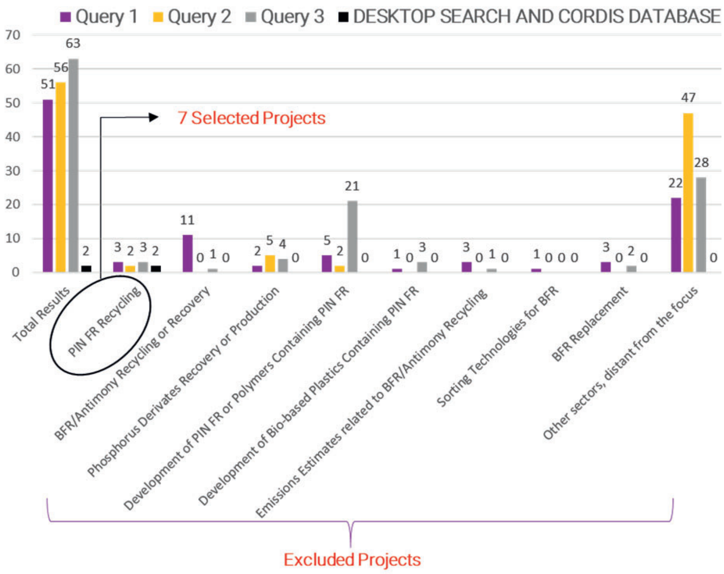


Figure 9-5: Funded projects selection overview

Innovators: the participants in relevant funded projects with a focus on PIN FRs and plastic recycling.

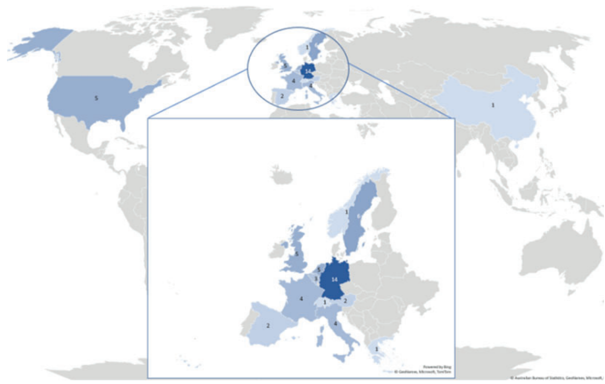


Figure 9-6: Projects participants by countries

Using different queries and different sources (e.g. Wheesbee[32], Cordis) the selection process isolated 7 relevant projects out of an initial corpus of 172 that included different topics, such as, for instance, brominated or antimony recycling, development and production of PIN FRs or polymers containing PIN FRs, BFRs replacement and development of bio-based plastics containing PIN FRs. Only the funded projects focussed on PIN FRs polymers recycling have been retained and analysed in detail. Their topics are reported in Figure 9-5 and a detailed analysis is summarised in Table 9-2

Table 9-2: Selected projects characteristics heatmap

Sector	Waste Type	Recycling Type		
		Mechanical Recycling	Solvent-based Recycling	Chemical Recycling
WEEE	Electric and Electronic Household Appliances	4	0	0
	Cable	2	0	0
Transport	Cable	1	0	0
Building and Infrastructure	All subcategories	0	0	0
No Data		0	1	0
Halogen-Free Flame Retardant Category				
Phosphorus based FR		3	1	0
Nitrogen based FR		1	0	0
Inorganic based FR		1	0	0
Not Specified		1	0	0
Polymer Type				
PC/ABS		3	0	0
ABS		2	0	0
HIPS/PPE		1	0	0
PP		1	0	0
PA6		1	0	0
PA66		1	0	0
PET		1	0	0
PE/EVA		1	0	0
LLDPE		1	0	0
PVC		1	0	0
Not Specified		1	1	0
TRL				
LOW (1-3)		1	1	0
MEDIUM (4-6)		5	0	0
HIGH (7-9)		0	0	0

[32] Wheesbee© is a proprietary software developed by PNO Consultants for innovation intelligence: https://www.wheesbee.eu/

Investors: the patent’s IP owners and inventors, found through the patents analysis. Since patenting can ralate to investments in R&D (it also entails fees for maintaining the patent), all the patent owners have been classified as “potential Investors” in new technologies.

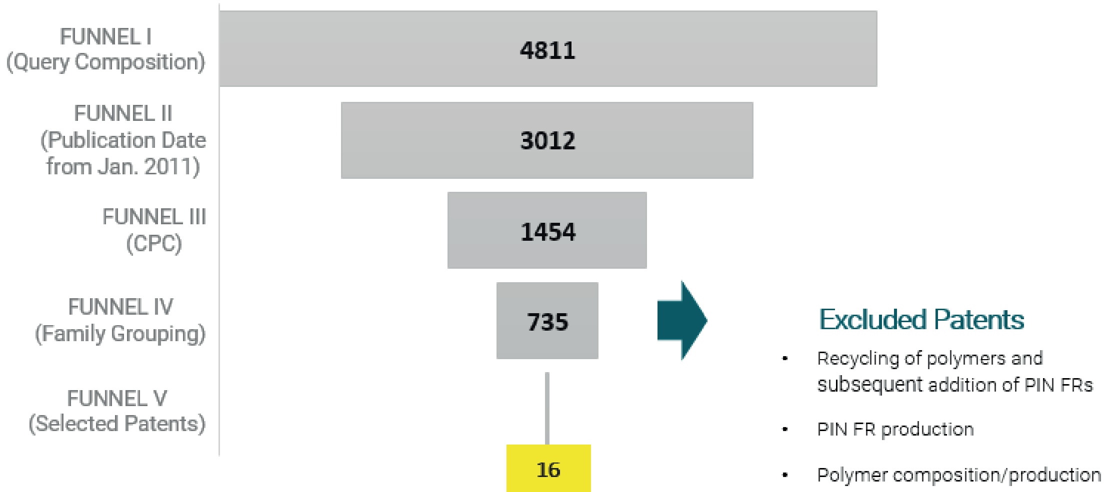


Figure 9-7: Funnels of Patent Documents Research

Using a high-level query, an initial set of 4811 patent documents was found. This was reduced to 735 patents by filtering upon different specific Cooperative Patent Classification numbers (CPC) tags, and then grouping the patents by family.

