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Recycling of Waste Fiber-Reinforced Plastic Composites: A Patent-Based Analysis

Beatrice Colombo [1,2,*], Paolo Gaiardelli [1], Stefano Dotti [1], Flavio Caretto [2] and Gaetano Coletta [3]

1 Department of Management, Information and Production Engineering, University of Bergamo, Viale Marconi 5, 24044 Bergamo, Italy; paolo.gaiardelli@unibg.it (P.G.); stefano.dotti@unibg.it (S.D.)
2 Division for Sustainable Materials, Brindisi Research Center, ENEA—Italian National Agency for New Technologies, Energy and Sustainable Economic Development, SS7 Appia Km 706,00, 72100 Brindisi, Italy; flavio.caretto@enea.it
3 Technology Transfer Unit, ENEA—Italian National Agency for New Technologies, Energy and Sustainable Economic Development, Via Martiri di Monte Sole 4, 40129 Bologna, Italy; gaetano.coletta@enea.it
* Correspondence: beatrice.colombo@unibg.it

Abstract: Fiber-reinforced plastic composite materials are increasingly used in many industrial applications, leading to an increase in the amount of waste that must be treated to avoid environmental problems. Currently, the scientific literature classifies existing recycling technologies into three macro-categories: mechanical, thermal, and chemical; however, none are identified as superior to the others. Therefore, scholars and companies struggle to understand where to focus their efforts. Patent analysis, by relying on quantitative data as a precursor to new technological developments, can contribute to fully grasping current applications of each recycling technology and provide insights about their future development perspectives. Based on these premises, this paper performs a patent technology roadmap to enhance knowledge about prior, current, and future use of the main recycling technologies. The results show that recycling macro-categories have different technology maturity levels and growth potentials. Specifically, mechanical recycling is the most mature, with the lowest growth potential, while thermal and chemical recycling are in their growth stage and present remarkable future opportunities. Moreover, the analysis depicts several perspectives for future development on recycling technologies applications within different industries and underline inter- and intra-category dependencies, thus providing valuable information for practitioners and both academic and non-academic backgrounds researchers interested in the topic.

Keywords: patent analysis; technology roadmap; fiber-reinforced plastic composite materials; recycling technologies

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1. Introduction

Fiber-reinforced plastic (FRP) composite materials consist of polymeric matrixes reinforced with fibers characterized by a high length-to-fineness ratio. Carbon fibers (CFs) and glass fibers (GFs) are two of the most commonly employed fibers as reinforcement. While the former is used for the production of carbon fiber-reinforced plastics (CFRPs), the latter are adopted for glass fiber-reinforced plastics (GFRPs). Thanks to their outstanding mechanical properties, low weight, and good fatigue behavior, CFRPs and GFRPs are increasingly being employed instead of traditional materials such as steel or aluminum [1–3] in many applications concerning the transport (e.g., automotive and aircraft), construction, and sport goods industries [4]. In 2017, approximately 1.12 million tons of GFRP were produced in Europe, reaching the sixth consecutive year of growth. In the same year, the volume of global demand for CFRP was around 114 thousand tons, about 14% higher than the value reached the previous year. Moreover, since 2010, the total demand for CFRP has been characterized by a continuous expansion, with an average annual growth rate (CAGR) of 12.8% [5].

The extensive use of FRP composite materials has resulted in a huge increase in generated waste at different stages of their life cycle, especially during production [6]. Indeed, around 30% of CFs is lost during the production phase [7], while losses of GFs generated during the production of GFRPs range between 5 and 10% [8].

Currently, the heterogeneous nature of FRP composite materials does not allow for a suitable recovery of produced waste [9], as the existing recycling technologies are unable to obtain a material of equivalent quality to the original [10]. Therefore, in the best-case scenario (i.e., when recycled), fibers are employed for the production of second-quality products suitable for specific niche applications, instead of structural ones [6,11]. In the worst-case scenario, it is necessary to dispose of the produced waste in landfills [9], causing both huge environmental problems and economic losses for companies. For instance, it is estimated that about 6000–8000 commercial aircrafts will be decommissioned by 2030 [7], thus creating large amounts of CFRP waste. Instead, recovering all this material would not only allow airlines to reduce environmental impacts, but also create new value by re-employing their high embodied energy [12].

Existing recycling technologies for FRP composite materials can be grouped into three main macro-categories, namely mechanical, thermal, and chemical [9]. As pointed out by the scientific literature, each category is characterized by specific advantages and drawbacks [6,13], and thus no technological solution is better than the others. Moreover, it is not yet clear for which industrial applications each recycling macro-category is potentially most effective [14]. This leads to companies not being fully aware of which technology is best to invest in to guide their strategic plan towards sustainability. Likewise, it makes it difficult for researchers to understand which technology they should address in their efforts and which industrial sectors they should concentrate their attention and actions on. Finally, for policymakers, uncertainty about the most promising technologies makes it more complex to establish proper guidelines to encourage sustainable development. Tracing the development of these technologies through a structured approach based on quantitative data would provide researchers and companies with a clearer and more comprehensive view of the potential of each technology. Leveraging objective data as precursors to new technological developments, patent analysis is therefore a research approach consistent with the needs arising in the field under investigation.

Based on these premises, this paper proposes an analysis based on patent data, providing a complementary perspective to traditional qualitative analysis (e.g., a scientific literature review). Consistent with the suggestion of Mehrotra et al. [15], this study employed a technology roadmap to fully grasp the current applications of each recycling technology and enhance knowledge about prior, current, and future use of the main recycling technologies. The analysis was carried out adopting three different techniques, namely time series analysis, patent trend analysis, and citation analysis. In detail, time series analysis was performed to investigate the current technology maturity level, forecast the expected remaining life, and estimate the growth potential of each technological macro-category. The patent trend analysis is proposed to identify the industries interested in the field under consideration and any changes in such interest over time. Finally, the introduction of citation analysis makes it possible to investigate potential connections and interdependencies between recycling technologies over time.

Overall, this analysis sheds light on the main evidence about the existing FRP recycling technologies, providing valuable suggestions for helping academic researchers in their theoretical research activities, non-academic researchers in the practical application of theoretical research, and companies in their strategic planning definition.

The reminder of this paper is organized as follows: Section 2 describes the theoretical background by providing a literature review concerning the recycling processes of FRP composite materials, while Section 3 presents the methodology adopted. In Section 4, the empirical results of the study are reported and discussed. Finally, limitations and further developments are presented in Section 5.

2. Theoretical Background

This section provides a brief review of the macro-categories of recycling technologies for FRP composite materials, as well as the usefulness of patent data as a source of technological information.

