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Review

Recycling of Plastic Waste: A Systematic Review Using Bibliometric Analysis

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Abstract: Research into plastic recycling is rapidly increasing as ocean and land pollution and ecosystem degradation from plastic waste is becoming a serious concern. In this study, we conducted a systematic review on emerging research topics, which were selected from 35,519 studies on plastic recycling by bibliometrics analysis. Our results show that research on the biodegradability of plastics, bioplastics, life cycle assessment, recycling of electrical and electronic equipment waste, and the use of recycled plastics in construction has increased rapidly in recent years, particularly since 2016. Especially, biodegradability is the most emerging topic with the average year of publication being 2018. Our key finding is that many research area is led by developed countries, while the use of recycled plastics in the construction sector is being actively explored in developing countries. Based on our results, we discuss two types of recycling systems: responsible recycling in the country where plastic waste is generated and promoting recycling through the international division of labor between developed and developing countries. We discuss the advantages and disadvantages of both approaches and propose necessary measures for sustainable and responsible production and consumption of plastics such as waste traceability system and technology transfer between developed and developing countries.

Keywords: plastic recycling; plastic waste; circular economy; plastic pollution; mechanical recycling; chemical recycling; biodegradation; bioplastics; e-waste; plastics in construction

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1. Introduction

Plastics are a lightweight, durable, and inexpensive material that can be easily processed into a wide range of products to be used in a variety of applications. Given the ubiquity of food packaging and daily necessities that surround us, plastics have become an indispensable material in many industries such as construction, engineering, medical, automotive, and aerospace. Economic development and growth have increased the demand for and dependence on plastics, and global production and consumption of plastics has increased dramatically over the past decades. Most of the plastics produced each year are used in short-lived products that are discarded within a year of manufacture, such as food packaging disposables. Much of this plastic waste is sent to landfills or dumped on to natural habitats, where it accumulates substantially due to its high durability. As a result, it causes serious environmental pollution problems such as groundwater contamination and hygiene-related problems and poses great risks to human health and ecosystems. In addition, due to inappropriate waste disposal, a large amount of plastic waste flows into the ocean, causing a serious marine plastic litter problem [1].

Sustainable and efficient treatment of plastic waste is essential to solve such problems, and interest in recycling plastic waste has been rapidly increasing worldwide. Based on the idea of using plastics in a cyclical manner, the EU has proposed the concept of a circular economy, and there are active movements outside the European Union (EU) to pursue circular economies. The International Organization for Standardization (ISO) is formulating international standards for the circular economy with the participation of many countries [2].

Under these circumstances, many papers on plastic recycling have been published, though researchers have differing classifications of plastic recycling methods. Some papers classify recycling into four categories: primary recycling (re-extrusion), secondary recycling (mechanical), tertiary recycling (chemical), and quaternary recycling (energy recovery) [3]. The difference between primary and secondary recycling is largely due to the degree of contamination of the plastic waste used, though there are many studies that collectively refer to these as mechanical recycling. Energy recovery does not reproduce new materials or raw materials, but the extraction of energy in the form of heat, so it is usually regarded as recovery rather than recycling. The ISO has created international standards for plastic waste disposal methods, in which plastic recycling is classified into three types: mechanical, chemical, and biological recycling [4]. In this study, we classify and review recycling methods according to this ISO classification.

Mechanical recycling is desirable in that it enables the recycling of plastic waste into plastics, but recycled plastics have weaknesses such as inferior quality and reduced strength compared to virgin plastics [5]. Especially in developed countries, it is necessary to promote consumer understanding of the environmental value of recycled plastics in order to promote the replacement of virgin plastics with recycled ones. Chemical recycling is an effective means of complementing mechanical recycling, but it mainly produces industrial products such as fuel and ammonia, and it is generally difficult to produce recycled plastics with this process. However, some developed countries have recently developed technology that can regenerate plastics from olefin plastic waste, and its implementation is desired in the near future [6].

Landfilling is the least desirable form of plastics processing in terms of the circular economy, and the high landfilling ratio in the United States and Europe (especially Eastern and Southern European countries) is a major issue [7,8]. Landfilling is also a source of secondary damage such as groundwater pollution and ecosystem destruction. In developed countries, it is strongly desired to replace landfilling with recycling. In Japan and some European countries, the percentage of energy recovery is high. Energy recovery is used for power generation and alternatives to fossil resources in industry and is highly effective in reducing carbon dioxide emissions, making it a useful method for processing plastics. However, if producing recycled plastics is prioritized, mechanical and chemical recycling are more desirable processing methods as compared to energy recovery. By improving recycling capacity in developed countries, it is possible to reduce environmental costs by an estimated 3.2 billion euros annually [9].

There is no universal solution to plastic waste recycling because each method has advantages and disadvantages. While mechanical recycling methods using unused plastics, post-consumer plastic waste or post-industry plastic waste is ideal, it has many problems. The disadvantage of mechanical recycling is that the recycled plastics is inferior to virgin plastics in terms of material properties such as strength, smell, purity, and color [10]. The difficulty in recycling dirty plastic waste and composite materials necessitates pre-sorting of waste [11], which increases costs.

Chemical recycling routes involving pyrolysis [12] and gasification [13] are highly applicable to recycling dirty waste and composite materials, which are difficult to mechanically recycle. However, it has the disadvantage of generally requiring a large amount of energy, resulting in higher carbon dioxide emissions than mechanical recycling.

In addition to the approaches mentioned above, there is an increasing interest in biological recycling. One method is to biologically decompose petroleum-derived plastics using microorganisms [14]. Another is a method of spontaneous decomposition under specific natural conditions as some plastics are biodegradable [15]. Since biological recycling does not use a large amount of energy, depending on the decomposing conditions, it has the advantage that the environmental burden is small. As a representative example of the former, research on the biodegradation of polyethylene terephthalate (PET) is being enthusiastically conducted, but it has not yet reached the stage of industrialization. As for the latter, there is a method for degrading bioplastics and another for degrading petroleum resources, and some methods are being industrialized.

It is not a rudimental task to synthesize research trends in plastic recycling. However, with the rapid increase in plastic consumption and application every year, establishing appropriate recycling system of plastic waste is an urgent task, and considerable research efforts are being made to achieve this. When we retrieved papers on plastic with the query of (plastic* OR chemical*) AND (recycl* OR "circular economy") in Web of Science, we approximately have 56,000 papers as of 2021 and approximately 7700 new papers in a year.

The bibliometric method is effective for synthesizing a comprehensive overview of a wide research area. This is a method developed to analyze research areas based on published data [16]. Bibliometrics is the analytics of bibliographic records of documents like papers, patents, newspapers, web pages, etc. Bibliometric methods include statistics, supervised learning, natural language processing, and citation analysis. They analyzed bibliographic records like information on the authors, publication date, keywords, abstract, and references.

Some previous studies have used bibliometric methods related to plastic waste. de Sousa (2021) conducted a bibliometric analysis using the keywords (waste management AND plastic*) and reported that the pollution of water bodies by plastic waste, especially microplastics, is a serious problem. He also points out that resolving this pollution is a major challenge in realizing a circular economy [17]. However, this study did not analyze research areas or review emerging topics on plastic recycling methods. Tsai et al. (2020) analyzed municipal solid waste management using a bibliometric method, and identified five themes for future research: incineration, life cycle assessment, plastic waste, sorting solid waste, and sustainability [18]. There is no analysis of research trends in plastics recycling in this study. Armenise et al. (2021) analyzed plastic pyrolysis research using a bibliometric method, and pointed out that important research issues include improving the performance of catalysts, pyrolyzing mixtures of plastics and biomass, and designing reaction paths [19]. However, there is no analysis of plastic recycling methods other than pyrolysis research. Wang et al. (2022) conducted a bibliometric analysis of the impact of the COVID-19 pandemic on plastic pollution, pointing out that plastic pollution has become more serious since the pandemic and that research on plastic pollution in developing countries has become active [20]. They have not done any research on plastic recycling other than the impact of the pandemic. Sandanayake et al. (2020) analyzed the five categories of recycled waste (plastics, glass, fly ash, slag, and construction waste) using the bibliometric method. They reported that such recycled waste is effective as part of the raw material for concrete, but they have not studied how to use recycled plastic for purposes other than concrete [21]. In this way, few publications have attempted to analyze the overall picture of plastic recycling. This study systematically analyzes and clarify the overview of research on plastic recycling, and reviews the latest research trend, which has not been the subject of previous studies.

The research objectives of this review are to comprehensively ascertain the following aspects of plastic recycling research using bibliometric methods: (1) the research areas that exist, (2) the emerging research topics, (3) and the way in which global researchers studying plastic recycling can cooperate to resolve the issues relating plastic waste.

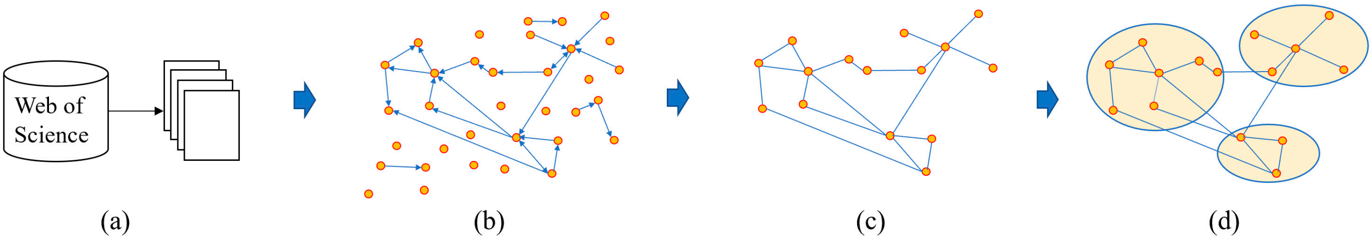
2. Materials and Methods

2.1. Data Collection

We collected bibliographic data from academic publications related to plastic recycle. We used data collected with the query (plastic* OR chemical*) AND (recycl* OR "circular economy"). The asterisk allows us to look for terms that begin with "plastic," "chemical," or "recycl" (e.g., recycle, recycles, recycled, recycling) in either the title, abstract, keywords or all fields. We retrieved bibliographic data with the above query from the Science Citation Index Expanded, the Social Sciences Citation Index, the Conference Proceedings Citation Index, and the Book Citation Index by using Web of Science. Data collection was carried out in February 2022. The number of publications collected for this review was 56,408.

2.2. Methods

We schematically show our methods in Figure 1. After acquiring relevant publications (Figure 1a), we created citation networks by treating the papers as nodes and the citations as links (Figure 1b). According to a 2009 study by Shibata et al., direct citation is the best approach for finding emerging trends [22]. Therefore, we also used the direct citation method. We removed irrelevant papers that were not connected to other papers in the largest component of the citation network. (Figure 1c). The component includes 35,519 papers (63.0%). We divided the network into clusters using the Newman's algorithm topological clustering method after obtaining the largest connected component (Figure 1d) [23]. Using this algorithm, we divided clusters into subclusters according to the rule of maximizing modularity, which has been used in previous bibliometric studies [24–28].



(a) (b) (c) (d)

Figure 1. Methodology overview. (**a**) Data acquisition from Web of science; (**b**) Creation of a citation network based on the citation relationships of papers; (**c**) Extraction of largest connected components; (**d**) Division of citation networks into clusters. In (**b**–**d**), each circle and line between circles represent a paper and a citation, respectively.

Modularity represents the strength of connections within a cluster. A high modularity means that the connections within a cluster are dense and the connections between clusters are sparse. It works well for citation clusters whose sizes are more than hundreds [29]. We characterized each cluster by analyzing the term frequency-inverse document frequency (tf-idf) [30,31]. However, a text-based approach is inferior to the citation-based approach to analyze corresponding relationships between papers [25]. Therefore, we used a text-based approach to complement the citation-based approach. We also calculated the ratio of the number of citations to those of papers in each cluster to assess relative importance among clusters. After the clusters were created, we named each cluster according to the content of the most cited papers within the cluster.

Plastic recycling research was divided into clusters that depend on their citation topology. We analyzed 21 clusters with more than 500 papers (total number of papers was 32,962, 92.8%), and we focused on six clusters related to plastic recycling. Since the query is very wide, 21 clusters contained those that are not related to plastic recycling (e.g., recycling of catalyst, chemical conversion of carbon dioxide, recycling of steel slag, crust, and mantle). The number of papers in the six remaining clusters related to plastic recycling was 13,248.

