

Plastic waste recycling via pyrolysis: A bibliometric survey and literature review

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ABSTRACT

Plastic materials have been crucial to the development of science, technology, and almost all aspects of modern progress since the mid-twentieth century. However, the increasingly unsustainable culture of plastic consumption and the accumulation of plastics in landfills, oceans, and broader ecosystems has also made negative, potentially irreversible environmental impacts. In recent decades, scientists and engineers have spent significant time and resources searching for more effective plastic waste management techniques based on thermochemical routes like pyrolysis. Indeed, plastic to fuel conversion has the potential to severely limit plastic pollution and to contribute to the circular economy, but industrial scale plastic pyrolysis has not been achieved. Therefore, this paper presents a bibliometric analysis and systematic literature review of pyrolysis-related articles in the Web of Science database published between 2001–2020. The resulting articles ($n = 670$) show that Spain is the most productive country in terms of total output and that there are an increasing number of researchers focused on this topic worldwide. The results also highlight the current landscape and future directions of plastic pyrolysis research based on the following hot topics: i) kinetic triplets as a vital component of plastic pyrolysis and scaling up processes, ii) catalysts syntheses and performance, iii) co-pyrolysis of plastic/biomass mixtures, and iv) reactor design and reaction parameters. In conclusion, the study offers a comprehensive overview of plastic pyrolysis progress, which will remain a major area of research for chemists and engineers in the coming decade and a powerful tool for environmental management.

1. Introduction

Plastics are cheap, pliable, and moldable materials. Since the 1940s, communities worldwide have developed a “plastic culture” [1]. The material has improved quality of life for many, but also changed global consumption patterns, increased demand for resources and production, and generated almost incalculable tons of waste and pollution. From food packaging to aerospace to medical instruments, our lives are metaphorically, and sometimes literally, wrapped in plastic. In fact, plastic consumption has increased twentyfold since the 1950s, and it is expected to reach to 720 million tons in the next 20 years [2]. To address this issue, in 2018 the European Commission the EU Strategy for Plastics in the Circular Economy, which establish the main goals for plastic

design, manufacture, use, re-use, and end-of-life management by 2030 [3].

In this line, several actors around world are focused on developing novel and holistic solid waste management plans that address the problems of plastic waste. The life cycles of all consumer products, and especially single-use plastics and personal electronics, must be reconsidered with attention to environmental management and sustainability. While there are numerous methods for recycling of plastic waste, only primary recycling (re-extrusion) and secondary recycling (mechanical) are well-established [4]. There are still challenges in the large-scale implementation of tertiary recycling (chemical recovery) and quaternary recycling (energy recovery) of plastic waste.

Despite the significant development of various technologies to

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handle homogeneous and relatively clean plastic waste in primary and secondary recycling, converting plastic waste into building block molecules, fuels, and energy remains a significant challenge [5]. Part of the challenge is separating plastic waste from other solid waste components; without this step, it is not possible to process used plastic and obtain high-quality outputs [6]. Several tertiary recycling methods and technologies have attracted attention and have the potential to be useful on the commercial stage. These include depolymerization (chemical feedstock recovery), as well as plasma arc gasification, and pyrolysis [7]. Among the available processes for converting plastics into fuel or any other value-added product, pyrolysis has drawn the most scientific attention between early-stage and mature technologies. Recently, Solis and Silveira [6] provided an extensive TRL assessment on eight technologies available for plastic recycling and reviews recent technological developments in pyrolysis, catalytic cracking, and gasification for TRL 8-9. Nevertheless, the full potentials of these technologies and their impact on the circular economy of plastics is still unclear and requires more full-scale projects to be subjected to critical examinations. Meanwhile, another review by Qureshi et al. [8] argues that pyrolysis can be an effective management tool that will complement mechanical recycling. Some of the primary challenges with elevating plastic to fuel pyrolysis to the industrial scale are feedstock quality, segregation of materials, reactor operations, and stability and standardization of the end products. Finally, Spreafico et al. [9] recently reviewed the innovations and the evolution of different technologies for plastic pyrolysis. Their analysis of patents and articles further indicates that pyrolysis technologies are changing from macro to micro, especially with the use of laser or microwave pyrolysis systems. Efforts to optimize raw material and energy input are advancing quickly. Therefore, the present review aims to offer further insights into how the state-of-the-art has and will develop.

Despite the abundance of scientific publications and reviews on plastic pyrolysis, we are not aware of any reviews showing the overall evolution of the state-of-the-art in recent decades and which clarify the current publication landscape of plastic waste pyrolysis research. To help fill this gap, we perform a bibliometric meta-analysis and systematic literature review of articles describing pyrolysis techniques and technologies published between 2001–2020.

Bibliometric meta-analysis and systematic literature reviews can help to identify research trends, hot topics, and new dynamics between research fields, countries, and researchers [10–12]. The information offers a broader view of a research landscape and, in addition to informing researchers, can inspire governments, financial institutions, and funding agencies to support new strategies to address emerging topics and to help fill anticipated gaps in research.

This bibliometric meta-analysis and systematic literature review provide readers with current information about *who*, *where* and *what* the scientific community are doing in order to improve plastic waste pyrolysis as a tertiary recycling method, which deals with conversion of the mixed plastic waste and impurities into useful fuels. The results suggest relevant hotspots in the state-of-the-art, which can help researchers in anticipating which processes and knowledge gaps must be addressed to scale up the plastic waste pyrolysis as a tertiary recycling method. In short, this review provides a baseline for further research in plastic pyrolysis technologies.

Our analysis adds to existing research by highlighting the unique potential for pyrolysis to treat plastic waste and, by extension, aid environmental management. Our review addresses the following dimensions of pyrolysis research: i) publications and citations, ii) top cited publications and journals, iii) researcher status by most cited authorship and networking, iv) productivity among country and institutions, and v) co-occurrence keywords. These keywords help to unravel the most important directions of plastic pyrolysis research and indicate new or underexplored topics.