2.1. Recycling Processes

As highlighted by Pimenta and Pinho [16], almost all recycling methods are characterized by two sequential stages: "fiber recovery" and "composite re-manufacturing". While fiber reclamation pertains to those activities performed to tear FRP composite material waste into pieces to recover fibers or fibrous products, composite re-manufacturing refers to those operations carried out to re-impregnate recycled fibers, in order to generate new composite materials. Although most recycled composites are GFRPs, the research has been mainly focused on CFRP recycling processes, probably because of their expensive components. FRP composite materials recycling technologies can be divided into three main macro-categories: mechanical, thermal, and chemical [9]. The adoption of a distinctive technology depends on both the material that needs to be recycled and its reuse application [13]. A brief description of each macro-category is reported in the following section.

2.1.1. Mechanical Recycling

As a first step, the mechanical recycling of FRP composites materials involves reducing large waste/scraps into smaller pieces (50–100 mm) by crushing or shredding. Then, through a high-speed mill or a hammer mill, the pieces are further ground (10 mm–50 μm). Finally, recycled elements, i.e., powders and coarse particles, are separated by sieving according to their dimension [9]. As the cost of virgin fillers is very low, the use of recycled powders is commercially inconvenient, especially when considering CFRPs. However, given their resin-rich nature, it is possible to use them as energy sources [13]. The reuse of fibrous fractions, instead, is very difficult and results in a significant reduction in mechanical properties because of the difficulty of adhesion between the recycled product and the virgin polymer [9]. Nevertheless, given its low cost, mechanical recycling is still considered as the best option for GFRP recycling by the extant literature [13,17]. Contrarily, it is not considered worthwhile for CFRP recycling, although the literature proposes some studies on this issue (e.g., [18]). Currently, examples of industrial applications of mechanical recycling are those of the Belgian company Reprocover, (https://en.reprocover.eu/, accessed on 8 December 2020) and Filon Products Ltd. (https://www.filon.co.uk/, accessed on 8 December 2020) in the United Kingdom [13].

2.1.2. Thermal Recycling

Thermal recycling technologies, including pyrolysis and fluidized beds, allow fiber recovery by breaking down the FRP composite material's polymer matrix. This process is activated at an operating temperature ranging from 450 °C to 700 °C. Pyrolysis, applied for the first time on the FRP composite materials by Cornacchia et al. in 1999, involves degrading the polymer matrix by heating the material in the near absence of oxygen ("Realization and testing of a pilot plant for pyrolysis for the recovery of carbon fibers from scrap and waste processing of the same reinforced with epoxy resins (CFRP). Validation of the technology and the process in environmental terms for industrial transfer". Program—"POP-FESR 1994–1999—Measure Research, Development and Innovation" of the Basilicata Region.) During this thermal process, gases and oil are produced. Instead, in the fluidized bed recycling process, developed for the first time by Pickering et al. [19], FRP composite material scraps feed a bed composed of silica sand fluidized by a stream of air that flows at an elevated temperature. Therefore, the process happens in an oxidant environment. The hot air decomposes the polymer matrix, allowing it to release the fibers and fillers that are carried away by the stream. Both processes imply the production of char that contaminates the fiber surfaces, making its recovery difficult. Thus, oxidation post-treatments are mandatory to burn the char and obtain clean fibers and fillers [13]. The fibers reclaimed by thermal processes appear discontinuous, short, and fluffy. However, they can also have modulus and tensile properties comparable to virgin fibers, especially for CFs [20,21]. Thus, if properly treated, these recycled fibers can be used as raw material to prepare new composites. Pyrolysis, among other thermal processes, is the most used in commercial applications; examples of companies implementing pyrolysis technology for FRP composite material recycling are ELG Carbon Fibre Ltd. (http://www.elgcf.com/, accessed on 14 December 2020) in the UK and Karborek Spa (http://www.karborekrcf.it/, accessed on 14 December 2020) in Italy [6].

2.1.3. Chemical Recycling

Chemical recycling encompasses all those treatments for degrading a polymer matrix to reclaim its fibers through the adoption of a chemical reagent. The depolymerization process, also called solvolysis, can be classified into hydrolysis, glycolysis, and acid digestion, when using, respectively, water, glycols, and acid as solvents. Each type of solvent requires adequate temperatures and pressures to dissolve the resin properly and to achieve good levels of efficiency [22]. Nevertheless, solvolysis is generally less energy intensive than pyrolysis, as it operates at lower temperatures, especially for epoxy resins and unsaturated polyester [13]. Moreover, the use of water and alcohol as solvents is environmentally friendly, as these materials are not hazardous and can be recovered after the reaction by evaporation and distillation [10]. Solvolysis allows the reclaiming of not only clean fibers and fillers but also the monomers deriving from resin depolymerization [10]. In addition, fibers recovered through solvolysis are characterized by high mechanical properties and fiber length [6]. However, they suffer from a reduction in bonding to the polymer matrix [23]. Finally, chemical recycling, unlike the fluidized bed, is characterized by a low tolerance to contamination [10,24].

2.2. Advantages and Drawbacks of Recycling Processes

Overall, scientific research underlines that when considering the high commercial value of CFs and the decrease in GFs' mechanical properties due to thermo-chemical technologies, mechanical recycling can be viewed as the best option for GFRPs, whereas pyrolysis and solvolysis can be considered as the best option for CFRPs. Notwithstanding, it is also clear from the literature that the choice of a proper technology depends on the material that is to be recycled and on its reuse applications [13], and that each macro-category is characterized by several strengths and disadvantages, as summarized in Table 1. Recent research has also suggested that the best route to overcome the disadvantages of thermal and chemical processes would be a new hybrid technology. For instance, the American company Adherent Technologies proved that making a three-step process composed of a thermal pre-treatment followed by two solvolysis steps, the first at a low temperature and the second at a high temperature, can produce recycled fibers with a very good quality, especially for CFs [25].

2.3. Patent as a Source of Technological Information

A patent is a form of intellectual property that grants its owner the exclusive right to exploit an invention, in a territory and for a specified period, and allows them to prevent others from producing, selling, or using the invention without authorization [29]. Consisting of different types of structured (i.e., assignee and citations) and unstructured data such as patent title, abstract, claims, and description [30], patents are also powerful sources of valuable information for industry, business, and society. According to Asche [31], about 80% of the technical information contained in a patent document is not available in other types of sources. Therefore, in-depth and appropriate analyses of patents can enable a better understanding of current business trends and help researchers and companies to better direct their research efforts [32].

Table 1. Advantages and drawbacks of recycling processes.