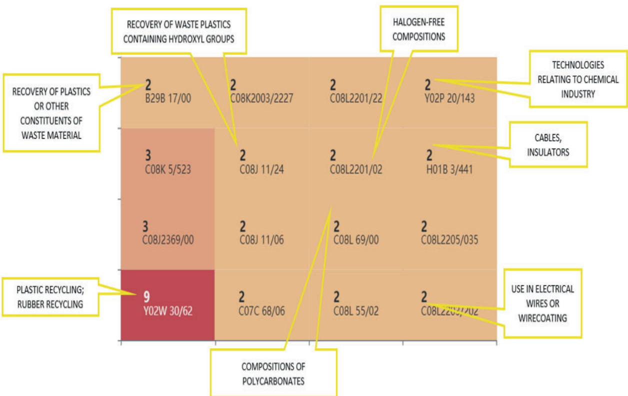


Figure 9-8: Selected Patents CPC

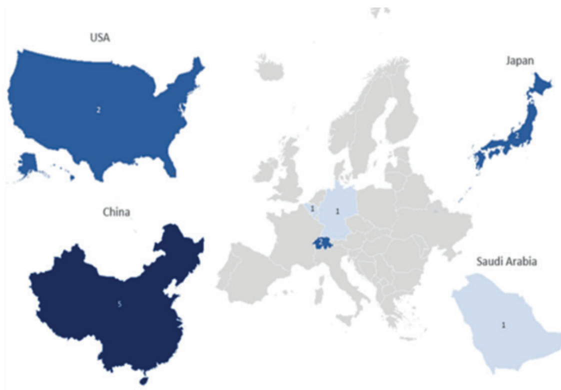


Figure 9-9: 16 top patents applicants by country

Eventually, **16 relevant patents related to PIN FRs polymer recycling have been spotted and analysed**. Many patents were excluded, since they refer to PIN FRs production or to polymer recycling, without considering FRs presence effects on it. More in detail, a large part of the 735 patents original sample aimed to (i) recycling polymers and subsequently add PIN FRs to the recycled polymer, (ii) producing PIN FRs or a polymer including PIN FRs. Differently, **the retained top**

16 patents, consider PIN FRs in the recycling process: their focus is indeed on recycling plastic containing PIN (halogen-free) FRs and their recovery. The **main emerging trends** are summarised in detail in Table 9-3.

Table 9-3: Selected Patents Heatmap

Sector	Waste Type	Recycling Type			
		Mechanical Recycling	Solvent-based Recycling	Chemical Recycling	Waste-to-Energy, Pyrolysis
WEEE	Electric and Electronic Household Appliances	1	0	4	0
	Circuit Board	1	0	1	0
	Cable	3	0	0	0
Transport	Interior parts, carpet	1	1	0	0
	Cable	3	0	0	0
Building and infrastructure	Cable	3	0	0	0
Packaging	Post-consume	3	0	1	1
Halogen-Free Flame Retardant Category					
Phosphorus based		4	0	5	1
Nitrogen based		0	0	0	0
Inorganic based		4	1	1	0
Not Specified		1	0	0	0
Flame Retardant Type					
Triphenyl phosphine		3	0	2	0
Bisphenol A Bis(diphenyl phosphate) (BDP)		2	0	3	0
Trimethyl phosphate (TPP)		1	0	0	0
Triethyl phosphate (TEP)		1	0	0	0
Tributyl Phosphate (TBP)		1	0	0	0
Tricresyl phosphate (TCP)		1	0	0	0
Cresyl diphenyl phosphate (CDP)		1	0	0	0
Resorcinol bis(diphenyl phosphate) (RDP)		1	0	3	0
Aluminium tri-hydroxide (ATH)		3	1	1	0
Boric acid (used as FR)		1	0	0	0
Triphenyl phosphine oxide (TPPO)		1	0	0	0
Magnesium hydroxide / Magnesium dihydroxide (MDH)		2	0	0	0
Red phosphorus (RP)		0	0	0	1
Not Specified		1	0	0	0
Polymer Type					
PC		1	0	2	0
PC/ABS		1	0	2	0
Fiber-reinforced poly(diketoenamine) composite		0	0	1	0
EPS (Expandable polystyrene)		1	0	0	0
PP		2	1	1	0
PET		3	0	0	0
PUR		1	0	0	0
PE			1	0	0

10 ANNEX-3: MOST RELEVANT SELECTED LITERATURE

10.1 Scientific Reports and Papers shortlisted as most pertinent for the R&D trends analysis, the Application Matrices and the Recycling Matrix