2. Methodology

To understand and appreciate the landscape of plastic pyrolysis, we analyzed published research using the PRISMA model or the “Preferred Reporting Items for Systematic Reviews and Meta-Analyses” [13]. The PRISMA method has helped to standardize literature review and bibliographic analysis, as indicated in our previous research [14] and summarized in Fig. 1. Adopting PRISMA requires following four systematic steps—identification, screening, eligibility, and qualitative synthesis—and completing a final checklist.

We focused our review and analysis on peer-reviewed journal articles with keywords related to plastic pyrolysis published within 2001–2020. Initially, we searched both the Web of Science (WoS) and Scopus databases using “TS” or “Title-Abs-Key” as a field tags searching, respectively. The query employed in both case was (((“plastic waste” or “polymer waste” or polyolefin* or polyethylene or polypropylene or polystyrene or “polyvinyl chloride” or “poly(ethylene terephthalate)”) and (“thermal pyrolysis” or “catalytic pyrolysis” or “thermal cracking” or “catalytic cracking” or “thermolysis” or “thermocatalytic”))). The timespan selected was 2001–2020 and the quotation marks used before (“”) were necessary to extract documents with exact phrases and to avoid lemmatization and synonyms. The Boolean operator “or” and “and” were required to ensure the appearance of at least one term between parenthesis.

An initial search with the same queries and restricted to only articles, proceedings, and reviews produced 1201 documents from WoS and 1284 documents from Scopus. A majority of the documents and all of the top cited papers (Table 1) were the same for both databases. We chose to focus our analysis and review solely on the results from WoS for three reasons. First, the top two papers in the results from Scopus did not relate to pyrolysis and would have been removed in the screening stage. Second, we noticed discrepancies in the citation counts for many of the specific articles with generally higher citation counts in Scopus, which would have impacted the accuracy of our analysis made by data-mining software (VOSViewer) of authors, country, and journals and the co-occurrence map. Finally, WoS is a thorough database with the largest number of indexed journals and conference proceedings (~12,000) and covers nearly 150 research disciplines. Our decision to use only results from WoS for our study was further validated after we completed our meta-analysis and review and realized that, of the 670 articles that were found to be eligible based on review of titles and abstract, 422 of these documents were in the results from Scopus, suggesting that we had not limited our results in any way and, instead, gained some clarity in the meta-analysis with regards to citation counts.

On November 5, 2020, all original data was extracted from the online version of the WoS database (indexes: Science Citation Index-Expanded) by selecting “marked list” function and exporting the data in “plain text” format file. A keyword co-occurrence map was generated using VOSviewer based on the extracted publications from WoS. A thesaurus file was used to remove duplicates and to standardize the variations of the spelling for authors name or keywords which refer to the same author or concept, respectively. The resulting 1201 documents were then screened using the WoS function so as NOT to exclude papers related to “carbon based materials” OR “nanotube*” OR “CNT*” or “algae” OR “asphalt”. After this screening phase, we had 897 documents. In the next step, eligibility, we read titles and abstracts to eliminate publications related to a range of topics adjacent to pyrolysis, including but not limited to the following areas: hydrothermal treatment of plastic, plastic life-cycle assessments, and thermochemical treatments of solid waste. After the eligibility phase, the 670 eligible documents were included in the meta-analysis and review. Other reviews on urban solid waste pyrolysis [15] and tire waste pyrolysis [16] have been recently published; therefore documents on these topics were excluded in our review.

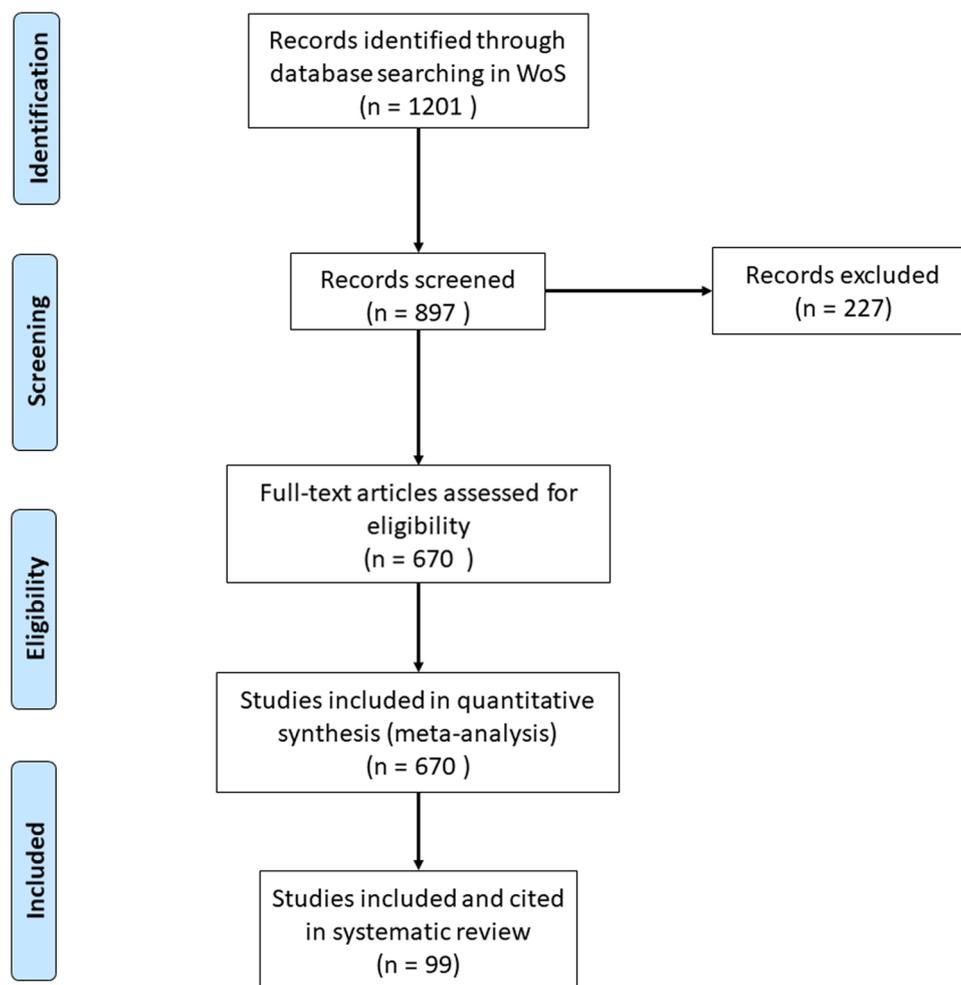


Fig. 1. Flow diagram illustrating literature research and selection process based on PRISMA model as adapted from Moher et al. [13].