	Advantages		Drawbacks	
Mechanical recycling	Relatively simple	[9]	Decrease in mechanical properties	[9,17]
	Environmentally friendly	[6]	Recovery of both fibers and resin as powders and coarse	[9,18]
			Few applications for remanufacturing	[6]
Thermal recycling	High mechanical properties	[6,20]	Fiber quality depends on process parameters	[26,27]
	Environmentally friendly	[6]	Recycled fibers in fluffy form	[9]
	With pyrolysis, production of oil from the polymer matrix	[9]	With pyrolysis, possibility of char on fiber surface	[26]
	With fluidized bed, clean fiber surface and good tolerance to contaminated materials	[9]	With fluidized bed, no recovery of polymer matrix and greater decrease in fibers strength and length	[13,19]
Chemical recycling	Recovery of both fibers and resin (the latter as monomers)	[10]	Generally, not environmentally friendly	[13,28]
	High mechanical properties and fiber length	[6]	Low tolerance to contaminated materials	[10]
			Reduction in bonding between fibers and new resin	[23]

Patent data can be analyzed in order to pursue different objectives. As stated by Abbas et al. [33], it can be exploited for determining the novelty and quality of inventions, determining the most promising technologies, including their vacuums and hotspots, forecasting developments and trends to support technological roadmaps and the implementation of strategic plans, and identifying infringements, as well as technological competitors. The scientific literature underlines that most studies use patent analysis with the aim of capturing the evolution of research and technology [34]. Among others, Mehrotra et al. [15] state that patent analysis can be of four approach categories, each one adopted to pursue specific objectives. Consistently distinctive techniques can be used to implement each type of patent analysis approach, as briefly summarized in Table 2.

Table 2. Summary of patent analysis types (authors' elaboration from Mehrotra et al. [15]).

Patent Analysis Type	Description	Techniques
Technology roadmap	To provide a commercial prospect and to understand perspectives, prior and current use for the considered technology	Time series analysis Patent trend analysis Citation analysis
Originality/R&D support	To compare a new patent with an existing one in a specific technical domain	Infringement analysis Novelty analysis White spot analysis
Technology classification	To analyze the perspectives of key technologies, research, development trends, and changes for a specific technical field	IPC analysis Family analysis Cluster analysis
Technology distribution	To provide information about technology concerning the main technology domain, competitors, assignees, etc.	Technology distribution by country Association rule analysis Assignee analysis Rival/competitor analysis

3. Methodology

To achieve the objective of this study, a patent technology roadmap was employed. Specifically, a time series analysis was carried out to highlight the technology maturity level of existing mechanical, thermal, and chemical recycling technologies of FRP composite materials, forecast their expected remaining life, and estimate the growth potential. A patent trend was then proposed to identify the main industries affected in the field under consideration and to explore their potential change in interest over time. Finally, a citation analysis was introduced to detect any connection and interdependencies between existing recycling technologies. A detailed description of each approach is reported in the following.

3.1. Time Series Analysis

Time series analysis was carried out to investigate the growth/decline rate of a technology over time [35]. Specifically, it was implemented by using a Gompertz growth curve model, as proposed by Yoon et al. [36] and confirmed by Bengisu and Nekhili [37]. In detail, the cumulative number of patents filed between 1991 and 2017 was used as input data. This was possible as the life cycle of a technology, whose development trend can be approximated by the number of patents accumulated over time, can be represented by an S-shaped curve [38]. The Gompertz model is defined by the following equation:

$$Y_t = Le^{-ae^{-bt}} \quad (1)$$

where L is the asymptotic maximum value (i.e., the saturation level) achievable by Y_t which represents the dependent variable of the model (i.e., the cumulative number of patents belonging to the single macro-category at time t), while a and b represent the location and the shape of the curve, respectively.

To identify the value of the three parameters, a least square estimation method was employed [36]. Moreover, to practically perform calculations and, accordingly, fit the collected data, the online Loglet Lab software (https://logletlab.com/, accessed on 17 February 2021) was adopted. The software allows us to test the statistical significance of the results and, by processing the curve fitting, automatically generate a S-shaped curve. Furthermore, it allows us to define the stage reached by each technology in its life cycle which, in accordance with Haupt et al. [39] and Mann [40], may assume four different configurations: birth, growth, maturity, and saturation, where each configuration is distinguished by distinctive shares of upper limits [41], as shown in Table 3.

Table 3. Stage of technology life cycle by share of upper limit (authors' elaboration from Ranaei et al. [42]).

Share of Upper Limit	Stage of Technology Life Cycle
$Yt/L \leq 10\%$	Birth
$10\% < Yt/L \leq 50\%$	Growth
$50\% < Yt/L < 90\%$	Maturity
$Yt/L \geq 90\%$	Saturation

3.2. Patent Trend Analysis

Patent trend analysis was carried out to achieve the second objective of this study. First, the assignees, i.e., the patent owners, were divided into two main groups: organizations operating in the public or private sector and individuals. The first group was further divided in two subgroups: the former includes both research centers and universities, while the latter private companies. Then, private organizations were classified considering their NACE code (Revision 2), as offered by the database Orbis (https://orbis.bvdinfo.com/, accessed on 6 March 2021) of the Bureau van Dijk. Second, each assignee was assigned a score ranging from 0 to 1 according to the share of ownership of the specific patent considered: for instance, in the case of a single owner the value assigned was 1, while if there were two owners the value became 0.5. This assumption was adopted in order to avoid overestimating the patents' ownership. Third, by adding up the scores calculated, the interest of each sector (i.e., NACE codes, Research and Individuals) was identified. Finally, to explore the potential change in interest over time, a year-by-year stratification was carried out.

3.3. Citation Analysis

Citation analysis was developed by using the backward citations contained in the cited patents. Since backward citations give information about the technological antecedents of an invention [43], they were considered particularly suitable for achieving the third objective of this study. Specifically, the backward citations (i.e., cited patents) of each patent in our database (i.e., citing patents) were classified considering their belongingness to one of the three main recycling macro-categories: mechanical, thermal, and chemical. A fourth macro-category ('others') was used to classify those citations that did not belong to any of the previous categories. Thus, the publication numbers of the cited patents were cross-checked with the publication numbers of the patents in the overall dataset. By comparing these groups with the total number of backward citations of the respective macro-category, an interdependence index was created. In detail, this index measures the degree of citation within and between each macro-category.