3. Results

3.1. Overview of Plastic Recycling Research

Table 1 summarizes the six clusters of plastic recycling. Cluster 1 is the largest cluster, and it is on plastic recycling in general. Cluster 2 is on waste electrical and electronic equipment (WEEE) and sorting of plastic waste. Cluster 2 has the second largest number of papers, but the number of papers has been increasing moderately in recent years. Cluster 1 and cluster 2 have relatively high citation per paper which implied significant scientific work and good knowledge sharing in these fields. Cluster 3 is on the use of plastic waste in the construction sector. Cluster 3 is an emerging cluster, which shows a younger average publication year (2016.8). Cluster 3 has the third largest number of papers, but the citation per paper ratio is relatively low (3.5), which implies less advanced knowledge sharing where the topic of research is quite specific. Cluster 4 focuses on chemical recycling of polyethylene terephthalate (PET). Cluster 4 has the highest citation per paper ratio (5.4) and is assumed to have good knowledge sharing because the average publication year is oldest (2013.9), and the research topic is more matured. Cluster 5, uses for wood-plastic composites, is tied with the oldest cluster (2013.9), and the citation/paper ratio is the lowest (2.7). Cluster 6 is on the recycling of fiber reinforced plastic (FRP) and is the youngest among the clusters (2017.0). Average publication years for those six clusters are approximately 2015, and most clusters have seen a significant increase in the number of papers since around 2015. The EU announced their Circular Economy Action Plan in December 2015, which may have triggered an increase in the number of papers. The increase in the number of papers in cluster 1, cluster 3, and cluster 6 are particularly remarkable.

Table 1. Information summary of six clusters. Tf-idf is a keyword that characterizes each cluster. "#" represents a number.

Cluster #	Cluster Name	Average Publication Year	# Papers	# Citation	Citation/Paper Ratio	Keywords
1	Plastic recycling	2015.2	4442	20,342	4.6	Plastic, pyrolysis, packaging, polyethylene, PET
2	Waste electrical and electronic equipment (WEEE) and sorting of plastic waste	2014.6	2287	10,771	4.7	PBDEs, plastic, WEEE, electronic, polybrominated
3	Use of plastic waste in the construction sector	2016.8	2023	7075	3.5	concrete, asphalt, aggregate, mortar, cement
4	Chemical recycling of polyethylene terephthalate (PET)	2013.9	1393	7455	5.4	PET, terephthalate, ethylene terephthalate, glycolysis, depolymerization
5	Use for wood-plastic composites	2013.9	1266	3458	2.7	composite, wood, fiber, plastic composite, wood plastic composite
6	Recycling of fiber reinforced plastic (FRP)	2017.0	1072	5627	5.2	vitrimers, epoxy, carbon fiber, CFRP, fiber

In the following sections, we conducted a detailed survey of each cluster. For this review, clusters with 2000 or more papers (cluster 1–3) were divided into subclusters, and clusters 4–6 were reviewed as they are.

3.2. Cluster 1: Plastic Recycling

Cluster 1 was classified into six subclusters. A summary of the six subclusters is shown in Table 2. Subcluster 1-1, recycling by pyrolysis, has the largest number of papers and the citation per paper ratio is also the largest (4.8), which makes this subcluster the central theme in cluster 1. Subcluster 1-2 which is on life cycle assessment (LCA) of plastic recycling is relatively young (2015.7). In recent years, in response to the growing social interest in recycling plastic waste, LCA research has been vigorously conducted to quantify the environmental impact of each recycling method from various angles. Subcluster 1-3, mechanical recycling, ranks third in number of papers but has only increased moderately in recent years. Subcluster 1-4, biodegradation of plastics, is the youngest among the six subclusters (2018.7). The citation per paper ratio is also high (3.9) and has shown increasing publication activity. Subclusters 1-5, bioplastics, is the second youngest of cluster 1 (2017.9). Subcluster 1-6 is on the recycling of polyvinyl chloride (PVC) and is rather old (2012.3).

Table 2. Summary of six subclusters in cluster 1. “#” represents a number.

Cluster #	Research Topic	Average Publication Year	# Papers	# Citation	Citation/Paper Ratio
1-1	Recycling by pyrolysis	2013.5	772	3736	4.8
1-2	life cycle assessment (LCA) of plastic recycling	2015.7	568	1839	3.2
1-3	Mechanical recycling	2009.9	547	1593	2.9
1-4	Biodegradation of plastics	2018.7	521	2028	3.9
1-5	Bioplastics	2017.9	396	885	2.2
1-6	Recycling of polyvinyl chloride (PVC)	2012.3	190	501	2.6

3.2.1. Subcluster 1-1: Recycling by Pyrolysis

The first subcluster is recycling by pyrolysis and thermochemical recycling. Plastics are polymers—repeating structures of carbon and hydrogen that can be broken down into either hydrocarbons such as fuels, monomers, or intermediate products in the chemical industry. This process is known as thermochemical recycling and is broadly classified into pyrolysis, gasification, depolymerization and upcycling [11]. Hereinafter, thermochemical recycling is referred to as pyrolysis in a broad sense. Mechanical recycling is an ideal recycling method in that it recycles plastics from plastic waste. However, it has the disadvantage that it is difficult to apply to the recycling of dirty plastic waste used for food containers and packaging, and plastic waste made of composite materials [3,32]. Thorough cleaning, separation, and sorting of plastic waste is required for mechanical recycling [11]. Pyrolysis has the advantage of being highly tolerant of dirty waste and composite materials. An inclusive approach of integrating mechanical recycling and recycling by pyrolysis may be the most effective approach to addressing plastic recycling challenges [11]. The global market size of pyrolysis technology was approximately 972.8 million USD in 2019, which is estimated to grow by 8.2% compound annual growth rate between 2020 and 2027 [33].

One of the most prominent research trends in pyrolysis worldwide is the production of fuel from plastic waste, with many commercialized plants already utilizing this method [34]. Considerable research has been conducted to optimize reaction conditions (e.g., temperature, residence time, pressure, equipment, process) and appropriate catalysts (e.g., cobalt, platinum, zeolite, aluminum chloride, organic matter) for producing fuel, depending on the type of plastic such as polyethylene (PE), polypropylene (PP), polystyrene (PS), and PET [3,12,35–38]. There is also research on recycling oils such as lubricants as products other than fuel [32]. Recently, research on upcycling to produce raw chemical materials other than fuel is increasing [36,39,40], and a commercial plant that produces ammonia and carbon monoxide by gasification is also emerging [41]. PS and PET are relatively easy to monomerize, and research is being conducted to regenerate PS and PET from these wastes [11,42,43]. There are commercialized plants for PET [43]. Olefin plastics waste such as PE and PP are difficult to monomerize, and there are not many publications on this subject. However, a pilot project has recently begun mixing oil produced from olefin waste plastic with fossil resources and introducing them into the petroleum refining process, regenerating them into plastics instead of processing them for fuel [6].

The major challenge of pyrolysis is its high energy consumption [36,44], which can be an obstacle to achieving carbon neutrality. To mitigate this, it is important to develop a highly efficient and inexpensive catalyst to realize thermal decomposition at lower temperatures [11]. To develop new catalysts and design highly efficient reaction systems, detailed research on the conversion mechanism of plastic waste is required [45]. In addition, recycled raw chemical materials and recycled plastics generated from plastic waste are often more expensive than virgin materials produced from fossil fuels and other raw materials, which constitutes a major research challenge [11,46].

3.2.2. Subcluster 1-2: LCA of Plastic Recycling

The second subcluster is LCA of plastic recycling. Plastic waste treatment methods are broadly divided into mechanical recycling, chemical recycling (which includes feedstock recycling), biological recycling, energy recovery, and landfill. There are several previous studies of LCA on these treatment methods from various viewpoints such as global warming potential, acidification potential, rich nutrition potential, energy consumption efficiency, and ecosystem impact potential [47–51]. The specifics of waste and treatment processes differ depending on each country and region, such as collection, transportation, and sorting methods, and the LCA of each previous study also reflects these regional characteristics [48,52]. In addition, the method of setting system boundaries also differs with study, and for these reasons it is difficult to compare and summarize the results of each study in a strict sense [52].

Regardless of the limitations, there are trends commonly observed in the literature. Energy recovery is mainly used as a benchmark in analysis, and the differences from other treatment methods are overviewed as global warming potential (GWP), energy use (EN), residual solid waste for landfill (SW), acidification potential (AP), and eutrophication potential (EP). Comparing mechanical recycling and energy recovery, mechanical recycling has less impact on the environment in GWP, EN, SW, AP, and EP [47,50]. Comparing chemical recycle and energy recovery, chemical recycle has a smaller environmental impact from the viewpoint of AP, EP, and EN, but energy recovery has a smaller environmental impact for GWP and SW [47]; though the impact in GWP has been found to be smaller for chemical recycling in some studies [53,54]. Comparing mechanical and chemical recycling, mechanical recycling has lesser impact on the environment in GWP, AP, EP, and EN, but a higher impact in SW [47]. Comparing energy recovery and landfilling, energy recovery has a smaller environmental impact in AP, EP, and EN, but a larger impact in GWP [47,50,55]. Mechanical recycling is considered the most desirable option when considering only LCA [47,48,50–53,55], but chemical recycling is considered the second most desirable when considering the other analyses mentioned [52], except for SW [51,55]. Mechanical recycling also has the disadvantage that it cannot recycle dirty plastic waste and composite waste. In addition, mechanical recycling always generates residue which usually cannot be recycled. Therefore, mechanical and chemical recycling should not be compared using LCA alone, and using these two recycling methods in a complementary manner is most desirable [55,56]. Energy recovery is considered the desirable option for treating waste for which mechanical and chemical recycling are not suitable. Despite their disadvantages, these processing methods have less environmental impact than non-recycling [51,52,57]. Deterioration of plastic waste in landfills is approximately 1–5% over 100 years, leading to potential air and groundwater emissions [52,58]. Over longer time periods, landfilling is likely to have a greater environmental impact and is considered to be the least desirable processing option [47,55].

Improvements in recycling technology may change these outcomes. For example, if automatic waste sorting methods become more sophisticated, mechanical recycling will be more advantageous. Similarly, technological progress into chemical recycling is increasing, for example there is rapid development in using pyrolysis at lower temperatures. Depending on the outcome of this work, the environmental impact of chemical recycling will be small. Therefore, it is necessary to promote the development of such techniques [55] and to continue research into LCA to monitor its progress [56]. In order to be able to compare and summarize LCA research results across methods, the system boundary conditions should be standardized as much as possible [56]. Furthermore, there are many LCA studies on post-consumer waste, but there are few LCA studies on post-industry waste, and further investigation is necessary.

3.2.3. Subcluster 1-3: Mechanical Recycling

The third subcluster is mechanical recycling. Mechanical recycling is the reprocessing of plastic waste into raw materials and products using physical methods [59]. Mechanical recycling requires a series of processing and preparation steps [60]. The first stages of the recycling process are collecting, sorting, shredding, milling, washing and drying. The waste is then shaped into recycled plastic pellets, powder, or flakes [3,59]. In the second step, the plastic pellets, powders, or flakes are melted and processed into the final product by resin molding. Resin molding methods are extrusion molding, injection molding, blow molding, vacuum molding, inflation molding, and melt spinning [59].

The advantage of mechanical recycling is that it is suitable for decentralized installations. Mechanical recycling plants are simple, inexpensive, and require less energy and resources to operate as compared to chemical recycling [10]. However, reprocessed plastics are inferior to virgin plastics in terms of material properties such as strength, smell, purity, and color [10].

Compared to virgin plastics, mechanically recycled plastics are divided into three groups based on the quality and price [10]. The first group is recycled material of the lowest quality. In this group, waste disposal is more important than maintaining the quality of recycled materials. Recycling businesses can earn money both by taking orders for recycling processes and by selling recycled products. Recycled products of this group are used for non-functional applications such as simple fillers. The second group consists of recycled products whose quality is not so low, but whose price is lower than that of virgin plastic in relation to its performance. There is a limit to how much the quality of recycled products can be improved while maintaining price competitiveness. Lower prices for virgin plastics will also constrain the price competitiveness of the recycled products in this group. The third group is recycled products that have the same high performance as virgin plastics but are priced higher than virgin plastics. These are used in high-performance fields such as food packaging [10]. Recycled bottles from discarded PET bottles are a typical example. If "Design for Recycling" (designing virgin plastics to be easy to recycle) is steadily introduced into the plastics industry and sorting technologies of plastic waste mixtures become more sophisticated, several problems in groups 2 and 3 can be considerably improved, reducing the cost of processing recycled products [61].