3. Publication and citation trends

The temporal publication and citation trends are among the important bibliometric indicators of a research topic. These indices illustrate the productivity of scholars in this field as well as the associated influence [17]. The earliest works on plastic pyrolysis were published by Kiang et al. [18], and Okui et al. [19] focused on waste recovery from industrial processes. Between 2001–2020, 549 articles (82 %), 90 proceedings (14 %), and 31 reviews (4.0 %) were published. Overall, an increased number of publications related to plastic pyrolysis in 2001–2020 is observed in Fig. 2 with a fluctuating trend, with a higher rate of increase observed since 2016. A similar trend is also observed for the total citations (T_C) received in different years. A total of 16,374 citations (including self-citations) were received by these publications, with an average of 25.13 citations per publication. English is used in 661 publications (98.6 %), making it the dominant language. This is followed by Japanese (2, 0.3 %), Polish (2, 0.3 %), Portuguese (2, 0.3 %), Czech (1, 0.2 %), and Korean (1, 0.2 %).

The study of plastic pyrolysis as a management tool for plastic recycling has experienced a rapid increase of total publications and authors. The blue circles in Fig. 2 show the number of authors normalized by the number of documents published in the same year. This ratio between authors/documents has been used to help to trace and tag “hotspots” in different fields [6,20]. A rapid increase of the authors/documents ratio shows that not only is more work in this field being published, but that more researchers are actively participating in the research. Previous works have related these trends to the development of new research domains or frontier which are able to attract more

researchers in the earlier stage of progress [21,22].

3.1. Top cited publications

A total of 27 out of 670 publications have received more than 100 citations in the studied period. The ten-most cited publications in this subject field are displayed in Table 1. The top-cited papers published before 2009 are research papers that provide important evidence on the technical feasibility of plastic-to-fuel production. For example, the research by Bagri et al. [23] provides important insights on the possible conversion of polyethylene (PE), the most widely used plastic, to liquid and gaseous hydrocarbons over ZSM-5 and Y zeolites. Their work demonstrated that formation of aromatic compounds in the liquid product is promoted by increasing catalysis temperature, which has been verified by subsequent studies [24,25]. Sharypov et al. [26] revealed the potential of liquid fuel production from a mixture of plastics and biomass waste via co-pyrolysis. The synergistic effect on the liquid yield was observed at a high plastic percentage in the mixture, and the polymer tacticity played a significant role in liquid product distribution. Thermal pyrolysis of real plastic waste (consisting of PE, polypropylene (PP), and polystyrene (PS)) collected from landfills was investigated by Demirbas et al. [27]. During pyrolysis, PS decomposed to form monomers via end-chain scission and random chain scission mechanisms. The pyrolysis produced hydrocarbons with a lower molecular weight with a wide carbon number range. Thermal cracking at a higher temperature also led to higher gas yield (at the expense of liquid yield), as well as higher paraffin and aromatic fractions in the liquid product. However, the authors observed several issues which limited the

Table 1

Top ten cited papers related to plastic pyrolysis research and chemical reactor employed, ordered by publication year.

| Publication | T _c | Year | Chemical reactor or setup employed | Ref |
|--|----------------|------|------------------------------------|------|
| Catalytic pyrolysis of polyethylene | 161 | 2002 | FBR | [23] |
| Co-pyrolysis of wood biomass and synthetic polymer mixtures. Part 1: influence of experimental conditions on the evolution of solids, liquids, and gases | 168 | 2002 | TGA | [26] |
| Pyrolysis of municipal plastic wastes for recovery of gasoline-range hydrocarbons | 202 | 2004 | TGA | [27] |
| Chemical recycling of plastic wastes made from polyethylene (LDPE and HDPE) and polypropylene (PP) | 247 | 2007 | FBR | [28] |
| Recycling and recovery routes of plastic solid waste (PSW): A review | 822 | 2009 | na | [4] |
| The valorization of plastic solid waste (PSW) by primary to quaternary routes: From re-use to energy and chemicals | 189 | 2010 | na | [29] |
| Thermolysis of waste plastics to liquid fuel: A suitable method for plastic waste management and manufacture of value-added products-A world prospective | 293 | 2010 | na | [30] |
| Thermolysis of waste plastics to liquid fuel | | | | |
| Current state and future prospects of plastic waste as a source of fuel: A review | 180 | 2015 | na | [31] |
| Thermal degradation of PVC: A review | 198 | 2016 | na | [32] |
| A review on thermal and catalytic pyrolysis of plastic solid waste (PSW) | 181 | 2017 | na | [33] |

na = non-applicable (review paper).

potential of liquid products as a substitute of gasoline, including the low octane number and high chlorine content obtained from Polyvinyl Chloride (PVC). Achilia et al. [28] compared the performance of plastic waste recycling via the pyrolysis and dissolution/precipitation techniques that are conventionally used in plastic recycling. While

satisfactory performances were observed in both routes, the use of an organic solvent in large amounts was said to be a disadvantage for the latter method. Nevertheless, the pyrolysis products can be used as feedstock for new products or fuels, therefore further efforts should be devoted to the development of such technology. These publications provide important findings and insights which stimulate further investigations in pyrolysis.

The top-cited publications published since 2009 constitute literature reviews, mainly focused on the importance of pyrolysis in the management plastic recycling and advancements of plastic pyrolysis techniques. Al-Salem et al. [4] provides a detailed overview of the plastic waste recycling technologies, which can be categorized under primary (re-use), secondary (mechanical, re-extraction), tertiary (chemical), and quaternary (energy recovery) recycling. Based on their analysis of the research, development, and applications of the plastic recycling technologies at industry scale, tertiary recycling (including pyrolysis and gasification) is a sustainable solution for energy and/or material recovery from the plastic waste. Another review published by Al-Salem et al. [29] gives more attention to the kinetic modelling in the plastic pyrolysis reactions and process design for plastic waste incinerators.