3.4. Workflow of the Study

Overall, the study research procedure includes five-stages: database definition, search strategy definition, patent pool retrieval, patent analysis, and discussion of results. In the first stage, the search database was defined. In particular, data were collected using the Intellectual Property Portal of Questel Orbit Company (https://orbit.com/ accessed on 4 December 2020) which is a software providing data mining tools to analyze individual databases with unique features, e.g., legal status. Furthermore, Orbit is based on patent families, namely a set of granted patents and patent applications protecting the same invention [29]. This avoids any duplication in the analysis of patent documents, which generally occurs when companies patent the same invention in multiple countries [44]. The second stage involved the identification of the search strategy. In particular, a mixed strategy was adopted to overcome the limitations of both the keywords strategy and classification code strategy. First of all, a search by keywords was carried out. Specifically, keywords related to the FRP composite material recycling processes domain were typed in the English language considering all possible variants, and were searched in the title, abstract, and claims field within the patent record. (Note: more information on keywords can be found in Appendix A, Table A1.) Concurrently, an attempt was made to identify the relevant International Patent Classification (IPC) codes. To this end, a classification code tree was browsed, followed by a reverse engineering analysis to identify the IPC codes relevant for the analysis. The results of the two analyses were combined and a group of experts in the field was then involved to refine the sample of selected patents. Albeit being time consuming, this activity allowed us to identify those words producing noise in the sample. Moreover, an algorithm based on keyword characteristics, their proximity in the document, and their recurrence was adopted to minimize the remaining noise following the manual screening. Subsequently, this sample was divided in agreement with the three recycling macro-categories (mechanical, thermal, and chemical) by using specific keywords (Note: more information on keywords can be found in Appendix A, Table A2.). The three resulting samples contained 205 patent families for mechanical recycling processes, 389 for thermal recycling processes, and 494 for chemical recycling processes. In the fourth stage, the patent analysis was carried out through times series, patent trend, and citation study. Finally, in the fifth stage, an in-depth discussion of the obtained results was executed.

All patents considered in this study were characterized by a priority date, i.e., the year in which the patent was first filed in a legal office, between 1991 and 2020. This choice was made because before 1991 patent activity concerning this topic was insignificant when compared to the period evaluated. The authors are also aware that the two-year period 2019–2020 is not representative of the trends under analysis due to the 18 months delay between the filing and publication. Lastly, the patent capture took place in December 2020.

4. Empirical Results and Discussion

This Section sheds light on the main results obtained from the patent technology roadmap. In particular, the main findings of the time series analysis are provided in Section 4.1, while Sections 4.2 and 4.3 report the main outcomes of patent trend and citation analysis, respectively. In conclusion, Section 4.4 provides some glimpses into future trends.

4.1. Time Series Analysis

Patenting activity on FRP composite materials was characterized over the last 30 years by a continuous growth, reaching the maximum number of published patents (82) in 2018, as shown in Figure 1a. Such a trend underlines the up-surging interest of research in the development of new technologies for FRP composite materials recycling. Nevertheless, the sharp decrease in patenting activity that marked the two-year period 2019–2020 cannot be considered relevant for the study, as it is unreliable. Indeed, many patents registered in this period might not have been published yet, as the average delay between patent registrations and publications was typically 18 months. The high interest of research in developing new and effective recycling techniques for FRP composite materials can be also inferred by comparing the historical trend of the relevant patenting activity at a worldwide level (Figure 1b). Moreover, such a comparison shows that patenting activity on recycling technologies for FRP composite material is growing slightly faster than patenting activity worldwide. The underlying motivation is manifold and might depend on internal and external forces impacting companies' decisions. Indeed, legislation has increasingly pushed towards environmental sustainability [45]. In addition, society as a whole is becoming more and more environmentally sensitive [46]. Customers, whether B2B or B2C, are also encouraged to buy or have products in line with the principles of sustainability and circular economy for many reasons, such as remaining competitive on the market and increasing their social status, respectively [47,48]. Such rapid growth also derives from internal factors, mainly related to the need of companies to find technological solutions able to improve production efficiency and therefore lower their costs. Nevertheless, recycling can help companies be less susceptible to market fluctuation. For instance, the possibility of recycling larger amounts of CFs would allow companies to be less dependent on the virgin raw material price, whose value is highly correlated with that of oil which, by its nature, is characterized by high volatility [49].

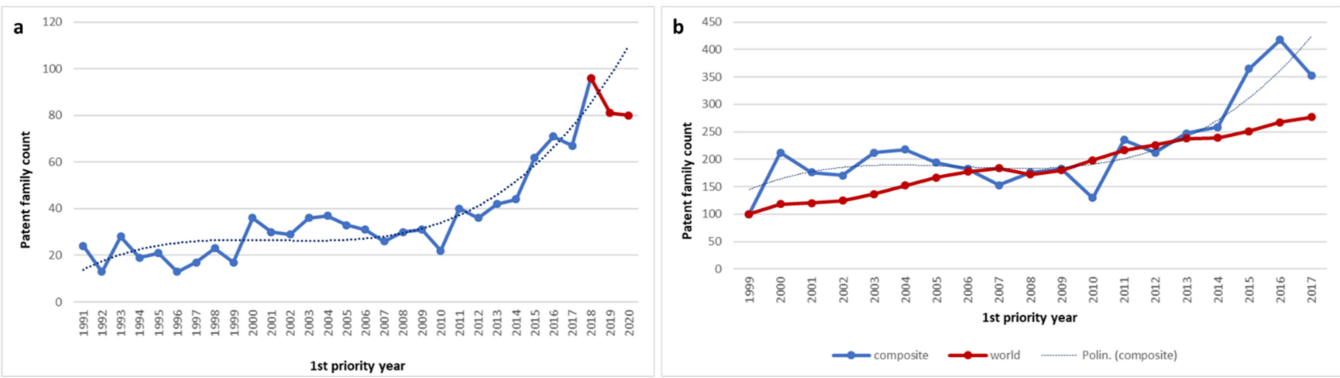

Figure 1. (**a**) Historical trend of patenting activity by first priority year (blue line represents the trend of patents filed between 1991-2018, red line represents the trend of patents filed during the non-representative two-year period 2019–2020, and dotted line represents the overall trendline); (**b**) benchmark between composite recycling processes patent activity and worldwide patent activity.

As depicted by the Gompertz growth curve presented in Figure 2, the FRP recycling technologies market is still in its growth stage (technology maturity level (TML) = 36.57%). Nevertheless, the number of patents is expected to continue this trend in the following years until about 2060 (expected remaining life (ERL) = 46 years). Finally, the potential patents to appear (PPA = 1,511) indicates that the future patenting activity in this market will be more active than in the past, suggesting academic and industrial researchers will continue focusing on the topic.