Paper Title	Year	Organisations (Publishers)
Chemical Recycling of Mixed Plastics in Electronic Waste Using Solvent-Based Processing Ref. (27)	2022	University of Massachusetts Lowell
Closed loop recycling of plastics containing Flame Retardants Ref. (22)	2013	KU Leuven; ICL-IP Europe; Recycling Consult BV; University of Cuenca
Enhanced PET Processing with Organophosphorus Additive: Flame Retardant Products with Added-Value for Recycling Ref. (46)	2019	EMPA - Swiss Federal Laboratories for Materials Science and Technology
Fast pyrolysis of polyurethanes and polyisocyanurate with and without flame retardant: Compounds of interest for chemical rec ycling Ref. (26)	2021	Ghent University
Intrinsic flame retardant phosphonate-based vitrimers as a recyclable alternative for commodity polymers in composite materials Ref. (47)	2020	Max Planck Institute for Polymer Research; Bundesanstalt für Materialforschung und -prüfung (BAM); Fraunhofer Institute for Manufacturing Technology and Advanced Materials IFAM
Investigating thermomechanical recycling of poly(ethylene terephthalate) containing phosphorus flame retardants Ref. (48)	2021	EMPA - Swiss Federal Laboratories for Materials Science and Technology; Montanuniversitaet Leoben
On the role of flame retardants in mechanical recycling of solid plastic waste Ref. (49)	2018	Ghent University
Product Clustering for Closed Loop Recycling of Flame Retardant Plastics: A Case Study for Flat Screen TVs Ref. (50)	2013	KU Leuven; University of Cuenca
Recyclable Flame retardant Epoxy Composites Based on Disulfide Bonds: Flammability and Recyclability	2021	IMDEA Materials Institute; Universidad Politecnica de Madrid; Central South University; CIDETEC
Recycling Halogen-Free Flame Retardant Plastics. Example: Polyamide Ref. (51)	2018	Kunststoffe International, Fraunhofer LBF
Recycling of flame retardant plastics from WEEE, technical and environmental challenges Ref. (52)	2013	ICL-IP Europe; Recycling Consult BV; University of Cuenca; KU Leuven
Recycling of phosphorus-containing plastic based on the dual effects of switchable hydrophilicity solvents Ref. (53)	2020	Southwest Petroleum University
Pyrolysis characteristics and pyrolysis products separation for recycling organic materials from waste liquid crystal display panels Ref. (37)	2015	Shanghai Jiao Tong University
Halogen Free Flame Retardant 2021 Ref. (40)	2021	Graft Polymer
Innovative and Sustainable Flame Retardants in Transportation Ref. (9)	2021	PINFA
Innovative and Sustainable Flame Retardants in Building and Construction Ref. (8)	2016	PINFA
Improving mechanical properties and reaction to fire of EVA/LLDPE blends for cable applications with melamine triazine and bentonite cl ay Ref. (41)	2020	CIATEC; Universidad Nacional Autónoma de México ; Università degli Studi di Milano; Politecnico di Torino
Flame retardant PP/PA6 blends: A recipe for recycled wastes Ref. (45)	2019	Université de Lorraine; Ecole des Mines d'Alès
Non-halogenated Flame Retardant Handbook, 2nd Edition Ref. (42)	2021	University of Dayton
Innovative Flame Retardants in Electronics & Electrical Applications Ref. (5)	2017	PINFA
Towards a more circular economy for WEEE plastics – Part B: Assessment of the technical feasibility of recycling strategies Ref. (33)	2019	KU Leuven; Campine NV
Towards Selection Charts for Epoxy Resin, Unsaturated Polyester Resin and Their Fibre -Fabric Composites with Flame Retardants Ref. (13)	2021	Ain Shams University; Norwegian University of Science and Technology; Galala University
Synergistic effects of aluminum diethylphosphinate and melamine on improving the flame retardancy of phenolic resin	2020	Nanjing Tech University; China University of Geosciences
Synergistic of ammonium polyphosphate and alumina trihydrate as fire retardants for natural fiber reinforced epoxy composite Ref. (44)	2017	The University of Nottingham; University of New Orleans

10.2 R&D funded projects shortlisted as most pertinent for the R&D trends analysis, the Application Matrices and the Recycling Matrix