Among the disadvantages of mechanical recycling, the deterioration of strength is significant. These limitations are caused by degradation of the polymer's molecular structure due to shear during the extrusion process at high temperatures and pressures [59]. Degradation mechanisms vary by polymer type, but changes in polymer chain length and mechanical properties are common challenges [5]. The term "downcycling" is used as a comprehensive term to describe the deterioration of material quality after recycling [62]. Antioxidants, chain extenders, blending technologies, fillers, and polymerizers are used to prevent deterioration in mechanical recycling [5,10,59,63]. For example, stabilizers are frequently used to control thermo-oxidative degradation during the melting process of PE and to maintain the quality of recycled products [64]. Stabilizers are also used in PP to prevent aggressive degradation of the polymer chains during the extrusion process. These suppression mechanisms are common [5,65]. Stabilizers are commonly used in PVC to neutralize the generated hydrogen chloride and prevent degradation of the polymer chains [5,66,67]. PS is susceptible to produce harmful substances to human health and the quality of the product. Unlike other packaging polymers, the use of fillers can strengthen the polymer structure of PS [68]. Deterioration of strength in mechanical recycling is a major problem, and additional work to develop novel low-cost and effective additives is required [5,63].

3.2.4. Subcluster 1-4: Biodegradation of Plastics

The fourth subcluster is the biodegradation of plastics. Biodegradation is the biological breakdown of materials with the help of microorganisms [69]. This process is an environmentally friendly cycle that converts materials into carbon dioxide, methane, and salts through microbial metabolism [70]. Because plastics are solid polymers linked by covalent bonds, they degrade slowly in the natural world and have a long lifespan [71]. In recent years, research on biodegradation has been vigorously pursued, but a practical system capable of biodegrading existing plastics produced from fossil resources has not yet been developed [71].

Existing fossil-based plastics can be divided into the following three categories based on biodegradation mechanism [72]. The first is the category of polymers with a carbon skeleton such as PE, PS, PP, and PVC. The second is the category of polymers such as PET with ester-bonded backbones and side chains. The third is a polymer with a hetero/carbonate (urethane) bond such as polyurethane (PU).

Using PE as an example of the first category, PE is biodegraded in four stages: carbonyl group formation [72], conversion to carboxylic acid [73], hydrolysis or fragmentation [74], and microbial metabolism [75], though the detailed biodegradation flow is still unclear [72]. Although many studies have reported that carbon-skeletal plastics are degraded by various microorganisms, investigations should be conducted to identify the depolymerizing enzymes that are key to the biodegradation process. Once identified, biodegradation research results can be applied industrially [76].

As an example of the second category, studies on the biodegradation mechanism of PET have mainly focused on bacteria that can digest PET and their functional enzymes. In 2016, Yoshida et al. reported the discovery of a bacterial strain called Ideonella sakaiensis 201-F6 that secretes two enzymes that hydrolyze PET (PETase and MHETase) [77]. This finding has stimulated researchers in generating significant progress and resulting in the rapid evolution of structural, kinetics, engineering, and evolution studies [71]. For example, a thermophilic hydrolase has been identified that is thermally stable at 70 degrees Celsius (°C) [78]. This temperature is close to the glass transition temperature of PET, which is advantageous for PET decomposition. The bacterium Pseudomonas putida has been shown to enzymatically hydrolyze PET to produce polyhydroxyalkanoate (PHA), a raw material for surfactants [72]. An enzyme produced by the strain HR29 has recently been shown to be the most robust method for hydrolyzing PET waste due to its excellent activity and thermal stability [79]. Proposals have also been made for a biodegradation system for PET waste in a saltwater-based environment by using eukaryotic microalgae instead of bacteria [80].

In the third category, the urethane bond of PU is hydrolyzed during biodegradation [81]. It has been reported that fungi and bacteria can break down polyester-PU blends with the help of enzymes that can hydrolyze ester bonds [82].

While biodegradation systems for existing fossil resource-derived plastics have not been put to practical use, biodegradable plastics made from fossil resources have been put to practical use. These are mainly used in combination with starch and other bioplastics. The biodegradability and mechanical properties of the combined compounds improve the performance of starch and other bioplastics [69].

Biodegradable polymers from bioplastics are also being industrialized. Polylactic acid (PLA), a biodegradable bioplastic, has inferior mechanical/barrier properties when compared to the existing petroleum-derived plastics. This limits the applications of PLA. Blending with tough polymers or plasticizing block copolymerization improves the vulnerability of PLA [83]. This increases strain at break but decreases tensile strength [84]. Despite these restrictions, PLA remains a promising biodegradable plastic. The mechanical properties of PLA are similar to those of PS, making it a potentially more sustainable alternative [83].

Biodegradable PHAs have better mechanical/barrier properties than PLA, but only account for 1.4% of the bioplastics market. [85]. However, its production level is projected to quadruple by 2023 [86]. The disadvantage of PHA is its high production cost [87]. PHA has the potential to substitute PET in bottle applications due to its biodegradability and outstanding barrier properties [88].

By further improving the applicability of microorganisms, it is possible to develop microbial cell factories that artificially control the biodegradation of plastic waste. This will require research to elucidate the biodegradation mechanisms of various types of plastic waste and to identify and manipulate the optimal microbes [72]. A groundbreaking biodegradation project is about to be put to practical use. This project will gasify municipal waste including plastic waste, remove impurities, and then biodegrade with microorganisms to produce ethyl alcohol [89], and finally produce plastics by processing ethyl alcohol [90]. We expect that plastic waste treatment will advance considerably if innovative efforts are made using new biodegradation technologies.

3.2.5. Subcluster 1-5: Bioplastics

The fifth subcluster is bioplastics. Research on bioplastics has increased rapidly in recent years due to the serious problems of plastic pollution and to move away from fossil resources. However, the definition of bioplastics varies from study to study [69,83]. There are three dominant definitions. The first is that bioplastics are polymers derived from renewable resources and materials or synthesized by microbial metabolism [72]. Simply stated, these are produced from biomass, and hereinafter will be referred to as bio-based. The second definition is that in addition to bio-based polymers, all biodegradable polymers are called bioplastics, including polymers derived from fossil resources [91]. The third definition is that only polymers that are bio-based and biodegradable are called bioplastic [69,83]. This excludes bio-based non-biodegradable polymers and biodegradable polymers derived from petroleum resources. Disagreement among researchers may have resulted in consumer confusion. A survey in Australia reported that consumers wanted many of the products they consume to be biodegradable, and 62% of the people surveyed dumped bioplastics in miscellaneous waste bins [92]. Australian consumers mistakenly believe that all bioplastics are biodegradable. In this study, we adopt the first definition and divide bio-based plastics into non-biodegradable and biodegradable polymers. We review them mainly in terms of recyclability.

Since bioplastics are derived from non-fossil resources such as plants that absorb carbon dioxide, they have the advantage of being generally carbon-neutral even if bioplastic waste is energy recovered or incinerated [83,93]. However, energy recovery is not a desirable treatment because it does not take advantage of the biodegradability of many types of bioplastics [83].

Examples of non-biodegradable bioplastics are bio-PET and bio-PE. Some beverage companies, such as Coca-Cola, Pepsi, and Nestle, already sell some beverages partially (30%) in bio-PET bottles [94]. As a method of producing bio-PET, its precursor, terephthalic acid (TA), is derived from fossil resources, but since the monomers necessary for production can be obtained from renewable resources, they can be called bio-based PET [95]. In most of the bio-PET currently produced, only one of its monomers, ethylene glycol (EG), is obtained from biomass, and bio-PET is partially bio-based in that respect. Due to technical problems, TA can only be produced from fossil resources [96]. Extracting lignocellulosic biomass from forest residues to produce bio-TA could be a solution for manufacturing 100% bio-based PET [97,98].

Theoretically, bio-PE can be produced utilizing the existing petrochemical-based PE plants [99]. Bio-PET waste with low organic waste contamination should preferably be mechanically recycled. It is appropriate to recover low-grade waste through chemical recycling. Due to the strong interaction between cellulose and PE, mixing PE with lignocellulosic waste prior to pyrolysis results in an efficient reaction [100].

Bio-PET and bio-PE have the same properties and characteristics as PET and PE derived from fossil resources, and the same recycling method and equipment can be utilized [93]. Therefore, it is not necessary to separate bioplastic waste and fossil resource-derived PET waste for recycling, which is a great practical advantage. Conversely, there are challenges when attempting to recycle biodegradable bioplastics with other plastic wastes. Especially in the case of mechanical recycling, it is necessary to sort and separate only biodegradable bioplastics from other wastes [93].

PLA, a biodegradable bioplastic, is suitable for food packaging because it is permeable to water, but it is also used to some extent for bottles. Trying to separate PLA bottles from PET bottles is difficult because both materials are transparent and similar in appearance [93]. In order to properly recycle PET, it is important to separate PLA from PET [93]. When PLA is mixed in PET at a concentration of 2–5%, the PET clumps and sticks to the walls inside the apparatus. As little as 0.1% PLA will make recycled PET opaque, and if the PLA content exceeds 0.3%, the recycled PET will turn yellow [101].

In principle, PLA and PHA waste can be mechanically and chemically recycled when they are sorted from other wastes, but it is questionable to what extent such sorting can be achieved at the site of waste collection. If these wastes are separated, they can be composted at the household or industrial level [93]. However, in household-level composting, care must be taken that if the composting system is not well managed, it becomes anaerobic and generates methane, which is harmful to the environment. It should also be noted that biodegradability and compostability are not always the same. A biodegradable polymer is one that can be biodegraded, but without a time limit. Compostable means that the polymer is degraded within the composting time limit and mineralization is initiated in time for food waste decomposition. Only compostable polymers do not produce materials with unknown environmental impacts [93]. From a waste management perspective, a reasonably short time required for biodegradation of the waste is a prerequisite for proper treatment [102].

Global bioplastics production in 2018 was 2.11 million tons (MT) and is projected to reach 2.62 MT by 2023 [85]. Despite this rapid market growth, bioplastics still account for less than 1% of the total plastic production [103]. While bioplastics are environmentally friendly, they are more expensive to manufacture and have inferior mechanical properties compared to existing plastics. Biodegradable plastics also have problems such as being decomposable by a limited number of microorganisms and the inability to control the environmental conditions and speed of biodegradation. Further research is required to resolve these issues [104,105].

3.2.6. Subcluster 1-6: Recycling of PVC

The sixth subcluster is PVC recycling. PVC is one of the most used thermoplastics materials [106]. The unique properties of PVC, high performance, low cost, and combined with a wide range of applications by a variety of processing conditions and methdologies, have made PVC a universal polymer [107–109]. PVC is used in a variety of short-life products such as packaging materials used in foods, cleansing materials, beverage packaging bottles, textiles, medical devices, etc. In addition, PVC is also used in long-life products such as pipes, flooring, window frames, wallpaper, cable insulation, and roofing sheets [110]. In recent years, PVC waste has rapidly increased; therefore, effective treatment has become more and more important [66,107]. Long-life PVC products have a long-time lag between use and waste discharge, but eventually become waste [54,108,111,112]. As a result, it should be noted that the amount of waste will increase rapidly in the future as the end of the life expectancy of long-life PVC products approaches [113].

The presence of chlorine is what characterizes the structure and predominant properties of PVC, but the inclusion of chlorine is what makes PVC processing difficult. There are difficult challenges to overcome, such as hydrogen chloride generation in the process of PVC treatment, which damages the equipment, and generation of harmful substances in the energy recovery and landfill processes [107,114,115]. Currently, the realistic recycling method for PVC is limited to mechanical recycling, and only a small percentage of PVC waste is recycled worldwide [110].

Post-consumer PVC waste is mixed with other plastic waste, and there remains the challenge of sorting; therefore, it is difficult to establish economical and efficient mechanical recycling [114]. In fact, products mechanically recycled from mixed waste materials have low mechanical properties and little applicability [116]. Additionally, when waste plastic mixtures with PVC are chemically recycled, chlorine must be removed as much as possible in the recycling process [110]. To avoid such restrictions on mechanical recycling, research on techniques for chemical recycling of PVC and research on chlorine removal methods has vigorously pursued in recent years [117,118].

Techniques for separating with different polymer densities have also been studied [110]. One example is a method using a liquid cyclone based on the principle of sorting by centrifugal force. The problem is that the specific gravities of PVC and PET are very similar, so they cannot be separated solely by density. For automatic separation of PVC and PET, it is necessary to add a melt filtration system. PVC and PET can be separated at a temperature of 204 °C, which is below the melting point of PET. Higher temperatures are unsuitable, as PVC deteriorates at the melting temperature of PET [116,119].