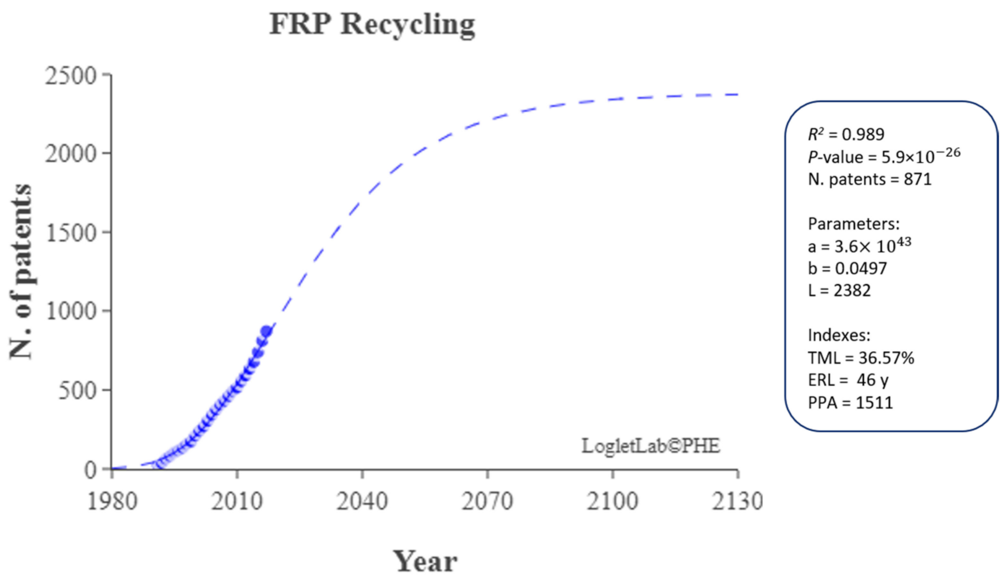

Figure 2. Gompertz growth curve and its parameters for FRP recycling.

Figure 3 depicts the development of the patents filed on mechanical, thermal, and chemical recycling processes from 1991 to 2020. The data show that the patenting activity has been continuously growing in all three recycling macro-categories since 2010. Instead, the analysis of the Gompertz growth curve (Figure 4) reveals that mechanical recycling technologies have already entered the maturity stage (TML = 61.51%), while thermal (TML = 36.54%) and chemical ones are still in their growth phase. Specifically, chemical recycling technologies have the lowest level of maturity (TML = 30.96%). Furthermore, the results show that chemical (ERL = 48 years) and thermal recycling (ERL = 51 years) technologies have an estimated remaining life about twice that of mechanical recycling technologies (ERL = 24 years). In summary, the data analysis indicates that the progressive growth in the number of patents takes on different forms when viewed from the perspective of the three recycling macro-categories, pointing out the existence of technologies that have reached their maturity faster than others. This behavior may be due to the complexity and history of each technology. For instance, the mechanical recycling of CFs has grown more quickly than thermal and chemical recycling, as it is simpler and has been developed based on existing knowledge and technologies already established in other industries. In addition, being at an advanced stage of maturity, it is about to reach the technological upper limit of the technological transformation process, intended as the set of technical actions necessary to recover the fibers. Vice versa, there is still ample room for improvement for the technological transformation system, defined as the set of processes, tools, and equipment suitable for an effective, efficient, and reliable recycling process. Contrariwise, thermal and chemical recycling, as they are in the growth phase of their life cycle, are still characterized by opportunities for growth and improvement from a technological perspective.

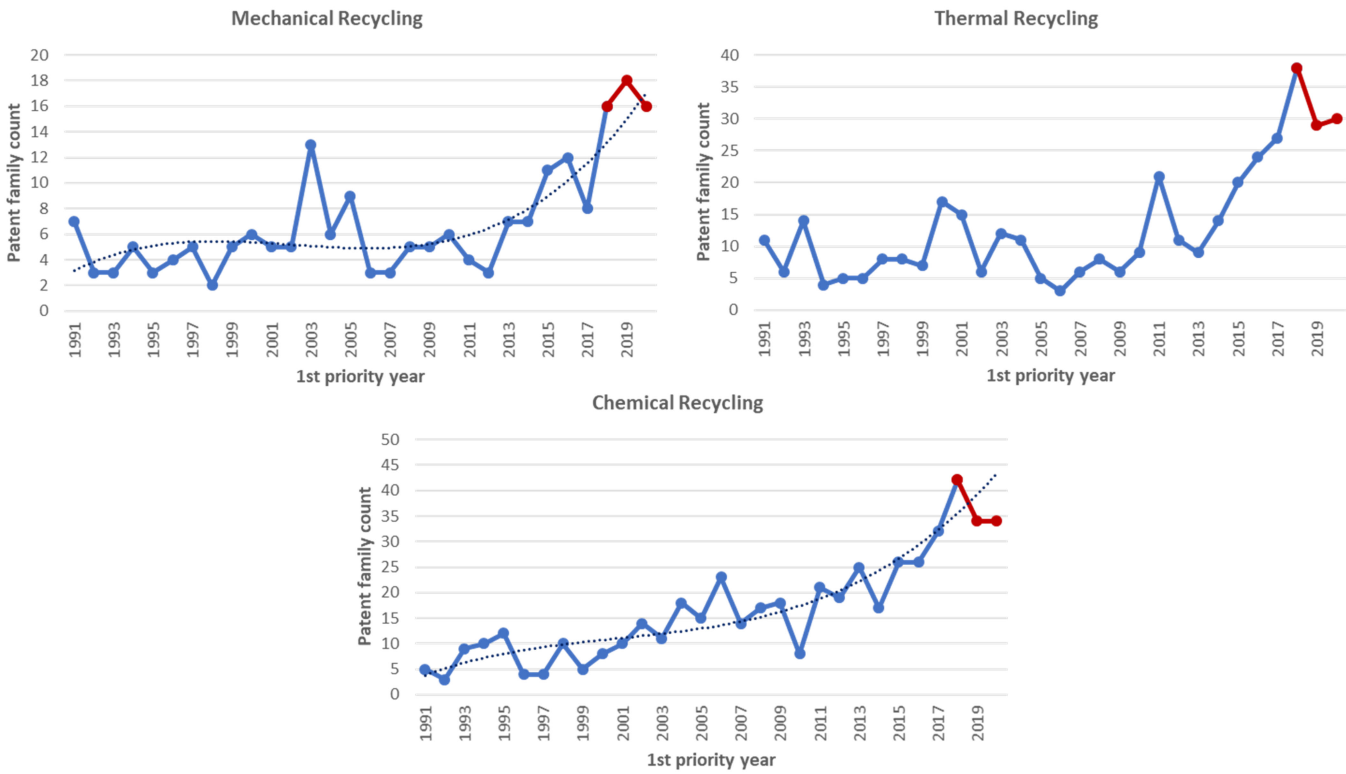

Figure 3. Historical trend of mechanical, thermal, and chemical patenting activity by first priority year (blue line represents the trend of patents filed between 1991–2018, red line represents the trend of patents filed during the non-representative two-year period 2019–2020, and dotted line represents the overall trendline).