Project	Period	Funding Country / Programme	Description	Coordinator	Other Participants
REWARD-WEEE	2009 2012	EUROPE Eco Innovation Funding Programme	To demonstrate in a prototype facility the generation of **recyclable products from WEEE**., demonstrating advanced **separation and sorting techniques by mechanical recycling** as verified by earlier laboratory analyses (Polymer sensor sorting of brominated and **phosphorus flame retardant containing polymers**)	RECYCLING CONSULT BV	Dolphin Metal Separation; Coolrec Bv(Phb); Brgm
Recycling of halogen-free flame retardant plastics	2015 2018	GERMANY German Federation of Industrial Research Associations (AiF)	To demonstrate the **recyclability of several thermoplastics formulations using PIN flame retardants**. Testing polypropylene**, PC/ABS, polyamides (PA-6 and PA-6.6**), and **polyethylenes**, using phosphorus, nitrogen and mineral based PIN flame retardants provided **by PINFA member companies**	FRAUNHOFER LBF	Adeka Europe Gmbh; Basf; Budenheim; Clariant; Nabaltec; A. Schulman; Byk Chemie; Ctf 2000; Dartex; Dsm; Dupont; Everkem; Frx Polymers; Italmatch Chemicals; Lanxess; Metadynea; Perstorp; Presafer; Schill & Seilacher; Solvay; Thor; William Blythe; Inemi
NANOFRABS	2012 2014	EUROPE FP7 Programme	The project aims at developing new **halogen free flame retardant (HFFR) for ABS (acrylonitrile-butadiene-styrene) compounds**. The **recyclability of the final HFFR-ABS compound** has been studied by its re-processing (**extrusion-injection**) in four different cycles.	TECNALIA	Masterbatch Srl; Sitraplas Gmbh; Prolabin & Tefarm Srl; Dasyc Sa; Politecnico Di Torino
Recycling of electrical cables with focus on mechanical recycling of polymers in end-of-life cable (38)	2014	SWEDEN Funded by Vinnova	Development of know-how and technology for **mechanical recycling of electrical cables** with focus on recycling the various polymers in the cable waste; The **recycling** of **PVC containing PIN FR** has been tested but other tests must still be carried out.	RISE	Nexans; Ab Volvo; Hellermann Tyton; Riflex Film; Norner; Axjo Plastic; Stena Recycling; Hamos Gmbh Recycling Und Separationstechnik, Gneuss Gmbh
An innovative catalytic system for the exchange reaction of phosphate esters with N- nucleophiles towards the production of sustainable and recyclable phosphorus flame-retardant additives	2020 2023	BELGIUM FRIS (Flanders Research Information Space)	Development of new and **recyclable phosphorus-based flame-retardant** additives, in view of the need to replace brominated compounds (**phosphate esters with N-nucleophiles** (like anilines)). The fate of different flame retardants will be evaluated in **solvent-based recycling** and reprocessing operations	KU LEUVEN	\
CloseWEEE	2014 2018	EUROPE H2020 Programme	Innovative solutions for **closing the loop of post-consumer high-grade plastics from WEEE**, for new EEE through advanced recovery of valuable plastic streams which do not have a **recycling system** yet, and subsequent **replacement of halogenated flame retardants by halogen-free flame retardants in new EEE**	FRAUNHOFER	Vertech Group; Coolrec Bv; Fundacion Gaiker; Argus Additive Plastics Gmbh; Tecnalia; Exergy Ltd; Ifixit Gmbh; Accurec-Recycling Gmbh; Die Wiener Volkshochschulen Gmbh; Sitraplas Gmbh; Philips Consumer Lifestyle Bv
Closing material loops for plastics from Waste Electrical and Electronic Equipment	2016 2020	BELGIUM FRIS (Flanders Research Information Space)	Development of **new mechanical recycling** strategies to re-apply post-consumer plastics in high-end thin-walled **injection moulded products**. To achieve this objective, separation based on dismantling and subsequent spectroscopic sorting of large plastic components is investigated. Results demonstrate that this strategy **allows to produce high-quality recycled plastics with flame retardants**, as well as other special engineering plastics.	LEUVEN	\

10.3 Patents shortlisted as most pertinent for the R&D trends analysis, the Application Matrices and the Recycling Matrix

Patent ID and Title	Applicant	Publication Date	Brief Description
Method for recovering waste circuit board flame retardant **CN111704632 (21)**	ANHUI CHAOYUE ENVIRONMENTAL PROTECTION TECHNOLOGY COMPANY UNIVERSITY OF SCIENCE AND TECHNOLOGY OF CHINA	25/09/2020	The invention discloses a waste circuit board flame retardant recovery method which comprises the following steps: (1) coarse crushing; (2) fine crushing; (3) electrostatic separation; and (4) supercritical extraction of CO2. According to the method, CO2 is used as an extraction agent to recover and analyse the flame retardant in the supercritical fluid, the flame retardant is composed of triphenyl phosphate, the occupation amount of triphenyl phosphate is about 90% or above, and the purity of the recycled flame retardant is achieved.
Manufacture of dihydroxy aromatic compounds by alcoholysis of flame retardant-containing polycarbonate compositions **EP2746249**	SAUDI BASIC INDUSTRIES CORPORATION	25/06/2014	Heating a polycarbonate-containing composition (comprising PC and PFR) in the presence of an alcohol and a catalyst at a temperature of 70°C to 200°C and a pressure of 50 mbar to 40 bar for a time sufficient to depolymerize the polycarbonate to provide a dihydroxy aromatic compound and a dialkyl carbonate. Given the presence of PFR containing BPA, the use of a transesterification catalyst having non-neutralizable groups is recommended (the basic catalysts are unable to depolymerize the PC containing this PFR and therefore do not allow recovery of the BPA). Dissolution and separation of the compound and recovery of the bisphenol A
Method for alcoholysis of polycarbonate compositions containing flame retardant or acrylonitrile-butadiene-styrene **US8846858**	SAUDI BASIC INDUSTRIES CORPORATION	30/09/2014	Methods for the alcoholises of polycarbonate compositions containing flame retardants or acrylonitrile-butadiene-styrene, and in particular to methods of making bisphenol A by methanolysis of a bisphenol A polycarbonate composition containing phosphorus-containing flame retardants or acrylonitrile-butadiene-styrene. Dissolution of flame retardant through solvents, formation of a filterable suspension of FR and ABS and/or PC and use of a second solvent for new dissolution of the FR in one of the two polymers. Finally, PC and/or ABS are depolymerized by heating with alcohol (methanol) and a transesterification catalyst for recovering the bisphenol A and the dialkyl carbonate.
Method for direct ammonolysis of polycarbonate-containing materials and products **US9328046**	SAUDI BASIC INDUSTRIES CORPORATION	03/05/2016	Non-catalytic method for carrying out ammonolysis of polycarbonate-containing plastic that also contains acrylonitrile-butadiene-styrene (ABS), and/or a phosphorus-containing flame retardant, such as bisphenol A bis(diphenyl phosphate) (BPADP). A process is provided that surprisingly allows the separation and recovery of bisphenol A and urea from recycled plastics of low polycarbonate content through selective depolymerization. The ammonia solution is of sufficient strength to selectively sever the ester bond of the polycarbonate and to form the ammonium salt of bisphenol-A (ammonium phenolate) which is soluble in the aqueous phase. A two phase system is formed, namely a solid phase and a liquid phase of which the liquid phase contains both urea and the ammonium salt of bisphenol-A. Separation of the phases is achieved by filtration.
Recyclable and reconfigurable high-performance polymer networks and uses thereof **Wo2019099944**	UNIVERSITY OF CALIFORNIA	23/05/2019	Depolymerisation by hydrolysis of the fiber-reinforced poly (diketoenamine) composite containing TPP (25%), in the presence of an acid or a mixture of acids selected from, but not limited to, HC1, H2SO4, H3PO4, -toluenesul lonic acid, methane sulfonic acid, trifluoroacetic acid, trifluoromethanesulfonic acid. Poly(diketoenamine) waste can be selectively chemically recycled in the presence of common plastics, including but not limited to: 111 (poly(ethylene terepthalate) (PET), polyamide nylon-6,6 (PA), polyethylene (PE), polypropylene (PP), poly(vinyl chloride) (PVC), polycarbonate (PC), obviating laborious and expensive material sorting. After this process it is possible to recover pure triketone monomer, pure TPP FR and pure reinforced fiber.
	SULZER CHEMTECH AG	16/09/2015	The present invention relates to a process to recycle and/or formulate expandable plastic materials.
Self-reinforced composite made of recycled materials and process of making the same **US7887726**	NOVANA, INC.	15/02/2011	Since most plastic waste from carpet and car parts contains immiscible polymers (IP) that are difficult to identify and separate, this invention eliminates the initial steps of identifying and separating these thermoplastics and uses two sequential processes to recycle these polymers: extrusion, 2. Melt-filtration. Through melt-filtration, flame retardant (such as alumina hydroxide) and other additives can be separated and completely removed from the polymers. FR is removed from the polymers after melt-filtration process. The recycled polymers have nice mechanical properties
Flame retardant thermoplastic elastomer (TPE) made from waste crosslinked polyethylene cable materials and preparation method thereof **CN102898768**	JIANGSU XINGHAI WIRE AND CABLE CO., LTD. (Jiangsu keriman Cable Technology Co., Ltd)	30/01/2013	The present invention relates to a cable material with waste XLPE production of a flame retardant thermoplastic elastomer and a preparation method thereof. Phosphate-based FR is added during the recycling process, before the extrusion step.