Another method of physical separation is based on various spectroscopies [66,120–122]. For example, X-ray fluorescence can be used to detect characteristic backscattering from chlorine atoms in PVC. However, since X-rays reflected from chlorine atoms are low in energy and cannot pass through paper labels often present on PVC waste, alternative methods, such as laser-induced plasma spectroscopy, have recently been proposed. Laser-based spectroscopy, however, has the disadvantage of being costly.

Electrostatic separation has been identified recently as a potential alternative to spectroscopic detection, which allows the separation of mixed plastics using a friction electrostatic process [123–125]. Different plastics can be either positively or negatively charged due to different work functions, and and electric fields can be used to separate these charged plastics.

By adopting both low-temperature dechlorination and mechanochemical treatment in base hydrolysis of PVC, a product with a low chlorinated compound content can be obtained. The resultant product does not form toxic chlorination-inducing compounds. Replacing the water-soluble medium with an organic solvent and chlorine can significantly reduce the temperature and time of the process [126–130].

The main advantage of hydrothermal treatment of PVC in subcritical water is that no chlorination-inducing compounds are produced. This is because the chlorine released from the PVC is converted into fully water-soluble hydrogen chloride. Research into this field is ongoing [131–138].

Dechlorination by catalytic hydrogenation is an environmentally friendly approach for the removal of organochlorines from chlorinated compounds. The main advantage of hydrodechlorination is that the presence of hydrogen effectively removes organic chlorines, greatly improving the product quality [139–142].

Gasification is the conversion of solid or liquid organic compounds into flammable gases by heating to high temperatures (1000–2000 °C) in the presence of oxidizing agents. Gasification of PVC waste in air, steam, carbon monoxide, carbon dioxide, and hydrogen make it possible to prepare hydrogen-rich gas with low organic chlorine content that can be used for power generation [111,143–157].

Practical recycling methods for PVC are currently limited to mechanical recycling; however, effective and economical pre-sorting is a challenge. Many chemical recycling methods have been proposed for the recycling and detoxification of PVC, but they have not reached the level of industrialization. Future research must focus on PVC preselection and chemical recycling to solve these problems.

3.3. Cluster 2: WEEE and Sorting of Plastic Waste

Cluster 2 was classified into four subclusters. A summary of the four subclusters is shown in Table 3. Subcluster 2-1, recycling of WEEE, includes general topics in the cluster and has the largest number of papers. WEEE is rapidly increasing in both developed and developing countries, and its proper disposal has become a major social problem. As will be discussed later, pre-sorting methods for plastics containing brominated flame retardants and safe treatment methods for chemical recycling are key issues in the recycling of plastics from WEEE. The presence of such challenging research issues and the high level of public interest are thought to be the factors behind the rapid increase in the number of papers published in this field.

Table 3. Summary of four subclusters in cluster 2. "#" represents a number.

Cluster #	Research Topic	Average Publication Year	# Papers	# Citation	Citation/Paper Ratio
2-1	Recycling of WEEE	2014.8	340	1283	3.8
2-2	Spectroscopy sorting	2014.1	260	977	3.8
2-3	Flotation separation	2014.3	187	1292	6.9
2-4	Electrostatic separation	2011.2	141	546	3.9

Subclusters 2-2, 2-3, and 2-4 are research topics on pre-sorting methods required before recycling plastic waste, which are important in both mechanical, chemical, and electrical and electronic equipment (EEE) waste recycling. Subcluster 2-2, spectroscopy sorting, has the second largest number of papers. Various methods have been proposed for spectroscopy sorting. Spectroscopy can handle many kinds of plastic waste, which may help explain the increase of recent publications. Subcluster 2-3, flotation separation, has the highest citation per paper ratio (6.9). Flotation separation is a technology that uses the hydrophobicity and wettability of plastics to separate them, and the principle is relatively simple. Subclusters 2-4 focus on electrostatic separation, which is an economically efficient way to separate plastic waste due to the relatively simple equipment required. Additionally, unlike flotation separation, there is no issue of wastewater treatment. However, subcluster 2-4 is a mature research field (2011.2), which seems to indicate that the flotation separation technology has reached the stage of practical application. The subclusters of cluster 2 generally have older average years of publication and a higher citation per paper ratio than the subclusters of cluster 1, suggesting that cluster 2 is a research topic that has attracted attention for a long time relative to cluster 1.

3.3.1. Subcluster 2-1: Recycling of WEEE

The first subcluster is the recycling of WEEE. Plastic from WEEE is increasing rapidly, including in developing countries, and potential treatment of WEEE is being heavily studied. WEEE is made up of a variety of recyclable components such as iron and non-iron materials, glass, and plastics. Plastic parts make up 10–30% by weight of WEEE and are the second largest component after iron [158]. A variety of plastics are used for EEE, of which acrylonitrile-butadiene-styrene (ABS), high impact polystyrene (HIPS), and PS are the most commonly used polymers. Flame retardants (FR) are used in EEE plastics to meet fire regulations. FR are chemicals added to the polymer structure to suppress ignition and reduce flammability of products [159]. FR pose a major challenge in the treatment of EEE plastic waste. A wide variety of chemicals are used as FR, including nonorganic and organic compounds. There are two types of organic FR: phosphorus-based FR and halogen-based FR. Halogen-based FR includes brominated FR (BFR) and chlorinated FR [160]. Approximately 30% of the plastics used for EEE contain FR [161]. The term BFR is a generic term meaning bromine-loaded FR. BFR include compounds such as aliphatic and aromatic compounds There are more than 75 types of BFR, and the main difference between them is the position of the bromine atom in the chemical structure [162].

Halogen-based FR are known to be problematic, and BFR is especially widely used. In particular, BFR have been found to be harmful to ecosystems and humans, accumulating in the body [162]. For this reason, some chemical substances in BFR are prohibited from being manufactured or used by international treaties [162–164]. These banned BFR (hereinafter referred to as legacy additives) are no longer used in new EEE; however, older EEE still contain legacy additives [164]. Due to the unregulated recycling of old EEE, BFR degrade into hazardous low-molecular-weight compounds in landfills. This can be a source of BFR leaking into the environment [165]. Hazardous BFR waste is transported both legally and illegally to areas where labor costs are low. Although cross-border movement of plastics containing legacy additives to developing countries is prohibited, due to changes in export declarations and inadequate cross-border management, these hazardous substances are still being carried to areas where waste management systems are less developed [164]. As much as 1818 kg of harmful brominated low-molecular-weight compounds are released into the environment every year around the world, especially in disposal sites in Asia [166]. In EEE, the utmost care must be taken when processing BFR-containing plastics [164,167].

Mechanical recycling is the most desirable method for treating EEE plastic waste, and most of the recycling currently performed is mechanical recycling [167,168]. Prior separation of BRF-containing plastics is required for mechanical recycling, which increases the processing cost. The most common automated sorting method is sorting by specific gravity [169].

A sorting method using infrared rays or X-rays has been extensively studied in recent years [170–174]. For mechanical recycling, it is also necessary to presort by type of plastic such as ABS, HIPS, and PS, which increases the processing cost. For this reason, there are many studies that mechanically recycle the blended plastics, which are often used in EEE, without sorting them [171,172,175–177].

Research on BFR chemical recycling can be broadly divided into three methods [178]. One is a method of dehalogenating plastics before pyrolyzing. The method of first dehalogenating and then decomposing the plastics is called the two-stage pyrolysis method [178–180]. The second is a method of simultaneously performing dehalogenation and thermal decomposition [178,181,182]. The third method is to first pyrolyze the plastics and then decontaminate the pyrolysized oil [178,183,184].

A phosphorus-based FR is used as a non-halogen-based FR; however, it has been pointed out that phosphorus-based FR is also harmful to the human body, and no safe and versatile FR has thus far been found. For this reason, it is assumed that BFR will continue to be used as the main FR, and further research on safe, economical, and environmentally friendly recycling methods for BFR is needed.

3.3.2. Subcluster 2-2: Spectroscopy Sorting

The second subcluster is spectroscopy sorting (SS). SS utilizes sensors to detect the presence and location of recyclable plastics within the waste and sorts the detected recyclables using automated machinery or robots [185]. SS can be broadly classified into three types. The first is spectral imaging-based sorting, the second is X-ray based sorting, and the third is laser-induced breakdown spectroscopy (LIBS) [185].

Spectral imaging consists of image processing technologies and spectral reflectance measurement [186]. There is considerable research in this area on near infrared radiation (NIR), visual image spectroscopy (VIS), and hyperspectral imaging (HSI). In spectroscopy-based techniques, light of a given frequency is projected onto the plastic waste. Due to the interaction between light and plastics, different types of plastics reflect different wavelengths of light. Various sensors, such as NIR sensors, read the reflected light to identify the type of plastics, which is then sorted [185]. A technique was developed to identify PP in mixed waste using VIS [187]. HSI-based approaches aim to recover high-purity PP and PE in the NIR range (1000–1700 nm) [188]. A method for rapid classification of PET, PE, PP, PS, and PLA was developed by combining NIR spectroscopy and independent component analysis [189]. However, this technique cannot effectively detect black polymer materials due to its high absorption.

As X-ray images can be captured within milliseconds, X-ray based sorting is fast [185]. A high intensity X-ray beam is used in the imaging module. X-rays penetrate the material and hit the bottom detector after being absorbed by the material. Information about the atomic density of materials is obtained by analysis of the detected radiation. In the sorting of plastic waste, X-ray fluorescence (XRF) type detectors are used. In XRF, an external laser source excites individual atoms, emitting X-ray photons. Although it has been pointed out that this technology can only be applied when recovering PVC from plastics such as PET and PP [190], it is an extremely important technology for recycling PVC. Energy dispersive X-ray fluorescence (EDXRF) has been proposed as an innovative technology that can improve the accuracy of PP sorting. EDXRF can recognize black polymer and surface contamination [191].

LIBS is considered to be the most promising novel elemental analysis method [192–194]. LIBS typically consists of an intense pulsed laser to generate plasma, an optical system to focus and collect the light, and a spectrometer [192]. It is reported that LIBS can distinguish various plastics such as PE, PP, PS, PET, and even PVC [195]. In addition, it has been reported that bromine can be detected [193], and attention is focused on its application to the separation and recycling of WEEE.

SS is one of the most widely used sorting methods at recycling sites; however, there are several types of plastics, and the shapes and characteristics of plastic waste are diverse. SS is a technology that has the potential to further improve the accuracy and economic efficiency of sorting a wide variety of plastic wastes.

3.3.3. Subcluster 2-3: Flotation Separation

The third subcluster is flotation separation. Flotation separation is a technology that uses the hydrophobicity of plastics to sort plastics based on the difference in their ability to float on water. The principle is to selectively hydrophobize the plastic surface to control its contact with air bubbles. The wettability between different plastics must be sufficiently different for selective contact with air bubbles [196]. There are two main approaches to flotation separation [197]. One is to selectively convert hydrophobic plastics to be wettable. Alternatively, wettable plastics can be selectively converted to be hydrophobic [196]. The main difficulty in plastic flotation is finding an effective way to selectively wet the plastics. For example, there are methods of reducing liquid-vapor surface tension (referred to as gamma flotation), methods of changing chemical conditions, and methods of surface treatment [198]. Flotation separation is categorized into four groups: gamma flotation, adsorption of reagents, surface modification, and physical regulation [196].

The first category, gamma flotation, can be achieved by lowering the liquid surface tension to some value between the two plastics [196]. With the help of plasticizers, PVC can be rendered hydrophobic and successfully separated from PET [199]. Various wetting agents such as sodium dodecyl sulfate have been investigated for the flotation separation of PET, PVC, polycarbonate (PC), and PS [200]. Gamma flotation often does not work well for sorting two or more plastics [196].

The second category, adsorption of hydrophilic reagents, is a promising option to selectively reduce the hydrophobicity of plastics. Flotation reagents are used to selectively wet plastics surfaces and are called wetting agents. Suitable wetting agents are molecular groups that have the ability to adsorb to plastics surfaces and make them hydrophobic [196]. Studies on wetting agents suitable for plastic flotation include tannic acid [201], methylcellulose [202] and lignosulfonates [196].

The third category, physical techniques, can change the wettability of plastics surfaces, and this method is called surface modification or surface treatment. There are many studies on physical methods to increase the hydrophobicity of plastics surfaces, such as flame treatment [203], wet oxidation [196], ozonation [204], and plasma treatment [197]. Separation of PET from other plastics has been achieved on the bench scale by involving an alkaline treatment [205]. By administering an ammonia treatment, ABS and PC could be separated by flotation separation, and the maximum purities of ABS and PC using this method were 99.72% and 99.23%, respectively [206].