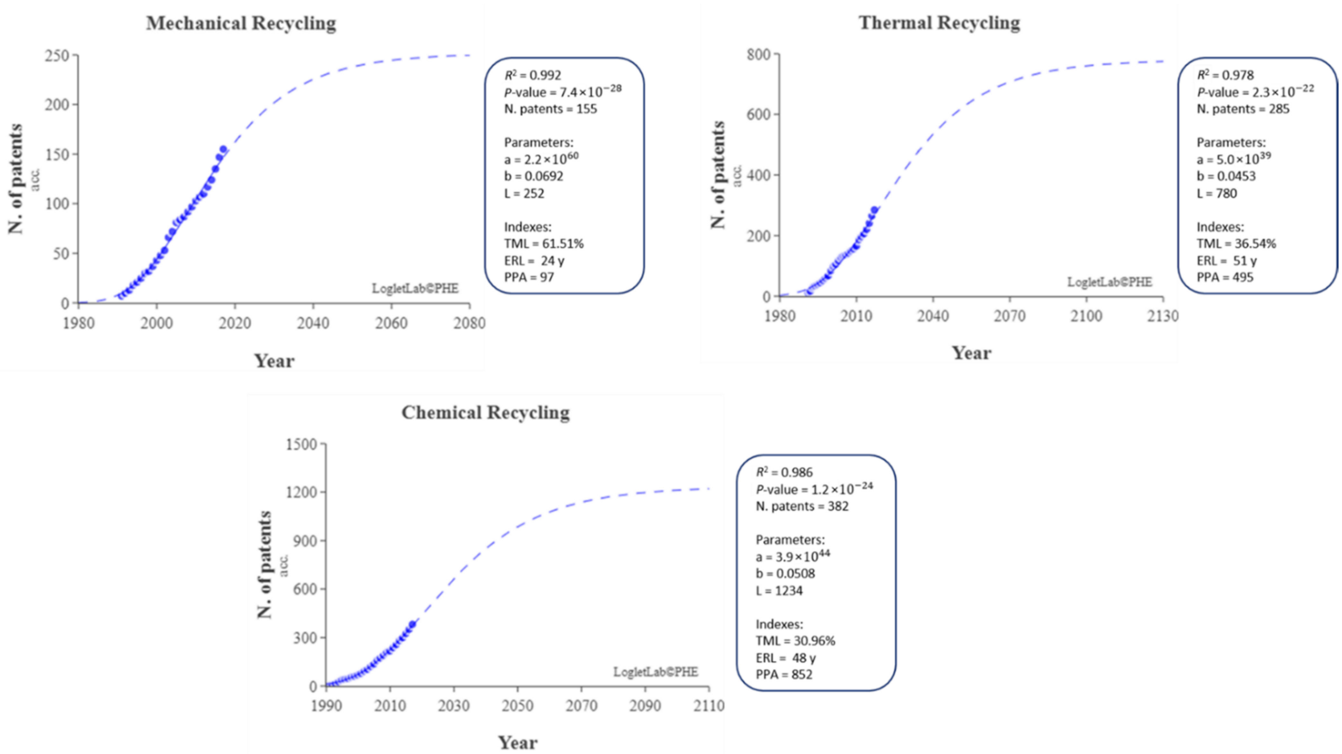

Figure 4. Gompertz growth curve and its parameters for mechanical, thermal, and chemical recycling technologies.

4.2. Patent Trend Analysis

Table 4 reveals the most active actors in this field, sorted by recycling macro-category.

Table 4. Patent holdings by recycling macro-category and assignee category.

Actors	Mechanical Recycling	Thermal Recycling	Chemical Recycling
Research	8.4%	15.0%	26.7%
Individuals	10.2%	9.8%	6.6%
Manufacture of chemicals and chemical products	14.8%	13.8%	22.5%
Manufacture of rubber and plastic products	6.4%	6.9%	4.0%
Manufacture of electrical equipment	3.8%	7.1%	13.0%
Manufacture of motor vehicles, trailers, and semi-trailers	10.1%	7.1%	3.1%
Construction of buildings	6.6%	2.7%	-
Other	39.7%	37.6%	24.1%

First, the data indicate that the top categories of assignees affected by the field under investigation are the same for each recycling macro-category, meaning an in-depth concentration of interest in the topic. Second, it can be noticed that 'Research' is the most important domain for both chemical and thermal recycling, with 26.7% and 15.0%, respectively, while for mechanical recycling it ranks only in fourth position, with 8.4%. These findings prove the level of maturity of the technologies highlighted in Section 4.1. Indeed, high percentages of patents developed at the research level testify that the development of chemical and thermal recycling technologies is still at an early stage. Furthermore, the data show that patents related to mechanical recycling are more evenly distributed across the industrial sectors than those for thermal and chemical recycling technologies. This result may be traced back to two different reasons: as specified, mechanical recycling is a more consolidated technology and therefore more widespread at an industrial level; in addition, it is mainly employed for the recycling of GFs [13,17] which are used for the production of several applications belonging to various industries [5].

The findings also highlight that mechanical recycling is interesting for sectors concerning industrial applications (i.e., 'Manufacture of motor vehicles, trailers and semi-trailers', and 'Construction of buildings'), as opposed to thermal and chemical recycling, whose patents mainly refer to 'Manufacture of chemicals and chemical products'. This result proves that the development of chemical and thermal technologies requires very strong competencies and skills that can be found in universities, research centers, or in those companies that deal with those specific processes and technologies.

More evidence clearly emerges when considering the patent evolution over time in the four main assignee categories. As shown in Figure 5, 'Research' suffered a continuous growth with a strong increase in the second decade of the 21st century, especially for thermal and chemical recycling. This is further evidence that these two recycling macro-categories are both in their growth stage and are based on much more complex underlying principles, thus requiring strong research skills. Contrariwise, for mechanical recycling, patents developed at the research level show much slower growth, underlining that the contribution to the development of this technology by the industrial field is more significant. Concerning industries, a flattening of the number of equivalent patents is observed for almost all the recycling macro-categories in "Manufacture of electrical equipment", highlighting that the technological development within this industry has almost reached its end. In addition, the increase in the number of equivalent patents for mechanical recycling was marginal in recent years, except for the 'Manufacture of motor vehicles, trailers and semi-trailers' sector, where there is a strong need to find solutions aimed at reducing product and production costs. Finally, 'Manufacture of chemicals and chemical products' shows an increasing trend only for thermal and chemical recycling, since, as already pointed out, it is precisely from such an industry that these recycling macro-categories draw.