A more detailed account of the main mechanisms in thermal and catalytic cracking of plastics, together with the effects of various reaction parameters on the process performance, is provided by another highly cited article by Panda et al. [30]. Due to the occurrence of various steps in random during plastic waste pyrolysis, proposal of a specific mechanism that applies for all plastic pyrolysis reactions is challenging. This work serves as an important reference of different approaches that have been utilized to model the plastic pyrolysis kinetics in different conditions (catalysts, temperature, heating rates, reactor setup, polymer/biomass mixture, etc.), which inspired different reaction mechanisms to be proposed.

Following the growing interests in plastic-to-fuel conversion, some reactor setups were developed and tested on the pyrolysis of plastics, as well as co-pyrolysis of plastic-biomass mixtures. Although Table 1 shows the most important engineering approaches employed for plastic pyrolysis in the top ten articles as relying on fixed bed reactor, other reactor types like fluidized bed reactors (FBR), batch and semi-batch reactors, and conical spouted bed reactors (CSBR) are the most frequently employed systems in laboratory settings and medium to large-scale studies. A brief analysis about the landscape of the most

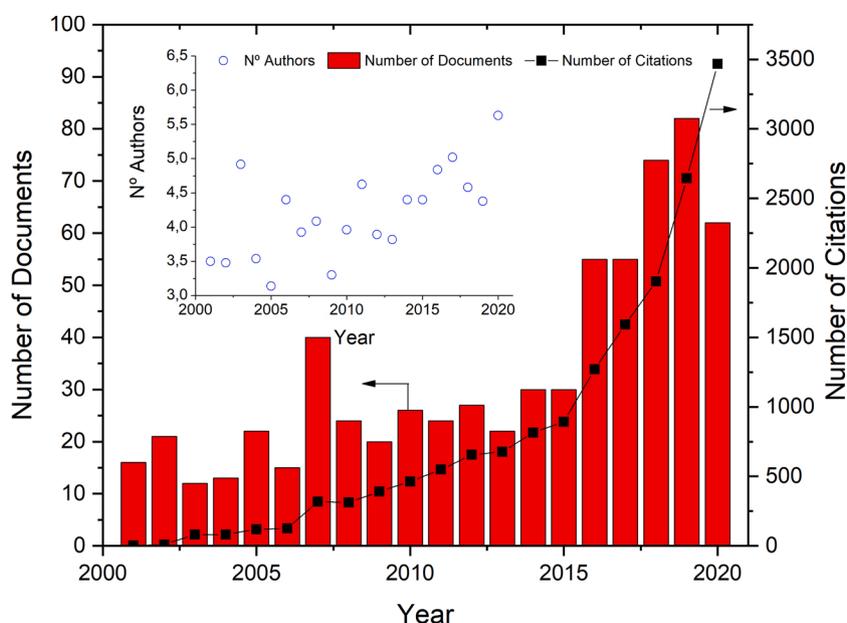


Fig. 2. Temporal trends of publications and citations related to plastic pyrolysis research. Bar graph and continuous line are associated with the left and right axis, respectively.

frequent reactors retrieved from the data analyzed here is offered in Section 4.4 together with the most recently reactors based on microwave [31] or plasma [32] systems as well.

Wong et al. [33] provide a concise summary of the performances of the various reactor and process designs used in the aforementioned processes, as well as the features of liquid fuels produced from plastics with different compositions and characteristics. The advances in analytical techniques for the investigation of plastic pyrolysis are also discussed in detail.

While pyrolysis is an effective method for plastic recycling, the pyrolysis of PVC often presents various complications. The chlorinated compounds in the polymer can be converted into hydrochloric acid, which increases the acidity of the liquid products and the probability of corroding the pyrolysis equipment. Also, the chlorinated hydrocarbons formed during the pyrolysis could be toxic, raising safety concerns related to the usage of such liquid as fuels. Yu et al. [34], offer a concise review of the product formation when PVC is pyrolyzed alone or with other polymers/biomasses. Various dechlorination technologies are also discussed, including the use of neutralization additives, alteration of pyrolysis steps and conditions, as well as pyrolysis in sub-critical/supercritical water. Torres et al. [35,36] analyzed the impact of applying several chemical removers (Na_2CO_3 -ZnO, zeolite, and aluminum, among others) into the pyrolysis or used as an external hot filter to reduce the chlorine content until 70 %wt. Given the disastrous effects related to plastic waste containing PVC, this publication serves as an important guideline for handling such waste.

Recently, Al-Salem et al. [37] shows plastic recovery via pyrolysis is the one of the most promising ways to overcome plastic waste issues. However, critical parameters such as temperature, particle size, pressure, and reactor design, among others, affect the final fuel products. Of these, reactor configurations are especially important. While fixed bed reactors are commonly examined, Al-Salem et al. in this work suggests a renewed focus on upgrading the products coming from the first reactor. Therefore, Al-Salem indicates that, while conical spouted bed reactor is a good choice to overcome traditional issues related to large particle feedstocks, microwave-assisted reactors, which are under early-stage of development, could be an effective alternative to reduce operational costs related to the energy consumption needed to heat up the systems to the desired reaction temperature.

In summary, the top-cited papers include four research papers published before 2009 and six review papers published after 2009. The first group provides evidence on the feasibility of pyrolysis for plastic-to-fuel conversion and the findings observed in these publications are verified in numerous independent studies. The second group provides researchers, policymakers, and industry partners with information about how these processes fit in the plastic recycling landscape, as well as abundant technical data for better processes design and control.