The fourth category is physical regulation. Unlike surface modification, physical regulation is based on essential physical properties and does not involve chemical reactions. Boiling treatment can effectively change the surface of ABS to be hydrophobic [207]. There are studies on the feasibility of separating chlorinated plastics from other plastic waste by selectively twisting PVC or PVC films [208].

A major challenge with flotation separation studies is that much of the work has been done on virgin plastics rather than plastic waste [196]. Compared to virgin plastics, plastic waste has differing properties, and the behavior of the flotation mechanism also differs. A better understanding of these differences is necessary.

3.3.4. Subcluster 2-4: Electrostatic Separation

The fourth subcluster is electrostatic separation. It is well known that two materials with different surface properties become charged when they come into contact. This is the tribocharging phenomenon, also known as contact electrification or frictional electrification [209]. Electrostatic separation is a method of separating two different plastics by charging them positively and negatively using the principle of tribocharging. Since plastics usually have different surface properties (i.e., effective surface work functions), electrostatic separation can ensure sorting of plastic waste [210]. Although it is difficult to accurately measure the work function of all plastic wastes with rough surfaces, electrostatic separation can be used to determine positive and negative charging only due to the relative work function differences between different plastic wastes [211].

PVC can be hazardous depending on how it is processed, making its sorting and recycling an important challenge. PVC, on the other hand, is the most highly negatively charged except for polytetrafluoroethylene and can be easily separated by electrostatic separation [212]. One of the advantages is that there is no need to worry about pollution from wastewater treatment, as water is not used for flotation separation. In addition, electrostatic separation requires a relatively simple apparatus; therefore, it is economical and can separate efficiently [210].

Plastic waste is usually pulverized to a suitable size, sieved, and the mixed particles are charged by a triboelectric charger. An electric field is then applied for deflection, and particles are then collected. In the device, there are mainly three types of collisions that affect the charge of the particles—contact between particles of different plastics, contact between particles of the same plastics, and contact between particles and the wall material of the device [213]. The polarity and magnitude of charge on plastic surfaces are affected by a combination of these three mechanisms, the most significant being contact between different types of plastics. In fact, tribocharging occurs only at a depth of as little as 30 nm on the plastics surface [214].

Humidity control is one challenge with electrostatic separation. In most cases, low humidity leads to high efficiency [210]. However, the performance of dehumidifiers in factories is generally limited, and it is difficult to maintain humidity below 20% [215]. In electrostatic separation, it is difficult to separate three or more types of plastics at once. A series of two or more repeated electrostatic separations can be a solution [211].

The mechanism of particle collision in the charging step still needs to be better understood. The ability to build a micro-level collision model to better understand the charging behavior of plastics particles will help control triboelectric charging efficiency. Several numerical models have been proposed, but there is much more that still needs to be investigated.

3.4. Cluster 3: Use of Plastic Waste in the Construction Sector

Cluster 3 is divided into two subclusters and both of them are relatively young. A summary of the two subclusters is shown in Table 4. Subcluster 3-1 is on the use of recycled plastics in concrete, and its citation per paper ratio (6.4) is higher than that of subcluster 3-2 (3.1). The advantage of this method is that a large amount of plastic waste can be treated when recycled plastics are available as aggregates for concrete, which may explain why research in this field is so vigorous. However, compared to the existing aggregates, mixing plastics into concrete deteriorates mechanical properties such as strength. It also reduces fluidity, making it difficult to handle at construction sites. There are several studies proposing solutions to these problems. Subcluster 3-2 is on use of recycled plastics in asphalt. Incorporation of recycled plastics into asphalt tends to improve the performance and durability of asphalt pavements and is being intensively researched for wider application on roads. Regarding the use of plastics in the construction sector, the use for asphalt is seemed to be a more feasible method than the use for concrete. It is thought that more researchers are interested in the use for concrete as it is a challenging topic that is difficult to apply to the real world.

Table 4. Summary of two subclusters in cluster 3. "#" represents a number.

Cluster #	Name of Cluster	Average Publication Year	# Papers	# Citation	Citation/Paper Ratio
3-1	Use of recycled plastics in concrete	2017.1	316	2028	6.4
3-2	Use of recycled plastics in asphalt	2017.5	236	734	3.1

3.4.1. Subcluster 3-1: Use of Recycled Plastics in Concrete

The first subcluster is the use of recycled plastics as raw material for concrete. Generally, concrete is mixed with aggregate (usually sand or gravel is used). By substituting plastics for a portion of sand and gravel, the plastics can be mixed into concrete as an aggregate at the concrete casting site. Mixing plastics into concrete as an aggregate suppresses heat generation and shrinkage when the concrete hardens, and helps prevent cracks. In addition, since cement paste and mortar, which are the main raw materials of concrete, are expensive, the amount of these expensive raw materials used can be reduced, and the construction cost can be suppressed by mixing plastics. Although much research has been done on the use of recycled plastics in concrete, there are few papers on their field applications. Demand for concrete is high around the world, and if recycled plastics can be used as aggregate, a large amount of plastic waste can be processed. For this reason, the existing body of research is large, and it is currently an active field of study [216–218]. As the proportion of plastics in concrete increases, mechanical properties such as compressive strength, flexural strength, tensile strength, and elastic modulus decrease [216]. Replacing 20% of the existing aggregate with plastics reduces compressive strength by 72%. However, a 5% replacement results in only a 23% decrease in compressive strength [219]. Substituting with PET at a rate of 15% decreased flexural strength by 16% for pellet-type PET and by 60% for thin-form PET [220]. When fine aggregate is replaced by 10%, the tensile strength decreases by 8.7%, and when it is replaced by 20%, the tensile strength decreases by 54% [221]. Several studies have reported that as the content of plastics increases, the ultrasonic pulse velocity (UPV), which reflects the quality of concrete, also decreases [216]. The value of UPV decreases with increasing content of PVC in concrete. However, the reduction is less than 16% if the PVC replacement rate is up to 45%. Replacing up to 85% reduces the UPV value by 30% [222]. Utilizing plastics as aggregate reduces concrete slump (i.e., reduces flowability) and results in concrete that is difficult to handle on construction sites. Replacing 20% of the fine aggregate with plastics has been reported to reduce slump values by up to 50% [223]. These characteristics are thought to be due to the low density of plastics, irregular shapes and sizes, and sharp corners of recycled plastic fragments. Plastics do not mix well with the existing aggregates, and water absorption, permeability, and carbonation of concrete enhance with increasing plastic content, adversely affecting the concrete durability [216].

Concrete made with recycled plastics is inferior to existing concrete in many respects. However, it is expected that concrete containing plastics will be used for non-structural materials that do not require high strength and applications that do not require high durability. Possible applications include highway median strips, temporary structures, and general-purpose bricks and blocks (for example, riverbanks) [216,217]. Applications in concrete pavement and sports courts are also mentioned. Concrete with plastic aggregate has a high water absorption rate, which helps with the proper drainage of rainwater. The use of additives such as superplasticizers can increase the flexibility of plastic-containing concrete and potentially improve the workability of concrete, thus reducing the challenges at construction sites. Plastic-containing concrete has a lower density, but lighter concrete could open up new uses. Furthermore, the possibility of applying new additives may complement the mechanical properties of plastic-containing concrete [216].

In solving the plastic waste problem, the use of recycled plastics as aggregate for concrete has great potential. Several issues remain, including the improvement of mechanical properties, long-term behavior change of mechanical properties, improvement of durability, development of additives to compensate for these shortcomings, elucidation of the optimal shape and size of plastics to mitigate performance degradation, and heat insulation and sound insulation properties [216–218] There are many themes in this field, which will require extensive research to resolve the numerous problems identified.

3.4.2. Subcluster 3-2: Use of Recycled Plastics in Asphalt

The second subcluster is the utilization of recycled plastics in asphalt. Asphalt is a hydrocarbon containing material with chemical similarities to plastics. There is a consensus among researchers that recycled plastics, when properly blended with asphalt under optimal conditions, significantly improves the performance and longevity of asphalt pavements [224]. For example, ethylene vinyl acetate (EVA) is a class of polymers that modifies asphalt by forming a tough, rigid, three-dimensional network that resists deformation, and virgin EVA has been used in road construction for many years [225]. The use of recycled plastics for asphalt provides a solution to the problem of waste treatment, improves the performance and economic efficiency of asphalt pavement, and may lead to cost reduction in the long term [226]. For example, it has been reported that approximately 1 ton of asphalt can be saved by constructing a 1 km long road (3.75 m wide) with asphalt using recycled plastics [227]. For these reasons, there has been increasing research into the utilization of recycled plastics in asphalt.

There are two methods of paving with asphalt-containing plastics [228]. One is the dry method, in which plastics are incorporated into hot aggregates prior to the addition of binders. This method applies in most cases to hard plastic types with high melting points such as high-density polyethylene (HDPE) and PET. The hardness and stiffness of recycled plastics particles play a role similar to the fine aggregate that is the skeleton of the asphalt mixture and contributes to its integrity [228]. Another method is the wet method, which involves adding plastics directly to the asphalt binder as a modifier before mixing it with aggregate. Low melting point plastics such as low density PE (LDPE) and PP are suitable for this method.

Since the effects and characteristics of asphalt mixtures differ depending on the type of plastics used, research has been conducted according to the type of plastic. PET is mainly used in dry processes, and when used as an aggregate substitute for asphalt mixtures, it increases stiffness and improves both rutting and fatigue resistance [229]. Conversely, it has been reported that thermal cracking and moisture resistance are impaired [230]. PET-modified asphalt can weaken the bond between aggregates and asphalt in asphalt mixtures [231]. This is due to the high stability and inert nature of plastics, and it is recommended to add an oxidizing agent to activate the plastic surface [232]. HDPE is mainly used in dry processes due to its high density and high rigidity. PS is mainly used in dry process. PS increases asphalt hardness and improves rutting resistance [233]. The addition of PS hardens the asphalt mix and improves its resistance to moisture damage, although its impact on resistance to rutting and fatigue cracking is inconclusive.

LDPE is mainly used in wet processes, which require high shear rates and high temperatures to fully dissolve the LDPE into the asphalt. Although it is generally accepted that the addition of LDPE to asphalt improves rutting, fatigue, and moisture resistance, the results of thermal cracking resistance differ from study to study [228]. PP is mainly used in wet processes, and when added to asphalt, it increases hardness and contributes to improving rutting resistance. On the other hand, PP reduces the ductility of asphalt, resulting in more air voids. One study demonstrated that increased air voids resulted in impaired rutting resistance [234].

As experimental levels of research, asphalt mixed with recycled plastics are likely to be stiffer, resulting in overall improvements in viscosity, strength, rutting resistance, and fatigue resistance. However, verification of the performance of asphalt mixed with recycled plastics ultimately needs to be confirmed by field projects that use it for road paving. Several field projects have so far been carried out in India, South Africa, New Zealand, Australia, Canada, the United Kingdom, the United States, and other countries, with positive performance results in terms of workability, constructability, and sustainability [228]. However, few field projects have studied long-term performance, and it is not clear whether the performance of asphalt mixed with recycled plastics will be sustained over longer time periods. Further research into the long-term viability of plastic-containing asphalt, as well as the effects of asphalt mixtures on parameters such as fatigue resistance, thermal crack resistance, and moisture resistance is needed.

3.5. Cluster 4: Chemical Recycling of PET

In cluster 4, recycling methods (chemical, mechanical, etc.) are being studied for various plastics such as PET, PLA, PU, and PC. Among these, the number of studies on chemical recycling of PET is the largest, and the number of studies has been increasing in recent years. Therefore, here we focus on the chemical recycling of PET. Chemical recycling methods for PET are roughly divided into pyrolysis, hydrolysis, methanolysis, glycolysis, and aminolysis [235,236].

Hydrolysis converts PET chains into value-added products, especially TA and EG. There are three types of hydrolysis: neutral hydrolysis, oxidative hydrolysis, and alkaline hydrolysis [237]. A significant disadvantage of the hydrolysis process is the need for high temperatures of 200–250 and high pressures of up to 1.4–2 MPa. Acid hydrolysis also has the drawback of being corrosive and contaminating [235].

PET is also decomposed by high-temperature, high-pressure methanolysis, and EG and dimethyl terephthalate are obtained as main products [238]. A disadvantage of this process is that the reaction products such as alcohols, glycols and terephthalate derivatives have to be separated and purified, which is very expensive [238].

Glycolysis is considered to be the best recycling method for PET due to its short reaction time and wide temperature range. However, glycolysis produces undesirable products such as diethylene glycol and dimers of EG, which pose challenges in terms of separation and purification [236].