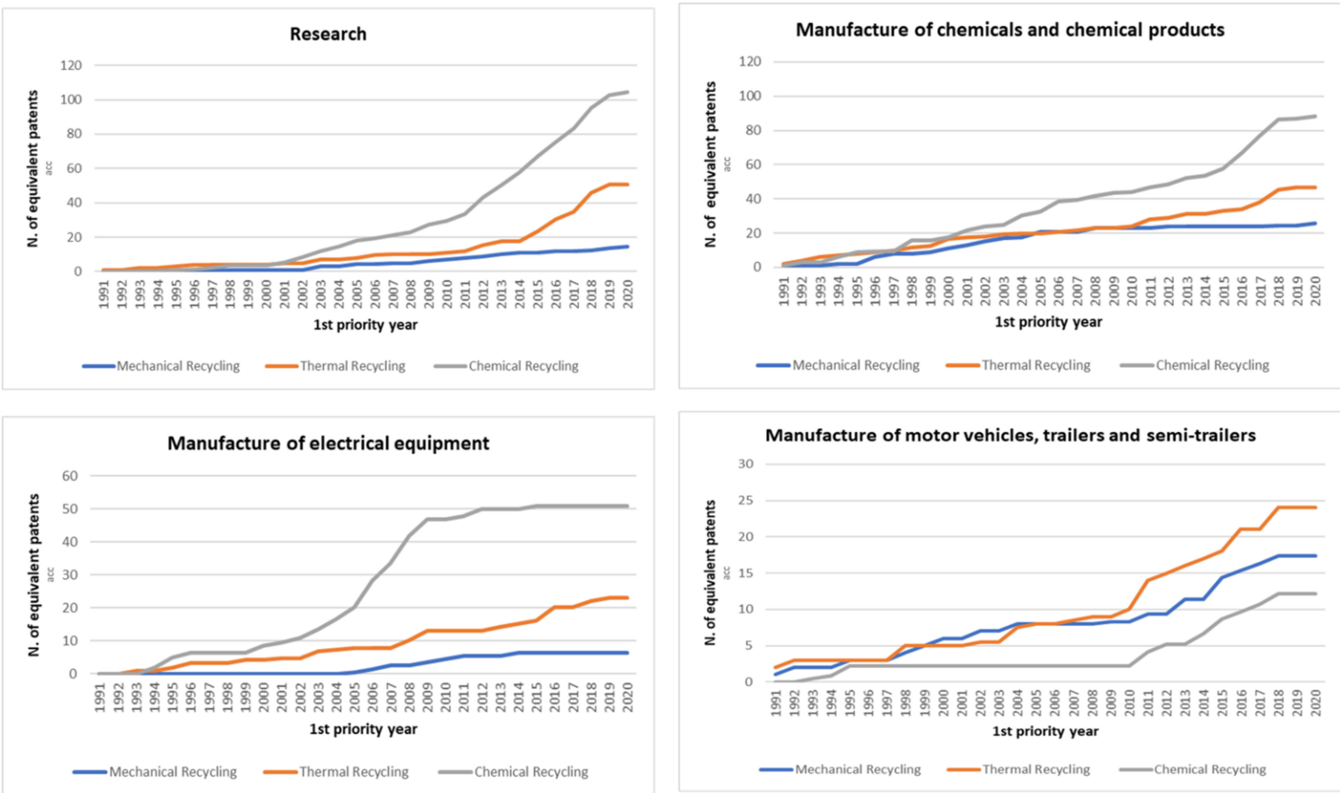

Figure 5. Cumulated patents for the main 4 assignee categories by recycling macro-category.

4.3. Citation Analysis

Backward citation analysis of patents related to FRP mechanical, thermal, and chemical recycling technologies allows for the identification of their potential existing interdependencies.

As reported in Table 5, where the rows represent the recycling macro-categories to which the citing patents belong (i.e., the patents that compose our database) and the columns specify the recycling macro-categories to which the cited patents belong, it may be argued that the average number of backward citations per patent is similar among the different recycling macro-categories. In detail, chemical recycling cites the most patents. Moreover, mechanical, thermal, and chemical interdependency shares appear very comparable, although the mechanical recycling macro-category cites patents referring to 'other' to a greater extent (90.5%) than the thermal (81.6%) and chemical (82.3%) recycling macro-categories.

Table 5. Distribution of backward citations per type of citation and citing patent and average number of backward citations per citing patent.

	Mechanical	Thermal	Chemical	Other	Average Number of Citations Per Patent
Mechanical	3.3%	3.0%	3.1%	90.5%	5.9
Thermal	1.4%	11.4%	5.6%	81.6%	5.6
Chemical	0.9%	2.8%	14.0%	82.3%	6.6

To better understand this difference, this overall picture is discussed in the light of the evolution of technology citation over time. In particular, as reported in Table 6, the distribution of backward mechanical recycling citations shows a decreasing dependence over time on the 'other' category, but is still high, due to the origin of these technologies that have developed by borrowing technologies from other industries.

Table 6. Distribution of backward mechanical recycling citations per type of patent cited.

Year		Mechanical	Thermal	Chemical	Other
1991	1995	2.1%	1.1%	1.1%	95.8%
1996	2000	5.3%	2.7%	0.7%	91.3%
2001	2005	3.6%	3.2%	1.6%	91.6%
2006	2010	2.5%	4.1%	4.1%	89.3%
2011	2015	2.6%	2.2%	5.5%	89.7%
2016	2020	3.7%	5.2%	4.4%	86.7%

On the contrary, although both chemical and thermal recycling macro-categories drew extensively from 'other' patents, this dependence sharply declined over time, in favor of a gradual increase in the number of citations within the same macro-category, as shown in Tables 7 and 8.

Table 7. Distribution of backward thermal recycling citations per type of patent cited.

Year		Mechanical	Thermal	Chemical	Other
1991	1995	0.5%	2.2%	1.1%	96.2%
1996	2000	0.0%	3.1%	2.1%	94.8%
2001	2005	0.0%	4.8%	4.3%	90.9%
2006	2010	1.9%	13.1%	6.3%	78.8%
2011	2015	1.6%	11.6%	3.3%	83.5%
2016	2020	2.9%	22.6%	13.6%	60.9%

Table 8. Distribution of backward chemical recycling citations per type of patent cited.