Patent ID and Title	Applicant	Publication Date	Brief Description
Recycled PC (Polycarbonate) and recycled PET (polyethylene terephthalate)-containing halogen-free flame retardant resin composition and preparation method thereof **CN102604352**	SHANGHAI KUMHO SUNNY PLASTICS CO., LTD.	25/07/2012	The invention relates to a recycled PC (polycarbonate) and recycled PET (polyethylene terephthalate)-containing halogen-free flame retardant resin composition and a preparation method thereof. TPP and BDP are added during the recycling process before the extrusion step.
Method of recycling a plastic **JP5006523**	PANASONIC	22/08/2012	To provide a method for recycling plastic by which plastic waste is recycled and boehmite can be produced at a low price. Plastic containing aluminium hydroxide as an inorganic filler is comminuted, heat-treated at 200-450°C in the air and recovered as boehmite.
Recyclable environment-friendly non-toxic sheath material for electronic product cable **CN108342011**	HEFEI LIXIAN ELECTRIC POWER ENGINEERING CO., LTD. (Hefei Li Xing Power Engineering Co Ltd)	31/07/2018	The present invention relates to the technical field of the cable jacket, in particular to a non-toxic recyclable and environment-friendly electronic product and wire cable jacket material. Aluminium and magnesium hydroxide are added during the recycling process before the extrusion step.
Recycling method for styrene-containing plastic waste **Wo2018224482**	INEOS STYROLUTION GROUP GMBH	13/12/2018	Decomposition of the styrene-containing plastic waste (with red phosphorus FR) is carried out in a suitable pyrolysis reactor (twin-screw extruder, fluidized bed reactors and microwave reactors) thermal energy (by heating or microwave irradiation) and optionally by the introduction of shear energy. Decomposition occurs at a preferable temperature between 250 °C and 500 °C. After the process, the separated styrene monomers are extracted and are available for the re-polymerization. Styrene oligomers can be split in a steam cracker, in presence of water vapor, to recover starting materials for plastics, such as ethene, propene or benzene.
Method of recycling waste of thermoplastic resin composition and recycled material **Jp2014214195**	SHARP CORP	17/11/2014	To provide a method of recycling waste of a thermoplastic resin composition (PC and ABS) containing a phosphorus-based fire retardant to obtain a recycled material having high fire retardancy. Phosphate-based FR is added during recycling before the extrusion step.
Oil-resistant, low-smoke, halogen-free and flame-retardant polyolefin cable material, and preparation method therefor **Wo2017177482**	YANGZHOU HAONIANHUA POLYMER MATERIALS CO. LTD.	19/10/2017	Disclosed are an oil-resistant, low-smoke, halogen-free and flame-retardant polyolefin cable material, and a preparation method therefor. This oil-resistant low smoke zero halogen flame-retardant polyolefin method for producing a cable material, the specific process includes sorting the stripped, crushing, cleaning, drying, internal mixer, extruder, pelletizing and packaging steps.
Flame-resistant coating for the rear side of a carpet **Ep2885362b1**	CLARIANT	26/10/2016	The invention relates to a flame-retardant preparation for bonding or fixing of textile sheet materials (e.g., floor covering constructions, Woven, Nonwoven, Tuftwaren, in particular carpets) and textile connected (textile laminations) equipped with this preparation and the products. 2 kg waste carpet were washed with 10 kg of a suitable solvent (xylene) which is added and heated to 80°C. The dissolution temperature of the Licocene PP 2602 is at 73°C. The flame-retardant equipment could be removed by Filtration. The textile fibers were not attacked by the solvent and remained completely obtained. The selectively dissolving time was below 20 min. The polyolefin wax was precipitated by lowering the temperature, squeezed out and dried in vacuo at 40°C. The thus recovered solvent was returned to the processing. After recycling process, the FR is removed by Filtration.