Aminolysis of PET produces diamides of TA and EG. The reaction is carried out in the temperature range of 20–100 °C by methods using amines and water, including methylamine, ethylamine, and ethanolamine [239]. Aminolysis has the advantage of being carried out under mild conditions, but the decomposition rate needs to be accelerated to reach the optimum reaction time [240].

Each chemical recycling method has advantages and disadvantages. Various research strategies should be pursued to develop more moderate reaction conditions for chemical recycling of PET and make the recycling method more feasible [235].

3.6. Cluster 5: Use for Wood-Plastics Composites

The fifth cluster is the use of plastic waste for wood-plastic composites (WPC). WPC is used for applications such as floor carpets, vases, waste baskets, lecture benches, and picnic tables. In recent years, the quantity of recycled plastics used for WPC has increased significantly in developed and developing countries [241]. WPC using recycled plastics have comparable or sometimes better flexural and tensile strengths than WPC using virgin plastics [242]. The absorption of water by WPC and the resulting thickness expansion are the most important properties when considering its end use in the natural environment [241]. In comparing the water absorption of WPC using recycled plastics with WPC using virgin plastics, studies have yielded mixed results. There are reports that the two are equivalent [243]. However, there are also contradicting reports that WPC using virgin plastics is superior [244] or that recycled plastic-based is superior [245].

In contrast to flexural strength, tensile strength, and hygroscopic properties, almost all studies show lower impact strength values for WPC made from recycled plastics than that made from virgin plastics [242]. Recycled plastics are available from various sources. Since the storage period and reprocessing conditions of recycled plastics also vary, different materials may exhibit different performance. Research on WPC using recycled plastics is still insufficient.

3.7. Cluster 6: Recycling of FRP

The sixth cluster is recycling of FRP. The plastics used in FRP are classified into thermoplastic resins (such as PE, PU, and PP) and thermosetting resins (epoxy, vinyl, etc.). Thermoplastic resins have a branched grain structure, soft properties, and the ability to retain their basic shape. Conversely, thermosetting resins are difficult to reshape or convert back to their monomers due to their tightly bound and crosslinked structures [241].

There are two methods of recycling FRP: mechanical and chemical [241]. In mechanical recycling, after sorting, cutting, shredding, and crushing, it is reduced in size in high-speed mills or rotary cutter sections into particles ranging from less than 50 µm to 10 mm. It is then separated into coarse (fibrous) and fine (powder) products using cyclones and sieves or by electrostatic separators. Coarse particles, which are usually mostly fibrous, can be reused in bulk molding compound composites. Microparticles, mostly powders, can be reused in bulk molding compound and sheet molding compound composites [246]. There are many studies attempting to determine the viability of using crushed FRP waste as concrete aggregate. Compressive strength, impact resistance, and bending strength are increased by mixing FRP waste into concrete. Increasing the ratio of mixed FRP waste to concrete enhances these properties.

Chemical recycling includes methods such as low temperature solvolysis, sub-supercritical solvolysis, glycolysis, and hydrolysis [247]. The primary purpose of chemical recycling of FRP is the collection and recycling of fibers rather than plastics, and several studies have been conducted on this process. Conversely, some studies report that plastics can also be decomposed and recovered in the process of chemical recycling. For example, ethylene oligomers depolymerized from thermosetting FRP can be reused as reactive components to produce advanced epoxy materials with high strength and elastic modulus [248]. Chemical recycling using zinc chloride and ethanol as catalysts yields decomposed matrix polymer (DMP) as an oligomer. With DMP as a reactive component, when 15% DMP by weight is added for FRP regeneration, the resulting crosslinked polymer maintains high strength and elastic modulus compared to the virgin polymer without DMP [249]. Glycolysis can break down epoxy resin into monomers, which can potentially be used to create chemicals [250].

Recycled FRP is beginning to be used in applications such as aircraft, automobiles, wind turbines, construction, household goods and sporting goods. Further research is needed to expand its application range and develop safe and economical recycling methods for various FRP wastes.

4. Discussion

In the earlier section, we have systematically reviewed the major fields and trends in plastic recycling research. In the following, the discussion is broadly divided into three perspectives. First, we discuss the characteristics and research trends of emerging clusters. Second, representative recycling methods are discussed from the perspective of sustainability and processing costs. Third, we discuss plastic recycling from a national and global perspective.

As a first perspective, we discuss the emerging research areas of plastic recycling. Table 5 lists the younger subcluster with an average publication year of 2017 and beyond.

There are two points common to each subcluster in Table 5. The first is that the research pursues a third recycling method that is neither mechanical nor chemical recycling, which are currently mainstream recycling methods. The average publication year for mechanical recycling is 2009.9, and the average publication year for chemical recycling (pyrolysis) is 2013.5, both of which are older than clusters listed in Table 5. The second is that the recycling methods studied in Table 5 emit little carbon dioxide. When biodegradable plastics are degraded by the action of microorganisms, the amount of carbon dioxide emitted is far less than mechanical or chemical recycling. Bioplastics are produced by renewable resources such as plants and by the action of microorganisms, and are naturally carbon-neutral materials [83,93]. Mixing recycled plastic into concrete or asphalt essentially emit less additional carbon dioxide.

Table 5. Summary of emerging research subcluster. "#" represents a number.

Cluster #	Research Topic	Average Publication Year	# Papers	# Citation	Citation/Paper Ratio
1-4	Biodegradation of plastics	2018.7	521	2028	3.9
1-5	Bioplastics	2017.9	396	885	2.2
3-1	Use of recycled plastics in concrete	2017.1	316	2028	6.4
3-2	Use of recycled plastics in asphalt	2017.5	236	734	3.1

There are remaining issues for these emerging research. For subcluster 1-4, "biodegradation of plastics," it will be necessary to elucidate the biodegradation mechanism according to the type of plastic, search for optimal microorganisms and operating conditions. For subcluster 1-5, "bioplastics," it is needed to develop less expensive production methods and bioplastics having improved mechanical properties. Regarding subcluster 3-1, "use of recycled plastics in concrete," improvement of strength and durability and development of additives to compensate for deterioration in properties are crucial issues. For subcluster 3-2, "use of recycled plastics in asphalt," it is important to demonstrate durability in actual use cases.

As a second perspective, we will discuss plastic methods from the view of sustainability and processing costs. Mechanical recycling is an ideal recycling method in that plastics can be recycled from plastics. However, mechanically recycled plastics generally deteriorate in quality and strength, and it is difficult to mechanically recycle the recycled plastics once again after use [10]. In particular, when post-consumer plastics are mechanically recycled, deterioration in quality and strength is remarkable. Mechanical recycling remains a challenge in terms of sustainability in that the plastics cannot be recycled over and over again. Since mechanical recycling can be produced in a relatively small-scale plant, the burden of equipment investment is light, and the amount of energy used is relatively small, the processing cost is lower than that of chemical and biological recycling [10]. User companies of recycled plastic are demanding lower prices than virgin plastic, and in this respect mechanical recycling is fully on a commercial basis.

Chemical recycling, such as pyrolysis, is more sustainable in that the same material can be recycled over and over again with little deterioration in quality or strength of the recycled plastics. However, with current technology, chemical recycling is more energy intensive [36,44] and emits more carbon dioxide than mechanical or biological recycling, which poses sustainability challenges in this regard. Chemical recycling requires the construction of a chemical plant, and the processing cost is generally high unless the plant is of a large production scale [11,46]. At present, there are many relatively small-scale chemical recycling plants, and processing cost tends to be higher than mechanical recycling.

Biological recycling can be broadly classified into two types. One method is to biologically decompose petroleum-derived plastics using microorganisms. Another is a method of spontaneous decomposition under specific natural conditions as some plastics are biodegradable. Both methods are superior from a sustainability point of view because they do not require a large amount of energy to decompose plastics [70]. The former is still at the stage of research on the optimum microorganisms and decomposition mechanism, and the processing cost is unknown because it has not been industrialized. As for the latter, since the recycling method for biodegradable plastics such as PLA is different from that for other plastics, it is premised on sorting by consumers, but it is difficult to distinguish the type of plastics just by looking at the appearance [93]. Therefore, there is a challenge from the viewpoint of sustainability in that recycling methods have not been established. Although biodegradable plastics have been industrialized, the market scale is still small [103], and processing cost remains at an extensively high level compared to general virgin plastics [87].

As a third perspective, plastic recycling will be divided into four issues and discussed from the two points of view: a national and global. The first issue is the discussion of countries with a large number of papers, comparing them with the plastic waste disposal situation, such as the recycling rate and landfill rate.

We collected data on current and past plastic waste and recycling research and analyzed the relationships amoung them, as well as quantified the number of papers by each country. Table 6 lists the countries in cluster 1 in descending order of the number of publications focused on each country, and describes the post-consumer waste amount, recycling rate, energy recovery rate, incineration rate, landfilling rate, environmental awareness, and population of each country.

Table 6. Various statistics on plastic waste management, relative publication frequency, and environmental awareness by country in Cluster 1. "#" represents a number.

Rank	Country	Share of Papers	Year	Waste Amount (mt)	Recycling	Energy Recovery	Incineration	Landfill	Untreated	Environmental Awareness	Population (m person)
1	China	12.4%	2020	130.30	28.0%	N.A.	32.0%	34.0%	6.0%	92%	1410.9
2	USA	11.1%	2018	35.68	8.7%	15.8%	0.0%	75.6%	<1.0%	71%	331.5
3	Italy	7.5%	2018	3.64	31.4%	32.8%	0.0%	35.8%	<1.0%	86%	59.4
4	Germany	6.9%	2018	5.32	38.6%	60.7%	0.0%	0.7%	<1.0%	81%	83.2
5	England	6.7%	2018	3.95	32.0%	45.7%	0.0%	22.4%	<1.0%	86%	67.2
6	Spain	6.2%	2018	2.57	41.9%	19.3%	0.0%	38.8%	<1.0%	85%	47.4
7	India	5.9%	2016	8.54	5–25%	N.A.	N.A.	N.A.	N.A.	82%	1380.0
8	Japan	5.2%	2020	4.10	23.2%	62.2%	12.0%	2.9%	<1.0%	56%	125.8
9	Brazil	4.7%	2018	7.90	2.2%	N.A.	N.A.	59.5%	24.4%	86%	212.6
10	Netherlands	3.2%	2018	0.94	33.7%	65.8%	0.0%	0.4%	<1.0%	73%	17.4

Data source: Data for the United States are taken from the US Environmental Protection Agency [7]. Data for European countries are taken from Plastics Europe [8]. The Japanese data quotes the research results of the Plastic Waste Management Institute [251]. Data for China are taken from a paper by Luan et al. [252]. Data for India are taken from Liang et al. [253] and Siddiqui et al. [254]. Data for Brazil are taken from a paper by Sandro et al. [255]. The Year column lists the year for which the data were obtained at these references. Environmental awareness indicates the percentage of people who want to buy products with as little plastic packaging as possible [256].

China, which ranks first in the number of publications, has a lower recycling rate than European countries listed and a higher landfill rate than the average of the top 10 countries. Six percent of China's plastic waste is not properly disposed and may have been released into the environment. The amount of post-consumer plastic waste per capita is second only to the United States. In these respects, China faces major challenges in realizing a circular economy. On the other hand, the public's environmental awareness is extremely high. To meet these challenges, in recent years, the Chinese government has tightened regulations on plastic waste and increased funding for research on how to deal with plastic waste, according to the head of a Chinese plastics industry association: ”China Plastics Processing Industry Association”. Such initiatives by the Chinese government are also thought to be a factor in boosting the number of papers published.

The United States, which ranks second in number of publications, has the highest landfill rate among the top 10 countries. Despite its high ranking in number of publications, such a high research capability seems not to be utilized for increasing their plastic recycling rate. Since the United States has a large land area, landfilling is considered to be the most economical disposal method. Per capita post-consumer plastic waste is the highest of the top 10 countries. Landfilling is the least favorable handling method in the circular economy, and in order to solve this problem the United States should be more enthusiastic about researching plastic recycling (e.g., researching into improving the economics of recycling to make it more economical than landfilling) than it is now. In addition, the results of advanced recycling research in the United States should be vigorously put to practical use, and the recycling rate and landfill rate should be improved. Until economically viable options are researched, the United States should intensively implement already existing technology through policy to mitigate the accumulation and environmental impacts of landfill waste.

The four European countries on this list (Italy, Germany, England, and Spain), which rank in the middle in terms of number of publications, have higher recycling rates than the Asian countries in the top 10 and the United States. The amount of post-consumer plastic waste per capita is close to the average of the top 10 countries. Environmental awareness is generally high, and these factors are thought to be increasing publication numbers.