3.2. Top productive journal and journal co-citation relationship

A detailed analysis of 670 publication published in 232 indexed journals, reveal that only 10 journals have reached the threshold of

fifteen or more documents and together, these ten journals published 48 % of the pyrolysis-focused research between 2001–2020. Table 2 shows the ten most influential journals, all of them with impact factor higher than 3 ($\text{IF}_{2019} > 3.0$). According to Lei et al. [38], the distribution of articles in journals related to a research topic follows the Bradford Law Scattering. The core journals, which are defined as journals which published at least 33 % of the accumulated documents, are the first six journals in Table 2 in this case.

As can be noted from Table 2, the *Journal of Analytical and Applied Pyrolysis* (JAAP) published 85 papers on pyrolysis, more than twice publications than any other journals. The 85 papers in JAAP were cited more than 3200 times. However, normalized citations by number of papers show that *Waste Management* (WM) has been more attractive in term of citations, around 68 citations per paper in comparison to 32 citations per paper from JAAP. While JAAP is the most productive journal, WM could be considered more impactful with three of its papers in the top ten most cited papers. Due to its strong relevance to the pyrolysis subject, its usurping that JAAP publishes more articles focused on pyrolysis; meanwhile the focus of MW is more transdisciplinary, with high affinity in management subject.

The underlying relationships between journals based on cross-citations are displayed in Fig. 3. This kind of networking map is created with fractional counting methodology [39]. A total of 3184 different sources cited by the 670 retrieved articles were considered. The map is distributed into four cluster colors, the blue one composed firstly by *Journal of Analytical and Applied Pyrolysis* in the middle of the map showing its relevance with several others journals from other subjects or topics. Articles related to polymer domains are also included in this blue cluster. The red cluster shows a great transdisciplinary topic composed by journal with focused on waste management, renewable resources and among others topics, led by *Waste Management Journal*. Meanwhile, the green cluster represents journals focused on catalytic conversion of plastic waste, such as *Industrial Engineering Chemistry Research*, *Applied Catalysis B*, etc. Finally, the yellow cluster is composed by *Energy & Fuel* and *Chemical Engineering Science* and its associated topics reported.

3.3. Authors productivity and relationship

From 670 documents, there are 1864 authors involved in pyrolysis research, whereof Table 3 display the most influential authors are ranked by the number of documents published between 2001–2020 (T_p), as well divided into four time periods. The most three productive researchers overall are Marcilla, A. with 39 documents, followed by Bilbao, J. and Serrano, D. with 36 and 33 documents, respectively. Nevertheless, the productivity along the time shows that Marcilla, A. was the most relevant researcher during the first two-periods, 2001–2005, and 2006–2010, with 27 % of all documents published in that time (35 of 130 documents), followed by Serrano, DP. and Aguado, J. with 15 % across the same period. The leading author trends change in the periods, 2011–2015, and 2016–2020 as researchers such as Olazar, M. Bilbao, J. and Park YK published impactful articles. In the early stages or a new research field like pyrolysis, a few researchers

Table 2
Top ten productive journals on plastic pyrolysis research.

| Journal | T_p | T_p (%) | Cumulative percentage (T_p ,%) | N° Citation | Citation/Document | Impact Factor (2019) | h-index |
|---|-------|-----------|-----------------------------------|--------------------|-------------------|----------------------|---------|
| Journal of Analytical and Applied Pyrolysis | 85 | 12.69 | 12.69 | 3210 | 38 | 3.905 | 35 |
| Energy & Fuel | 43 | 6.42 | 19.11 | 663 | 15 | 3.421 | 15 |
| Waste Management | 28 | 4.18 | 23.28 | 1907 | 68 | 5.448 | 17 |
| Polymer Degradation and Stability | 26 | 3.88 | 27.17 | 769 | 30 | 4.032 | 18 |
| Fuel Processing Technology | 24 | 3.58 | 30.75 | 528 | 22 | 4.982 | 14 |
| Fuel | 20 | 2.99 | 33.73 | 637 | 32 | 5.578 | 14 |
| Industrial Engineering Chemistry Research | 19 | 2.84 | 36.57 | 576 | 30 | 3.573 | 10 |
| Energy Conversion and Management | 18 | 2.69 | 39.26 | 490 | 27 | 8.208 | 13 |
| Chemical Engineering Journal | 16 | 2.39 | 41.64 | 433 | 27 | 10.652 | 10 |
| Applied Catalysis A: General | 15 | 2.24 | 43.88 | 492 | 33 | 5.006 | 13 |

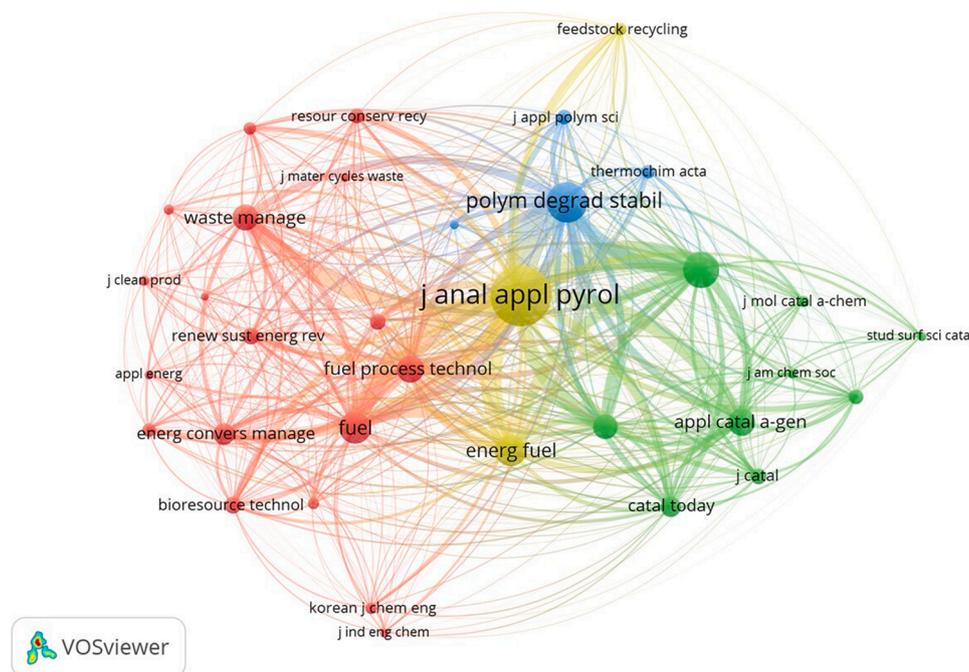


Fig. 3. Networking visualization map that illustrates the co-citation relationship of journals in plastic pyrolysis research (from 3184 journals, 35 sources meet a threshold of 100 citations a least).