Year		Mechanical	Thermal	Chemical	Other
1991	1995	0.4%	0.8%	1.2%	97.6%
1996	2000	0.0%	3.3%	1.6%	95.1%
2001	2005	0.4%	0.9%	11.4%	87.3%
2006	2010	1.1%	0.8%	15.6%	82.5%
2011	2015	1.0%	5.4%	15.9%	77.7%
2016	2020	1.6%	4.4%	23.0%	71.0%

This phenomenon of convergence, which also emerges clearly in the mechanical recycling macro-category, underlines a process of progressive focus of research, in accordance with [50]. Nevertheless, it should be pointed out that over time, the number of patents from a specific macro-category that cite patents from other categories increased. If with reference to the macro-category of mechanical recycling the citations of other categories increase in a very small way, underlining a greater orientation towards internal cohesion through the consolidation of knowledge taken from outside, it is different for the macro-categories of chemical and thermal recycling, characterized by similar self-citation patterns. In both cases, citations toward the mechanical area increase, underscoring its status as a functional precondition for the development of thermal and chemical skills. In addition, it is interesting to note that the citations of patents belonging to the macro-category of chemical recycling from thermal recycling patents have grown significantly (13.6% during the period 2016–2020), demonstrating a progressive dependence of research in the area of thermal technologies on that in the area of chemical technologies. Vice versa, citations of patents belonging to the macro-category of thermal recycling from chemical recycling

(4.4% over the 2016–2020 period) grew less significantly, demonstrating greater research independence in the area of chemical recycling.

4.4. Glimpses into Future Trends

Based on the outcomes emerging in the previous sections, some considerations about the future trends of FRP recycling technologies can be drawn.

First, it is feasible to expect that the near future will be characterized by a progressive and continuous growth in the number of patents, in particular for the thermal and chemical recycling macro-categories, due to their lower level of technological maturity. Overall, an acceleration in patent growth can be assumed due to increasing pressure from society, legislation, and the rising market sensitivity to the environmental sustainability dimension [51]. As regards mechanical recycling, having reached a technological limit that poses a not-so-high level of quality of the recycled raw material [9], it is reasonable to foresee that research in this area will have to be aimed primarily at production system reliability, barring disruptive innovations. Indeed, it is recognized that when a certain technological limit is reached, the research emphasis shifts to other dimensions that are more related to the manufacturing process and its reliability [40].

In contrast, in the case of thermal and chemical recycling processes, future research should still focus on overcoming the inefficiency of existing technologies [40], as there are still important margins of improvement, as suggested by the time series analysis.

Moreover, future research in mechanical recycling should be increasingly devoted to industry instead of basic research, specifically, focusing on those sectors where the need is not to have high-quality products for structural applications, but to establish the best conditions to minimize product and production costs. A clear example of this is the automotive industry [5], characterized by an increasing interest in reducing the weight of vehicles and improving driving dynamics by using cheaper and more sustainable materials to decrease their environmental impact and comply with existing regulations [50]. In such a context, it may be stated that, though marginal, innovations for mechanical recycling will probably come from this sector, where the interest to exploit the lightness of FRP composite materials also for non-structural components emerges as crucial.

Conversely, research concerning thermal and chemical recycling will continue to be used to recover raw material potentially suitable for the production of structural products. Furthermore, at least in the near future, research in these categories will continue to be developed by universities and research centers where basic research is more rooted, but also by those industries that have specific technology and process knowledge. Undoubtedly, these types of recycling require significant upfront investments in terms of both competences and physical and economic resources that cannot be borne by all companies, suggesting that research in this field will be conducted by incumbent players. However, as knowledge in this area advances, the research focus will progressively shift from the perspective of the transformation process to that of the transformation system, thereby stimulating diffusion into other industries typically covered by mechanical recycling technologies.

Furthermore, it is reasonable to expect that research on mechanical recycling will increasingly focus on itself. Indeed, unless disruptive technology is developed, the goal of potential new patents will be to increase recycling process reliability and once consolidated, research having reached its natural limit, will restrict its area of action to marginal activities. Contrariwise, in the next few years, research on thermal and chemical recycling macro-categories, still being in an expansion phase, will continue to maintain a lower level of cohesion than the mechanical technologies, but it is expected to grow over time as research will consolidate. Finally, as underlined by an increasing level of cross-citations between chemical and thermal macro-categories, it can be claimed that research entropy, namely the drawing extensively from different macro-categories in an unsteady manner over time [48], will significantly rise, suggesting that investigations on chemical and thermal recycling will tend to converge into a common field of interest.

5. Conclusions

In this paper, a patent technology roadmap of FRP composite material recycling processes was carried out. To this end, patents were retrieved from the Intellectual Property Portal of Questel Orbit Company database, and then analyzed through time series analysis, patent trend analysis, and citation analysis. To the best of the authors' knowledge, this research is the first to provide an overview of FRP composite material recycling technologies and potential future directions using a patent-based analysis, leveraging a set of quantitative data.

This study sheds light on some evidence. FRP composite material recycling is in its growth stage and its expected remaining life is quite long, thus warranting further studies. Moreover, this work highlights that thermal and chemical recycling are the most promising and challenging processes within the field. Instead, mechanical recycling seems to be less appealing since it has already reached its maturity stage. Patents filed within this recycling category are mainly developed by final industries. Furthermore, specific inter- and intra-category dependencies have been identified over time.

The findings of this study can provide valuable suggestions for practitioners to make decisions about future technological investments in the field of FRP composite material recycling, but also academic and non-academic researchers. Indeed, the former could understand where to target its research efforts as well as find new development areas, while the latter could identify where to focus its efforts towards application research (i.e., to turn scientific research output into practical applications). Nevertheless, this research should be extended through the application of other patent analysis methods to analyze the perspective of key technologies, and changes to a specific technical field, rather than provide information about the evolution of the technological domain and technological competition. In addition, this study has some limitations that should be acknowledged. First, the patent analysis was carried out assuming patents as a good and reliable source of technological information. However, not all innovations are able to be patented, since the patenting trend may vary from country to country and from sector to sector, and patenting is not always the best approach to protect an innovation [29]. Second, the strategy used to find relevant patents gives rise to some problems. On the one hand, the main limitation of the keyword-based approach regards the inconsistent terminology between companies, inventors, and patent grantors [44]. On the other hand, employing many IPC codes can both lead to a high number of irrelevant patents and make it difficult to study a particular field of interest [52]. The manual screening of the retrieved patents allowed us to overcome, as far as possible, these limitations. Despite these shortcomings, the objective set was achieved. However, for future research, it is suggested to perform potential industrial outlet sectors for recycled FRP composite materials, as well as the realization and management of potential industrial symbiosis, which could improve the composite industry's supply chain and the synergy between recycling technologies.

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Appendix A

Table A1. Search keywords for identifying the overall patent pool.

Search Keywords	
Overall patent pool	Fiber-reinforced plastic Fiber-reinforced polymer CFRP GFRP Recycling Recovering Reclaiming

Table A2. Search keywords for identifying the patent pool of each recycling macro-category.

Search Keywords	
Mechanical recycling	Mechanical Pulverizing Powdering Grinding Shredding Crushing
Thermal recycling	Thermal Thermic Pyrolysis Fluidized bed
Chemical recycling	Chemical Solvolysis Hydrolysis Glycolysis Acid Solvent

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