11 ANNEX-4: RECYCLING MATRIX RATIONALE

11.1 Recycling matrix design process

Starting from the analysis of the existing knowledge related to plastics recycling and including polymers, polymers mixtures and resins, an initial description of the substances and recycling process involved has been collected. The entry of data within the recycling matrix has been performed in parallel with the analysis of the defined innovation scoreboard in Chapter 3 (and ANNEX-3). This allowed to: (1) populate the matrix with new contents, (2) define more keyword to continuously refine the research activity, (3), (4) review the database structure and contents, (5) further refine the matrix quality by iterating this process.

To support the design of a scalable rationale, all the definitions within the matrix have been agreed in a series of dedicated sessions supported by the PINFA industry experts. Terms, definitions, and nomenclature have been proposed and discussed to avoid ambiguity or overlaps, thus preventing the redundancies which could compromise the overall informativeness of the map.

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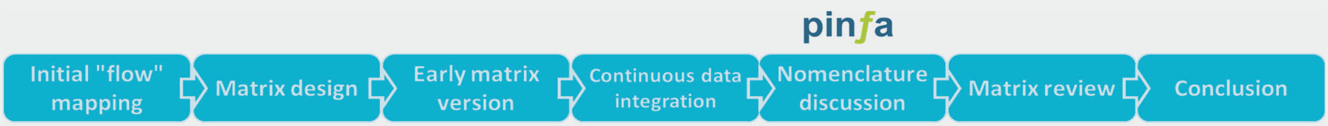

Figure 11-1: PIN FR recycling matrix development workflow

11.2 Data Sources

The data, which actually populate the recycling matrix, are sourced from ANNEX 3's documents corpus:

- **[L]** Technical or scientific literature
- **[J]** Funded Project
- **[P]** Patent literature

This is aligned with the stakeholders analysis process and related sections in Chapter 3.

When available, the stage of development of the analysed technology follows a standard TRL definition:

-
- **[L]** Lab-scale (TRL<6)
- **[P]** Piloting (TRL6-7)
- **[I]** Actual industrial scale application (TRL7-9)

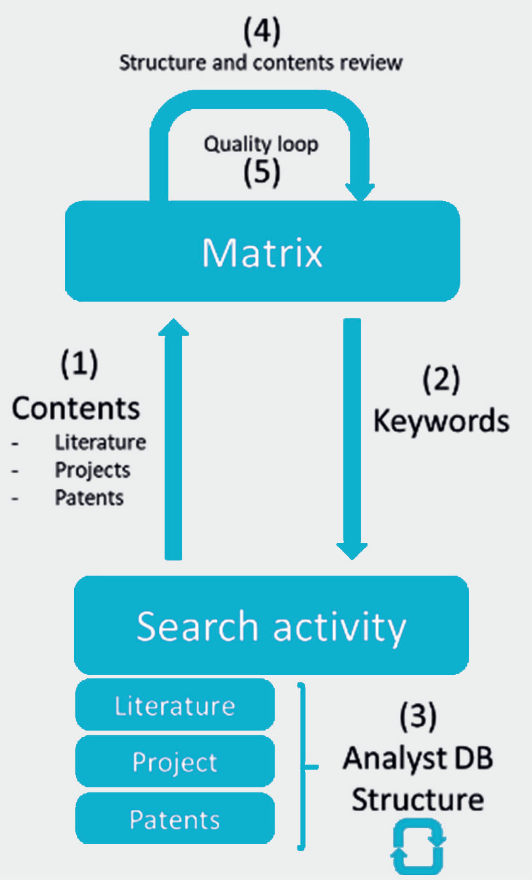

Figure 11-2: Iterative matrix population

11.3 TAXONOMY

11.3.1 Waste polymer sources

This category describes the possible sources of plastics waste considered in the recycling map.

We consider polymers, in pure composition, or as a mixture, or as resin(s) which may often contain flame retardants. The present types of **plastic sources** have been adopted:

- **Post-consume** – *Packaging*, waste from mass market application or house-holds
- **Post-consume** – *Yarns*, waste from mass market application or households
- **Transportation** – All applications, part, or components from any vehicle
- **WEEEs** – *All applications*
- **Building and infrastructure**, part, or components of buildings, including conduits / corrugates
- **Secondary raw materials** / Post-industrial residues, industrial residues, post-industrial (i.e., from cutting, grinding, operations)

- **Other / No-data / Other post consume**

The description level is quite high, allowing for more detailed subcategories to be filtered:

- **Plastic waste sources:** *ESR fraction containing TV sets, PC Boards, wires; LCD TVs post-consume; post-consume (yarns); building and infrastructure (flooring); vinylogous polyurethane (vitrimer); yarns, packaging; WEEES (general); building and infrastructure (other); post consume waste; automotive; flat screen TVs; WEEEs, automotive; flame retardant resins; LCD panel; non specified, NS; waste circuit boards; carpet, car interior parts; waste liners; adhesive labels; cables; packaging: OA equipment and electric and electronic components.*
- **Polymers/polymers mixtures/resins**: *PS, ABS, PC/ABS blend, HIPS-PPE, PET, PIR – polyisocyanurate, vinylogous polyurethane, PP/PA blend, PP-flax composite, PC, PP/EPR blend, PE/Tire rubber, PP/Wood floor, HIPS/PPE, EP, glass-fiber reinforced polyamide (PA), HIPS/PPE blend, CTA/PVA/TPP film, NS, PC/ABS, PP, PA6, PA66, PE/EVA, LLDPE, PVC, poly(diketoenamine) composite, EPS, PUR, PE, XPLE, polyamide, PMMA, epoxy resin, EVA*.
- **Compositions:** *When retrieving this information was possible, the amount of PIN FR contained in a plastic substrate has been expressed as %. However, this information is missing in most of the analysed cases.*

11.3.2 Flame retardants (FRs)

The described flame retardants are categorised accordingly to the main categories in the scope of our study.

- **[P]** Phosphorous flame retardant
- **[I]** Inorganic flame retardant
- **[N]** Nitrogen flame retardant
- **[X]** Non classified / data not available

Each chemical mentioned in the matrix is also associated, when possible, with the related CAS number reference.

The CAS number - Chemical Abstracts Service, allows to exactly identify any existing chemical substance and without occurring in any ambiguity. Ambiguities in chemical nomenclature are caused due to the often-adopted traditional naming, or outdated chemical nomenclature, which is still used especially in commercial applications.

CAS can be eventually linked with the EC Number (54) which is the ECHA

standard identifier for chemical substances and allows to retrieve useful information in regards with safety or regulations (i.e., REACH).
The correct reference with unique identifiers of chemical substances (i.e., CAS number) can amplify the overall informativeness of this tool, by linking further valuable technical information such as chemical/physical properties and toxicity or environmental data from dedicated international repositories.

Details of flame retardants are included in the matrix and can be filtered.

- **Flame retardant (alone or in mixtures)**: *triphenyl phospate (PFR) / (PPT); phosphorous - PFR - not further specified; DOPO-PEPA (DP); tris(2-chloropropyl) phosphate (TCPP) - (Note, TCPP is not anymore rated as "PIN" but instead as a "Chlorinated - Phosphorous FR); phosphonates; Aflammit PCO (900) (AF); intumescent ammonium; polyphosphate (APP); magnesium dihydroxide (MDH); potassium diphenylsulfone sulphonate (KSS); red phosphorus/PA6; magnesium oxides; organic phosphinate (Exolit / Red phosphorus), bisphenol A bis-(diphenylphosphate) – BDP; antimony trioxide; APP; DPER; melamine; DOPO; aluminium diethylphosphinate (AlPi); alumina trihydrate (ATH); piperazine pyrophosphate; phosphate ester; alkoxyamine; melamine cyanurate; DEPAL + P-synergist; bisphenol A bis(diphenyl phosphate); resorcinol bis(diphenyl phosphate); resorcinol tetraphenyl diphosphate (RDP); boric acid; triphenyl phosphine oxide; tri (3-hydroxypropyl) phosphine oxide; phosphonic acid ester; tricresyl phosphate ester, triethyl phosphate, triphenyl phosphate, dimethyl phenyl phosphate, tributyl phosphate or phosphoric; acid dimethyl phenyl diphenyl ester; magnesium hydroxide (MDH) diethyl phosphinic acid aluminate; NS - Other HFFR.*

11.3.3 Taxonomy: recycling processes

The present category includes all possible processes, existing at the state of the art, which concerns the recycling, thermal valorisation, or disposal, of the recycled plastic and resins materials. A sustainability indication is also reported, according to the EU pyramid approach.

- **Re-use** recovery of part or pieces from a recycled item, no further process phase foreseen
- **Depolymerisation** - chemical recycling, set of chemical recycling process technologies which allow the conversion of a recovered polymer into a monomer, to obtain a virgin-like polymer (i.e., crPET, monomers obtained from lysis recycling processes)
- **Pyrolysis** - *chemical recycling* (thermo-cracking, gasification)
- **Mechanical recycling** - *shredding + sorting + moulding (thermal)*, mainstream recycling methodologies, most based on physical processes
- **Solvent-based recycling** - *shredding + solvent*, recycling technology partly

based on mechanical means and then on the polymer dissolution within a chemical solvent– recovered by precipitation / filtering

- **Waste-to-energy**,
    - *Gasification*, recycling processes which allow the final conversion into gas for energy generation purposes (e.g., syngas)
    - *Pyrolysis* thermal decomposition processes
    - Other processes, no data available
- **Landfilling** (option not considered due to the Landfilling directive in EU[33])

The recycling process in the matrix can be further filtered. Example processes are: *thermo-mechanical recycling, including sorting, shredding, moulding and extrusion, etc.*

11.3.4 Taxonomy: End-of-life - Material (polymer, resin, etc.)

The present category concerns the plastics, or more likely polymers, end-of-life (EoL). This allows to classify the outcomes of a recycling process and possibly to understand the impacts caused by the PIN FR presence – which is contained in the polymer substrate, on the overall recycling process.
The EoL aspect is especially relevant considering the possible affection caused by the PIN FR traces on the recycling process itself or in terms of recycled materials final quality, which can impact on its end use and the related market demand.

The recycling process outcome, in terms of final quality of the recycled materials, is although not negligible considering the need of ensuring an economically sustainable recycling process. This must be indeed justified by an effective use of it and a consistent market demand for the recycled plastic materials which is linked with a profitable IRRs – Internal Return Rate, for any recycling process investment initiative.