Table 3

Most productive authors, most frequently employed technology, and technological readiness level (TLR) for plastic pyrolysis according to 670 articles conveying plastic pyrolysis research.

| Authors | T_p | T_p (%) | T_c | T_c/T_p | h-index | Timespan | | | | Most often Technology / TRL |
|-------------|-------|-----------|-------|-----------|---------|-----------|-----------|-----------|-----------|-----------------------------|
| | | | | | | 2001–2005 | 2006–2010 | 2011–2015 | 2016–2020 | |
| Marcilla A | 39 | 5.8 | 1001 | 25.67 | 21 | 12 | 23 | 4 | 0 | Batch / 9 |
| Bilbao J | 36 | 5.3 | 1432 | 38.78 | 22 | 5 | 10 | 14 | 7 | CSBR / 5-6 |
| Serrano DP | 33 | 4.9 | 1301 | 39.42 | 20 | 10 | 10 | 9 | 4 | FBR / 8-9 |
| Park YK | 30 | 4.4 | 418 | 13.93 | 11 | 1 | 1 | 7 | 21 | Batch / 9 |
| Olazar M | 29 | 4.3 | 1339 | 46.17 | 21 | 2 | 8 | 15 | 4 | CSBR / 5-6 |
| Aguado J | 26 | 3.8 | 1140 | 43.85 | 18 | 9 | 10 | 7 | 0 | FBR / 8-9 |
| Escola JM | 21 | 3.1 | 906 | 43.14 | 14 | 7 | 3 | 9 | 2 | RKR / 7 |
| Elordi G | 20 | 2.9 | 1022 | 51.10 | 16 | 0 | 6 | 12 | 2 | CSBR / 5-6 |
| López G | 19 | 2.8 | 1053 | 55.42 | 15 | 0 | 6 | 11 | 2 | Batch / 9 |
| Artetxe M | 16 | 2.3 | 811 | 50.69 | 13 | 0 | 2 | 12 | 2 | CSBR / 5-6 |
| Miskolczi N | 16 | 2.3 | 317 | 19.81 | 10 | 2 | 3 | 3 | 8 | Batch / 9 |

dominate the overall output. emerging researchers and teams group shows a rapid increase of number publications, which is closely related to the evolution of the total number of authors that was shown before in Fig. 2.

On the other hand, it is also possible to note researcher like Artetxe, M., Elordi, G., and López, G., shows T_c/T_p ratio bigger than the most productive authors. This strong capability to attract citation is frequently being closely linked to the top productive team and to hot research topics. In this line, these three authors match both of them, in one hand they are part of the same network and, on the other hand most of their published document as tagged as “hot paper” by WoS.

Fig. 4 show the expanded collaborative authorship network around the most productive author with 7 or more documents. As can be note, the landscape on plastic pyrolysis has been dominated by three principal team ladder by Bilbao, J., Marcilla, A., Serrano, DP., and recently new fourth incoming team lead by Park., KY. Additionally, from the network it is possible to underlying that the most influential researcher all of them are member of Bilbao’s team ($T_c/T_p > 50$, Table 3). The isolated pattern of research team without interconnection, except for a small one between Serrano, DP., and Marcilla, A., reveal a dominant research team focused on very specific topic. Others small and isolated

teams are also displayed in Fig. 4 with less impact in term of productivity than the mentioned before, but with strong relevance in term of citations as Al-Salem, SM, or López-Uriónaberrenchea, A.

Table 3, display also the readiness technology level of the most frequent reactors/technology used by these authors. It is possible to note the expertise for the most dominant research teams, is strongly focused on e.g, CSBR for Bilbao’s team, and FBR for Serrano’s team. This landscape suggests a deep understanding and development or improvement of such technology in each case, with is also noted during the analysis of data done in this work (not shown here).

3.4. Productivity and contribution among country and organization

Sixty-two countries contributed to the plastic pyrolysis research under review, with at least ten publications from fourteen countries. The top productive countries, together with their h-index and T_c , are depicted in Table 4. The numbers of total publications (T_p) produced by each country in different five-year periods are also displayed to visualize the research dynamics. The international collaborations in the form of co-authorship are displayed in an overlay visualization map (Fig. 5).

In general, all the listed countries produced between 21–135

publications, with h-indexes of 8–41 respectively. Spain is deemed the most productive country in this research topic due to the high number of total publications produced in 2010–2019. Researchers in Spain made early advances in this field, with their number of publications (29) far exceeding those of other countries for the period between 2001–2015. It is also worth noting that the articles were published in English, not Spanish, probably because the most influential journals publish just in English. This shows an implicit recognition of the global interest in pyrolysis research and that English-language publications were more likely to reach a larger audience. With an h-index of 41 and a total of 4605 citations received, Spain is also the most influential country in this research topic. Nevertheless, a decrease in the T_p is observed in 2016–2020 and a weak collaboration is observed among Spain and other countries, including Canada, Italy and Brazil, as shown in Fig. 5.

China is recognized as the second most productive country in plastic pyrolysis research. Despite the low productivity in the first five-year period, China showed increased research output in the following periods (9, 15, and 74 respectively). In 2016–2020, China emerged to be the most productive country in this topic, demonstrating the determination of the Chinese government in curbing environmental pollution (more green-yellow color displayed in Fig. 5). This strong gap in term of publications, which is directly related to new research efforts, could be associated to the implementation into 2017 of a policy in China banning importation of plastic waste [40]. Researchers in China have also worked to develop an advanced treatment for other wastes, including construction and demolition waste [41], municipal solid waste incineration ashes [42], and food waste [43]. As shown in Fig. 5, China has a close research collaboration with other productive countries listed in Table 4, including the USA, South Korea, Malaysia, and India. While publications by authors based in China have received far fewer citations than those from Spain or England, which were the earliest countries involved in this research topic. Such observation has been already pointed by Zhang and Wang [44], and related to the fact that citation accumulation takes time.