India, which ranks seventh in number of publications, has a low recycling rate, although there is a wide range. The amount of post-consumer plastic waste per capita is the lowest among the top 10 countries. India has the lowest per capita Gross Domestic Product (GDP) among the countries listed, and it can be said that India is doing well in producing publications given its economic size. In India, there are many unknown figures such as the landfill rate, energy recovery, and untreated waste rate, and it is necessary for the government to collate data for the necessary statistics. The Indian government has recently announced a ban on single-use plastics and may be encouraging research into plastic recycling.

Japan, which ranks eighth in the number of papers published, has a lower recycling rate than the four European countries and China. It is characterized by a high percentage of energy recovery and a low landfill rate. The amount of post-consumer plastic waste per capita is the second lowest of the ten countries. Japan’s environmental awareness is the lowest among the top 10 countries, and it is possible that the nation’s low interest in the environment is related to its low number of publications.

Brazil, which ranks ninth in the number of papers, has the lowest recycling rate of the top 10 countries and the second highest landfill rate after the United States. As much as 24.4% of plastic waste may have been released into the environment without proper treatment, which constitutes a significant issue for environment and human health. Brazil has major challenges in developing a circular economy. Despite the country’s small economic power, having the second smallest GDP per capita of countries on this list, its ranking in the top 10 in number of publications may suggest that researchers want to solve these problems.

The Netherlands, ranked 10th in number of publications, has a high recycling rate and the lowest landfill rate among the top 10 countries. Among European countries, the progression of their circular economy is comparable to Germany. The reason the number of publications ranks 10th could be that it has the smallest population among the top 10 countries.

The second issue is whether plastic waste should be treated nationally or globally. A fundamental question is whether the current trends in research can resolve the issues regarding plastic waste not only in individual countries but also globally. In Table 6, we only focus on the domestic waste treatment, but we must note that international trade has affected the burden of waste for many countries.

The burden by plastic waste has not been distributed equally, but mainly in China. Until the mid-2010s, China imported 40 to 50% of the world’s plastics exports, and as a result, plastic waste from developed countries such as the United States, Europe, and Japan has been stably managed [257]. It is speculated that the recycling rate figures reported for developed countries included the amount of waste that was recycled after being exported abroad. Since 2017, China has gradually tightened import restrictions on waste such as plastic waste, and in 2018 the amount of plastic waste imported was almost zero [253]. As a result, the plastic recycling rate in the United States decreased from 9.1% in 2015 to 4.4% in 2018 after China’s plastic waste import ban [258]. Developed countries that have depended on exports to China have fallen into a serious shortage of processing capacity for plastic waste, and the price of plastic waste, which is used as a raw material for recycling, has plummeted [259,260]. Developing countries such as those in Southeast Asia have become importers of plastic waste as there is no single country that can replace China’s former import volume [261]. The top exporting countries in 2019 are, in descending order, Germany, Japan, and the United States [262]. These countries are now increasing their exports to countries with relatively small economic margins within their respective regions (Germany to Eastern Europe, Japan to Southeast Asia, and the United States to Mexico and Asia) instead of exporting to China [262].

To combat this, governments in Southeast Asian countries are making efforts to restrict imports due to concerns of polluting the environment within their own countries, and exporting to these developing countries is becoming more difficult year by year [253]. In 2021, an international treaty called the Basel Convention has been implemented, requiring the consent of the importing country when exporting dirty plastic waste. This is a further headwind for the export of plastic waste from developed to developing countries [258].

An ideal solution for plastic waste is to be recycled and consumed as recycled plastics in their country of origin. In this review, we refer to this way of thinking as the principle of local waste treatment. In the principle of local waste treatment, the country where the plastic is consumed and discarded should be responsible for processing the waste. The advantage of this principle is a normative one. Developed countries have more consumed and wasted plastics than developing countries. The former should not impose their waste and the associated environmental and social burdens to the latter. The disadvantage of this principle is economic feasibility. Recycled plastics are often inferior to virgin plastics in terms of strength and quality, making it difficult to stimulate domestic demand for recycled plastics and tending to limit the amount of recycled plastic waste. Furthermore, the demand for recycled plastics is higher in developing countries, which emphasizes lower-priced plastic materials. In addition, the cost of handling plastic waste is generally higher in developed countries. Therefore, it is necessary to increase the processing capacity of plastic waste in developed countries, which will reduce the cost of handling [263]. In this situation, plastic waste is likely to be exported if waste disposal and processing companies in exporting countries only pursue economic rationality. However, local waste treatment is not always economically infeasible. Some plastic waste still has economic value as resources. Additionally, the (mainly developed) countries that generate waste often have nearly sufficient capabilities to appropriately treat their waste.

Another principle is global waste treatment. According to this principle, countries generating plastic waste do not have a responsibility to treat it within the country but should seek the most economically efficient way. This can include international trade of wastes. Some previous studies suggest that developed and developing countries should work together to build a global circular economy system in which high-quality waste is exported to developing countries for recycling [260,261]. If the principle of global waste treatment is implemented, it has the advantage of being able to minimize the total processing cost for both the exporting and importing country. Economic cooperation and business matching from developed to developing countries may improve the accuracy and capacity of recycling systems in the latter. Inhibiting international trade will shift economic equilibrium in an undesirable direction. For example, mechanically recycled plastics are economic and popular in China; thus, there is a large demand for plastic waste as a raw material. At present, when plastic waste cannot be imported, increasing the supply of plastic waste by improving the recovery rate (recycling rate) is an urgent issue in China [263]. Recently, Chinese mechanical recycling companies have expanded into Japan to obtain plastic waste. They process Japanese plastic waste into recycled plastic raw materials, such as pellets or flakes, in Japan and export them to China [264]. Because recycled raw materials are no longer considered waste, China can import them. Such efforts have increased in recent years. Disadvantages of the principle of global waste treatment are that many of the importing countries are developing countries, and they sometimes tend to lack the legal systems, infrastructure, financial capacity, expertise, and human resources needed to ensure proper treatment of plastic waste. There is a risk that this will lead to the release of waste into the environment in inappropriate way that will degrade human and ecosystem health.

In order for the principle of global waste treatment to function, the exporting country should have the responsibility in capability development of waste treatment rather than (or in addition to) waste treatment itself. In addition, it is important to ensure the traceability of the flow of waste and waste treatment. At present, developed countries do not know how the waste exported to developing countries is recycled and how the residue generated as a result of recycling is treated. Several other issues are concerns to be resolved by the developing countries. There is a concern that even the governments of developing countries are not aware of the actual state of plastic waste recycling within their territory. In order to ensure that importing companies treat plastic waste properly, it is important to make the actual state of treatment transparent. Therefore, it is necessary for both exporters and importers to take responsibility and build a traceability system for handling plastic waste. For example, an international manifesto system that tracks the movement of plastic waste in importing countries would be effective. The exporter issues a control sheet called a manifesto together with the waste to the importer (transporter). The importer describes in the manifesto when, by whom, and how the waste was transported and processed. The importer must return the manifesto to the exporter within a certain period of time. In order to ensure the accuracy of the contents of the manifesto, export companies or third-party organizations should conduct regular audits. Governments of exporting and importing countries should be involved in the establishment of this system, otherwise, international NGOs should be responsible for the system's operation. Such a manifesto system will greatly help ensure international traceability of plastic waste.

As a third issue, we focus on the ranking of the number of papers in each subcluster by country, and discuss the characteristics of each subcluster and trends by country. Table 7 shows the number of publications by each country in clusters and subclusters.

Table 7. Ranking by country and share of number of papers in cluster 1, 2 and 3. "#" represents a number.

Cluster #	Rank Research Topic	1	2	3	4	5	6	7	8	9	10
1	Plastic recycling	China 12.4%	USA 11.1%	Italy 7.5%	Germany 6.9%	UK 6.7%	Spain 6.2%	India 5.9%	Japan 5.2%	Brazil 4.7%	Netherlands 3.2%
1-1	Recycling by pyrolysis	China 11.0%	Spain 10.0%	India 9.5%	USA 8.0%	UK 6.0%	Japan 5.3%	Italy 4.1%	Porland 3.8%	Germany 3.5%	Malaysia 3.5%
1-2	LCA of plastic recycling	USA 16.4%	China 14.6%	Italy 9.0%	Germany 7.0%	UK 6.5%	Spain 6.5%	Japan 6.3%	Netherlands 4.4%	Denmark 3.7%	Switzerland 3.3%
1-3	Mechanical recycling	Brazil 9.7%	USA 8.2%	China 8.0%	Italy 6.9%	Spain 5.7%	Germany 4.6%	India 4.2%	France 4.2%	Japan 4.0%	Sweden 3.7%
1-4	Biodegradation of plastic	USA 17.7%	China 17.1%	Germany 13.8%	India 6.7%	UK 6.5%	Italy 4.6%	Brazil 4.4%	Japan 3.8%	Sweden 3.6%	Canada 3.6%
1-5	Bioplastic	USA 13.6%	Italy 12.4%	UK 9.3%	Spain 8.3%	China 8.1%	Germany 7.8%	Netherlands 5.8%	Poland 5.6%	India 5.1%	Portugal 3.8%
1-6	Recycling of PVC	Japan 29.5%	China 17.9%	India 8.4%	Korea 5.3%	USA 4.2%	Jordan 3.2%	Germany 3.2%	Russia 2.6%	Australia 2.1%	Italy 2.1%
2	WEEE and sorting of plastic waste	China 25.8%	USA 11.0%	Germany 6.5%	India 6.4%	Japan 6.4%	France 6.2%	Italy 6.1%	Korea 4.5%	UK 4.2%	Australia 3.7%
2-1	Recycling of WEEE	China 13.2%	Germany 12.6%	UK 9.4%	USA 7.9%	Belgium 7.4%	France 7.4%	India 6.8%	Italy 6.5%	Switzerland 5.0%	Brazil 4.7%
2-2	Spectroscopy sorting	Italy 17.3%	Germany 13.8%	France 10.4%	China 10.0%	USA 9.6%	Spain 5.0%	Brazil 4.6%	Malaysia 4.6%	Korea 3.8%	Japan 3.8%
2-3	Flotation separation	China 31.0%	Japan 15.0%	Korea 7.5%	Italy 6.4%	Australia 4.8%	USA 4.8%	Portugal 4.3%	UK 4.3%	Turkey 4.3%	Spain 3.2%
2-4	Electrostatic separation	France 39.0%	Algeria 26.2%	USA 15.6%	Romania 15.6%	China 9.2%	Korea 6.4%	Poland 4.3%	Japan 3.5%	Italy 2.8%	Canada 2.8%
3	Use of plastic waste in the construction sector	China 17.5%	USA 13.1%	Australia 8.4%	India 7.2%	Spain 7.0%	Italy 6.0%	Brazil 4.7%	Malaysia 4.0%	UK 4.0%	Portugal 3.3%
3-1	Use of recycled plastics in concrete	India 12.7%	USA 7.0%	IRAQ 6.3%	China 5.7%	Malaysia 5.4%	Saudi Arabia 5.4%	UK 5.1%	Italy 4.7%	Australia 4.7%	Algeria 4.4%
3-2	Use of recycled plastics in asphalt	China 14.0%	USA 11.4%	Spain 10.6%	Australia 10.6%	India 8.1%	Italy 7.6%	Malaysia 5.9%	Saudi Arabia 4.2%	Turkey 4.2%	Portugal 3.8%

The United States, which ranks second in number of papers in cluster 1 as a whole, ranks first in several subclusters, with a young average age of publication and a rapid increase in the number of papers in recent years. The subclusters in which the United States ranks first and the associated average age of publication are: subcluster 1-4, biodegradation of plastics (2018.7), subcluster 1-5, bioplastics (2017.9) and subcluster 1-2, LCA of plastic recycling (2015.7). In contrast, China, which ranks first in the number of papers overall, ranks first in only one subcluster with a relatively old average publication year: subcluster 1-1, recycling by pyrolysis (2013.5). The United States is considered to be more advanced than China in research into new academic fields. Given the high landfilling rate in the United States, it is possible that there is intense research into the biodegradation of plastics and bioplastics aimed at the environmentally friendly disposal of landfilled plastics.