Understanding the quality of a recycling process output is therefore key to understand the economics of a recycling process and therefore to sustain its wider adoption, or scale-up, by levering on future investments by both private and publics stakeholders (55).

- **Full recyclability through value chain** – effective circularity of the polymers / recycled materials, quality and functionalities are preserved even after multiple recycling iterations
- **Downgraded** polymer is recycled but deemed to lower value application / FR applications of recycled materials / only partial loss of property and quality

[33] https://ec.europa.eu/environment/topics/waste-and-recycling/landfill-waste_it

in recycled material

- **Partial recyclability** the polymer recyclability is affected / a significant loss of technical properties and quality is noticed / degradation of material due to the FR content itself
- **Feasible waste-to-energy conversion** polymer is converted to energy through pyrolysis or other waste-to-energy means
- **Recyclability compromised** due to FR contamination in polymer matrix, polymer cannot be recycled

11.3.5 End-of-life – Material (PIN – FR)

Commonly, PIN Flame Retardant chemicals are not effectively separated from plastics after the recycling process, however there is empirical evidence which support a growing interest in separating and recycling FRs themselves and especially for the most valuable FRs.

- **[S]** FR is separated from the recycled material / polymer
- **[R]** in case of FR it is separated, the PIN FR is purified and then recycled itself for new FR applications
- **[D]** FR is destroyed, i.e., in thermal processes due to chemical decomposition
- **[U]** Unseparated, the FR is integrated within the recycled polymer chains which can be eventually fully recycled or downgraded to lower value applications (i.e., FR materials).
- **[A]** Added, a virgin flame retardant is introduced as additive in recycled a polymer matrix, post-recycling addition of Flame Retardant to enable the recycling of plastics / polymers
- **[L]** Lost, unwanted loss of FR in environment causing drawbacks such as pollution / contamination / harmful effects on personnel or environment / unknown fate or EoL cannot be understood from the information available,

11.4 POSSIBLE FUTURE DEVELOPMENTS OF THE RECYCLING MATRIX DATABASE

11.4.1 From small-data to big-data and data analysis

The structure of the matrix potentially allows to identify the relationships described above, with the concrete possibility of identifying many more correlation patterns existing among different combinations of waste/polymer/treatment/FRs/EoL, etc.
More data are however needed to provide robust conclusions and to reduce the noise effect in this exercise, preventing any possible bias.

With a larger availability of data, more advanced data analytics methodologies can also be used to identify statistically relevant patterns and map more strictly the correlations which may occur.

11.4.2 Link with external relevant resources

The utilization of CAS numbers, which are assigned to each PIN FR substance, allows to leverage on different scientific and technical resources (public databases). This link will support a better understanding of the environmental and safety profile of each PIN FRs.

The CAS number identifier also permits to exactly identify the chemical substance and the further link with relevant toxicological or environmental information including the link with ECHA-REACH chemical substance database.

Possible links given a CAS identifier:
CAS Registry - https://www.cas.org/cas-data/cas-registry
ECHA-REACH - Registered Substances Factsheets: https://echa.europa.eu-/it/information-on-chemicals
ECHA-SVHC - Substance of very high Concerns: https://echa.europa.eu/it/substances-of-very-high-concern-identification

11.4.3 Data extraction and interoperability/replicability

The logic of the developed matrix can be easily translated from a spreadsheet to a relational-based database tool, i.e., SQLlike database, for online publication purposes or to develop other data-driven applications.

The matrix framework offers the possibility of being replicated into similar maps that can be useful to analyse different plastic additives behaviour within the existing recycling processes. This knowledge would be industrially relevant, especially considering the possible impacts of additives on the plastic circularity as well as on the plastic value-chain itself and to sustain any further effort concerning sustainability by design.

The utilization of more advanced data processing means would also allow to obtain many other different types of information and to enable further data analysis methodologies like clustering to identify similarities.
Finally, the present rationale can support the identification of keywords suitable for being applied in data-driven applications such as scientific literature mining (text mining) at very high throughput.

11.4.4 Infographics and data visualisation (DataVis)

Graphical visualisations (DataVis) can further simplify the process of understanding, especially for divulgative purposes (i.e., by assigning symbols or logos). Visualisation could be also easily translated into online media.

11.4.5 Mapping exercise replicability

The approach used for mapping the applications and treatments of plastics containing PIN FR can be translated toward other families of additives which are commonly used for conferring and modulating the specific properties of plastic and polymer substrates. These substances can be analysed with the present matrix/map approach and rationale.

When the presence of these additives in a polymer is linked with major impacts on the recycling process (safety, quality, environmental sustainability) the organization of the available information within a similar map could become highly relevant for identifying any possible interference with the related recycling process, or even to understand when their separation from a polymer substrate results economically viable at larger scales. Commonly used additives regard plasticiser, antioxidants, pigments, heat stabilisers, etc. are indeed used in a range of 100T/year[34][35], and concern almost all existing plastic materials.

Considering the pervasive use of additives in plastic materials it may be highly relevant to address more recycling cases by applying and extending the proposed application / recycling matrix approach. Additives of potential interest for future mapping exercises[36] are Light stabiliser, other stabiliser, nucleating agents, antistatic agents, other functions, heat stabiliser, antioxidants, pigment agents, flame retardants (PIN FR already assessed), plasticisers.

[34] https://echa.europa.eu/it/mapping-exercise-plastic-additives-initiative . Last accessed: 13 April 2022
[35] https://cefic.org/media-corner/newsroom/eu-inventory-of-all-plastic-additives-will-further-improve-safety-of-chemicals/
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