In term of organization, as can be expected, universities in Spain show the most important contributions in this field. On the top, University of Basque Country, University of Alicante, and King Juan Carlos University are the most productive, followed by Korean and Chinese

universities. Meanwhile funding agency seem Chinese institution (National Natural Science Foundation of China) is the most relevant agency, even more than Spanish one (non-shown here). In contrast to the cross-linked research programs and numerous funding grants available for European countries and universities, research funding in China seems to be concentrated in just one institution.

4. Keyword co-occurrence analysis

Keyword co-occurrence refers to the common presence of two keywords in the title, abstract, and keyword list of a publication. The larger the co-occurrence frequency of two keywords, the closer their relationship in a research topic will be. Therefore, mapping the knowledge structure of a research topic (e.g. “pyrolysis”) via keyword co-occurrence has become a popular practice to reveal the attention of researchers stretches beyond the specific topic [45]. Fig. 6 depicts a network visualization map that contain 27 keywords, each with a minimum occurrence of 31. “PE” is the keyword with the highest total link strength (2179) in the figure. This observation is attributed to the popular use of low-density polyethylene and high-density polyethylene in plastic pyrolysis research, followed by “PP” (1041) and “PS” (463). “Pyrolysis” (1854), “degradation” (1717) and “plastic waste” (1545) are the other keywords with high total link strengths, due to their relevance to the main theme of the research topic. The keywords are grouped into five categories, represented by different colors. Analysis of the co-occurrence map reveals four research hotspots, which have not been fully discussed in previously published reviews.

4.1. Research hotspot 1

The first research hotspot (indicated by Cluster I in red colour) is thermal and kinetic studies on the decomposition and degradation of different plastic types. “Pyrolysis” is the central node of this cluster. This cluster represents the researchers’ efforts in determining the kinetic triplet parameters: activation energy (E_a), pre-exponential factor (A) and reaction model [$f(\alpha)$] in plastic degradation. The results, including the numerical values of the aforementioned parameters, provide vital insights on the possible thermal/catalytic degradation mechanism of

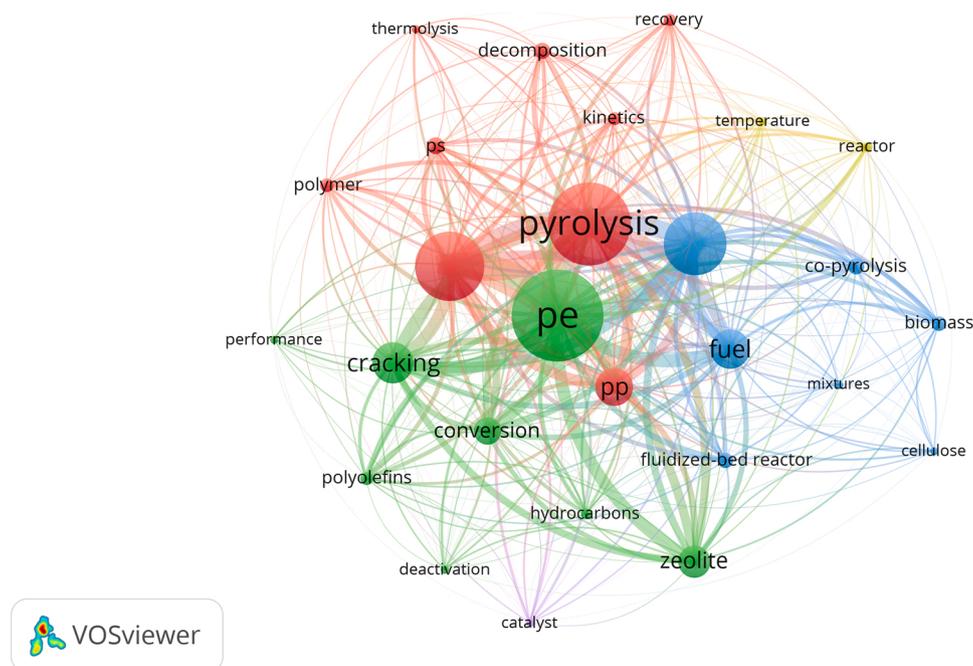


Fig. 6. Co-occurrence of keywords extracted from plastic pyrolysis research documents (minimum number of co-occurrence: 31, node size denotes total link strength).

plastic mixed with additives, impurities, or contaminants. The accurate prediction and optimization of plastic pyrolysis process relies on the precise determination of the kinetic parameters [46]. In literature, thermogravimetric analysis (TGA) is the most important analysis technique to obtain reliable data on the plastic mass loss upon temperature increase. Fitting of the TGA data and its first derivative (DTG) using model-fitting method and model-free method could then be performed to calculate the kinetic parameters with as much accuracy as a Costa et al. [47] have reported. Recently, the use of combined analysis techniques has gained increased attention, as such combinations enable extraction of more information from the plastics degradation reactions. For example, Karimpour-Motlagh et al. [48] successfully demonstrated the use of TGA-differential scanning calorimetry (DSC) (to determine the heat flux types) and TGA-Fourier-transform Infrared (FTIR) spectroscopy (to determine the characteristics of plastic degradation product gas) to investigate the effects of PP and nanoclays on the decomposition of poly(lactic acid).

The coupling of TGA-FTIR-MS is another strategy to achieve a more accurate estimation on the product/intermediate species during plastic degradation, which adds more details to the kinetic and mechanism analysis [49]. Researchers recognize the importance of determining thermodynamic parameters in plastic degradation, especially changes in Gibbs's free energy (ΔG°), enthalpy (ΔH°) and entropy (ΔS°) [50]. These parameters could be calculated based on the TGA data to reveal the feasibility of the pyrolysis process and the related heating/cooling energy requirement during the pyrolysis process [51].