India and Brazil are not included in the top 10 countries of subcluster 1-2, LCA of plastic recycling, and it is possible that countries with low GDP per capita are not enthusiastically conducting research on the LCA of plastic recycling. Brazil has the highest number of papers in subcluster 1-3 mechanical recycling, indicating that the promotion of mechanical recycling is a major issue in Brazil, where the recycling rate is low, and the landfilling rate is high. In subcluster 1-6, recycling of PVC, countries other than the United States and those in Europe occupy the top positions, and the trend is very different from other clusters. In order of number of publications, Japan, China, India, Korea, Jordan, and Russia all make the top 10 in subcluster 1-6. In particular, the number of publications from Japan is the highest, and due to the number of papers in this subcluster, Japan ranks eighth in cluster 1. In Japan, it is thought that the processing of PVC is recognized as a major issue in promoting recycling. By contrast, in Europe and the United States, contamination with PVC may not be regarded as a major hindrance to recycling.

In subcluster 2-1, recycling of EEE waste, China ranked first, followed by several European countries, and India and Brazil ranked within the top 10. Subcluster 2-1 is similar to cluster 1 in this respect. Countries where plastic recycling has become a social issue may indicate that EEE waste is also of high social concern. Subcluster 2-2, spectroscopy sorting, is characterized by the fact that European countries, specifically Italy, Germany, and France, occupy the top three positions, while China and the United States are at the fourth and fifth place, respectively. Spectroscopy sorting is being intensively studied in Europe. Asian countries such as Malaysia, South Korea, and Japan are also well represented. Subcluster 2-3, flotation separation, is characterized by the fact that Japan (2nd), South Korea (3rd), and Turkey (9th) are taking vigorous efforts. Subcluster 2-4, electrostatic separation, is characterized by European countries such as France, Romania, Poland, and Italy being prominent. We need to improve the recycling capacity to realize a circular economy and to shift the scope of circulation from local to global, and then to planetary scales. As reviewed in this paper, there a variety of both established and emerging recycling technologies, including mechanical, chemical, and biological recycling. In addition, recent subclusters within cluster 1 on plastic recycling focus on the circular economy at the planetary scale. As already seen, emerging subclusters in cluster 1 are subcluster 1-4 biodegradation of plastic (2018.7), and subcluster 1-5, bioplastics (2017.9); which are younger than the other subclusters such as subcluster 1-1, recycling by pyrolysis (2013.5).

As the fourth issue, we will discuss the necessity of global technology transfer of the research results of plastic recycling. Another point clarified through our analysis is the necessity of international technological cooperations. We need global technology transfer for efficient and effective waste treatment, as capability of recycling and waste treatment are not equally distributed.

It became clear that research on plastic recycling (cluster 1) is actively being carried out in developed countries such as China and the United States. Of course, it is desirable that such research results be put to practical use in China and the United States, particularly in the latter, where the recycling rate is low and landfilling rate is high. However, in order to promote the international division of labor, it is critical that these research results be transferred to developing countries that import waste. Expected methods of technology transfer include the dispatch of experts from developed to developing countries, capacity building by inviting human resources from developing to developed countries, and international technology conferences. We hope that researchers and companies with recycling technology in developed countries, such as China and the United States, will actively engage in technology transfer to developing countries. Furthermore, in order to solve the problem of lack of funds in developing countries, we expect for companies from developed countries to enter developing countries and establish plastic recycling businesses.

In contrast, countries other than Europe, the United States, and China (mostly developing countries) are in the top 10 of cluster 3, a notable divergence from clusters 1 and 2. For example, in subcluster 3-1, "Use of recycled plastics in concrete," India ranks first, followed by Iraq (2nd), Malaysia (5th), Saudi Arabia (6th), and Algeria (10th). In subcluster 3-2, "Use of recycled plastics in asphalt," India ranks fifth, followed by Malaysia (7th), Saudi Arabia (8th), and Turkey (9th). Recycling methods for utilizing plastics in the construction sector tend to be vigorously researched in the developing and emerging countries. Recycling methods that mix plastic waste into building materials are characterized by relatively low processing costs and the ability to process a large amount of plastic waste. It appears that countries with low economic capacity have high expectations for these technologies. According to the head of India's plastic industry association: "Plastindia Foundation", the government policy in India is to actively use plastic waste in road construction (up to 20% of all plastic waste produced in India shall be used in road construction by regulation). In addition, the average publication year of these subclusters is relatively young (subcluster 3-1: 2017.1, subcluster 3-2: 2017.5) and these topics are areas of active research and attention.

There is a possibility that these topics will result in reverse innovation where these technologies are transferred from developing to developed countries. Concerning the use of recycled plastics in concrete, there are still remaining issues regarding strength and handling at construction sites. However, there are high expectations for its use in structures that do not require high strength and for new applications that take advantage of its lightweight concrete properties. Numerous studies have reported that the use of plastics in asphalt improves strength and durability, and there are even examples of its real-world application in road construction. In addition, studies have reported that the use of recycled plastics for asphalt reduces road construction costs and improves the economic efficiency of road construction [226,227]. Another study reported that mixing recycled plastics into asphalt improves the strength and durability of the asphalt mixture, extends the life of roads, and reduces the environmental impact of road construction [228].

We must also note that because the use of plastics in the construction sector is one-way use and not circular, if we prefer circularity in developed countries, it is preferable to recycle mechanically or chemically as much as possible, and to utilize only waste that is not suitable for recycling using other methods in the construction sector. However, from the point of view of reusing plastic waste at the lowest possible cost without releasing it into the environment (including landfilling), using plastics in the construction sector is a promising option, and there are high expectations for it in developing countries.

5. Conclusions

Using bibliometrics analysis, we synthesized an overview of 35,519 publications on plastic recycling, identified emerging topics, and conducted a comprehensive review to elucidate research trends and key issues. We collected bibliographic data from academic publications related to plastic recycle. We used data collected with the query (plastic* OR chemical*) AND (recycl* OR "circular economy") by using academic database "Web of Science". After acquiring relevant publications, we created citation networks by treating the papers as nodes and the citations as links. We used the direct citation method. We removed irrelevant papers that were not connected to other papers in the largest component of the citation network. We divided the network into clusters using the Newman's algorithm topological clustering method after obtaining the largest connected component. Using this algorithm, we divided clusters into subclusters according to the rule of maximizing modularity, which has been used in previous bibliometric studies.

We found that research topics on plastic recycling can be broadly classified into the following six clusters: general issues of plastic recycling; waste electrical and electronic equipment (WEEE); use of plastic waste in the construction sector; chemical recycling of polyethylene terephthalate; use for wood-plastic composites; and recycling of fiber reinforced polymers. After extracting the above clusters, we conducted a comprehensive review on each cluster as well as subclusters of the larger three clusters.

The largest cluster (cluster 1) is on general issues of plastic recycling and includes subclusters such as the biodegradability of plastics, bioplastics, pyrolysis, and life cycle assessment (LCA). Among them, the biodegradability of plastics is the youngest subcluster (average publication year, 2018.7) and the most active topic. Many studies on biodegradation of plastics derived from fossil resources are being conducted, and at the same time, research on biodegradable plastics is also attracting attention. The former is still in the research stage and has not been industrialized, while the latter, such as PLA, PHA, has been industrialized, but the production cost is extensively high. Consequently, the market share is low. In general, biodegradable plastics need to be sorted by consumers because the recycling method differs from that of other plastics. We pointed out the problem that it is difficult to distinguish the type of plastics just by the appearance, and that recycling methods have not been established. Bioplastics is the second youngest subcluster (average publication years, 2017.9), with a rapidly increasing number of papers. Definitions of bioplastics differ among papers, and we clarified that three different definitions were used. In this study, we defined bioplastics as polymers derived from renewable resources and materials or synthesized by microbial metabolism. Pyrolysis is a relatively old subcluster (average publication year, 2013.5), but has the largest number of papers in cluster 1 (number of papers, 772). The citation per paper ratio is also the largest (4.8), which makes this subcluster the central theme in cluster 1. LCA is a relatively young subcluster (average publication years, 2015.7) with the second largest number of papers in cluster 1 (number of papers, 568). The combined results of many studies on LCA reveal that mechanical recycling is superior to chemical recycling in terms of global warming potential, but inferior in terms of residual solid waste for landfill. We proposed that mechanical recycling and chemical recycling should not compete with each other, but should be used in a complementary manner depending on the type and condition of plastic waste.

In the second largest cluster (cluster 2), research regarding WEEE recycling is increasing rapidly (average publication years, 2014.8). The brominated flame retardants (BFR) used in WEEE plastics is hazardous to human health and ecosystems. Hazardous BFR waste is transported both legally and illegally to areas where labor costs are low. As much as 1818 kg of harmful brominated low-molecular-weight compounds are released into the environment every year around the world, especially in disposal sites in Asia. The treatment of BFR make recycling difficult, and considerable effort is being taken to address this. Mechanical recycling is the most desirable method for treating WEEE plastic, and most of the recycling currently performed is mechanical recycling. The separation of BFR from WEEE by chemical recycling has been intensively researched but not industrialized.

In the third largest cluster (cluster 3), there is increasing research into the use of recycled plastic waste in the construction sector. Cluster 3 consists of two subclusters, "Use of recycled plastics in concrete" (average publication year, 2017.1) and "Use of recycled plastics in asphalt" (average publication year, 2017.5). Both are young research fields. The number of papers on the use of recycled plastics in concrete (2028) is higher than the number of papers on their use in asphalt (734). Citation/paper ratio of concrete is 6.4, which is higher than 3.1 of asphalt. Concrete applications are a more intensely studied research topic than asphalt applications. On the other hand, the use of recycled plastic mixed with concrete results in inferior strength and durability, and there are few reports of actual field applications. In the case of asphalt use, many studies have reported that strength, durability, and economic efficiency are improved, and there are practical examples in actual road construction.

By country, we found that China and the United States had the highest number of papers. Specifically, in cluster 1, China ranked first with a share of 12.4% of the total number of papers, and the United States ranked second with a share of 11.1%. In cluster

2, China ranked first (25.8% share of papers) and the United States second (11.0% share of papers). Our first key finding is that China and the United States are global leaders in many research fields. In contrast, countries other than Europe, the United States, and China (mostly developing countries) are in the top 10 of cluster 3, a notable divergence from clusters 1 and 2. For example, in subcluster 3-1, "Use of recycled plastics in concrete," India ranks first, followed by Iraq (2nd), Malaysia (5th), Saudi Arabia (6th), and Algeria (10th). In subcluster 3-2, "Use of recycled plastics in asphalt," India ranks fifth, followed by Malaysia (7th), Saudi Arabia (8th), and Turkey (9th). These studies are being actively carried out in developing countries, and it is thought that they are attracting attention due to their high economic efficiency as a recycling method. These are reverse innovation that should be considered as methods of using waste that are not suitable for recycling using other methods, even in developed countries. Our second key finding is that research on the use of recycled plastics in the construction sector is actively being conducted in developing countries.

In order to realize a global circular economy, we proposed and discussed the principle of local waste treatment, the principle of global waste treatment, and global technology transfer. In the principle of local waste treatment, plastic waste should be handled responsibly and appropriately in the country where it is generated. According to this principle, the environmental burden associated with waste treatment may be minimized, but the economic rationality is questionable. In the principle of global waste treatment, the international trade of waste resources is allowed and requires a division of labor between developed and developing countries. Although the principle of global waste treatment has the advantage of minimizing the cost of processing plastic waste globally, there remain concerns that it may promote environmental pollution associated with improper waste processing in importing countries. We also highlighted the necessary measures to promote both principles, such as building a traceability system and transferring technology in both directions between the developed and developing countries.

We proposed that an international manifesto system which tracks the movement of plastic waste in importing countries is an effective way for buiding a traceability system and ensuring appropriate plastics waste reatment. The exporter issues a control sheet called a manifesto together with the waste to the importer (transporter). The importer describes in the manifesto when, by whom, and how the waste was transported and processed. The importer must return the manifesto to the exporter within a certain period of time. In order to ensure the accuracy of the contents of the manifesto, export companies or third-party organizations should conduct regular audits. Such manifesto system will greatly help ensure international traceability of plastic waste. The international manifesto system is our research contribution for global plastic waste treatment. Further research is required to identify the means to realistically advance in both principles.

In addition, we discussed the necessity of global technology transfer. Especially research on the use of plastics in the field of construction that is actively being conducted in developing countries. Although there are criticisms that the use of plastic waste in the construction sector is not circular, considering the economic efficiency and environmental improvement effects associated with using recycled plastics in the construction sector, plastic waste that is not suitable for recycling can be used in construction. Even in developed countries, the use of such plastics in the construction sector has a certain rationality. For this reason, the technology transfer (reverse innovation) of research in this field from developing to developed countries should also be actively promoted. In the theory of international cooperation, technology transfer in both directions between developed and developing countries is essential for realizing proper plastic waste treatment and recycling systems as well as to promote a circular economy.

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