Thermal and kinetic analysis on the decompositions of virgin plastics have been reported by many research teams. Some examples include analysis on linear low-density polyethylene [52], polypropylene [53] and poly(vinyl chloride) [54]. Some interesting waste materials under investigation are aluminum-containing plastic wastes, including tetrapack [55] and metallized food packaging waste [56]. Due to the high binding strengths of the aluminum-plastic layers in these waste products, efficient metal recovery using physical and mechanical methods proves to be difficult. Pyrolysis, on the other hand, conveniently converts plastic layers, adhesives, and cellulosic components into liquid products, while the metal can be recovered from the solid residue. Despite the great advantage of pyrolysis in handling the aluminum-containing plastic wastes, kinetic modelling on such decomposition presents a challenge. Yousef et al. [56] demonstrated the use of an independent parallel reaction (IPR) model to describe the simultaneous decomposition of individual components during thermal degradation of different metallized food packaging waste. Upon comparison, the authors observed high deviations between the kinetic parameters calculated using experimental kinetic data and the kinetic parameters predicted with the IPR model (7–12 %). Such deviations were related to the presence of abundant pseudo-components and pseudo-subcomponents during the waste decomposition. To solve such issue, the IPR model was modified according to the method described by Sfakiotakis et al. [57], and the deviation between the calculated and predicted kinetic parameters was decreased to 2.35 %. It is expected that the kinetic modelling of real commingled plastic waste would become more challenging following the increasingly complex compositions of modern consumer products. Nevertheless, such efforts will provide more practical insights on the application of pyrolysis in commingled plastic waste recycling.

From the analysis carried out in this work, it has been possible to note a nascent interest in the use of such computational tools like machine learning and others supported by artificial intelligence. These programs help to model and analyze the kinetics and reaction mechanism in plastic pyrolysis process [58]. This new strategy could be helpful in understanding complex plastic mixtures, where the reaction mechanisms and the kinetics involved are too complex to track with physical models.

4.2. Research hotspot 2

The second research hotspot (indicated by Cluster 2 in green in Fig. 6) refers to the investigations on the synthesis and application of catalysts in plastic pyrolysis. Thermal pyrolysis of plastic is often associated with high energy requirements and low product quality. The presence of catalysts can significantly improve plastic pyrolysis in the following ways: reduction of plastic degradation temperature and time, improved plastic conversion, enhanced liquid and gas product yield and selectivity, removal of contaminants via adsorption. Therefore, development and applications of catalysts are important elements in plastic pyrolysis research [59,60]. Commercial aluminosilicate materials such as those with mesoporous, microporous, or hierarchical structures, especially zeolites ZSM-5 [45,61], BETA [62], silica-alumina [63], and MCM-41 [64] are known to possess excellent catalytic performance for plastic pyrolysis. These catalysts enhance cracking, oligomerization, cyclization, aromatization, and isomerization reactions and have been shown to result in liquid products with high quality [65]. The catalyst acidity (denoted by Si/Al ratio) and textural properties (surface area and porosity features) are the two main factors which influence the catalytic performance. Modifications of catalyst properties have also been performed in attempts to further maximize the yield and quality of liquid products [66]. Catalyst impregnation with transition metals, including Ni and Fe, improve the aromatic compounds in the liquid products [67]. However, formation of coke on the catalyst particles during plastic pyrolysis hinders the reaction intermediates from reaching the catalyst active sites and pore systems, hence render the catalyst useless [68]. In this view, the economic viability of commercial plastic pyrolysis process is highly reliant on the zeolite cost, and the possibility of multiple catalyst regeneration with minimum deterioration on the regenerated catalyst performance. Therefore, investigations on the deactivation and regeneration of used catalysts is on-going to formulate suitable strategies to extend the catalyst lifetime [69,70].

As a response to the circular economy concept that has gained remarkable importance recently, several research teams proposed the conversion of waste materials from agricultural and industrial sectors into catalysts for plastic pyrolysis. Such an innovative idea is expected to reduce the burden of waste management sector, while significantly reducing the material cost in plastic recycling process. Coal fly ash (CFA) is an example of waste generated from coal-fired power plants. Although application of such ash in cement production is possible, the abundance of fly ash and bottom ash from other sources [42] limit the total amount of CFA recycled via this route. Due to the high alumina/silica content, CFA can be converted to zeolites for various applications. Cocchi et al. [71] demonstrated the application of CFA and CFA-derived zeolites in pyrolysis of plastic waste (PE films mixed with 2–5 % PP). Thermal pyrolysis at 723 K produced 70 wt.% of condensable fraction (which mainly consisted of tar, wax, and oil), 25 wt.% of gas product, and ~5 wt.% of char. Nevertheless, only 5 wt.% of condensable fraction resulted in oil formation in the cold trap, while the tar and wax were trapped in the reactor during the reaction. Such an observation is attributed to the relatively low pyrolysis temperature when compared to the maximum decomposition temperature of the plastic waste (753 K). The use of CFA had little effect on the reduction of plastic degradation temperature (753 K vs 680 K), but drastically increased the oil obtained into the cold trap from 5.0 wt.% (thermal pyrolysis) to 36.2 wt.%. In comparison, the use of CFA-derived zeolite effectively reduced the plastic degradation temperature from 753 K (thermal pyrolysis) to 680 K, and increased the oil yield further to 44 wt.%.

Following the development of polymer science, PVC and plastics containing brominated flame retardants have been widely applied in human society. However, the presence of halogen atoms in such plastic waste can hinder the recycling via pyrolysis, as the use of pyrolysis oil containing halogenated compounds as fuel causes engine corrosion and air pollution [72]. In addition, generation of HCl during pyrolysis of PVC is a known cause of pyrolysis system deterioration [34]. Thus,

development of a pyrolysis system which can minimize the adverse impacts of halogenated plastics is an important branch in plastic pyrolysis research. Removal of halogenated compounds by calcium-based compounds has been identified as a critical strategy to minimize the presence of halogenated moieties in the reaction intermediates and oil product [34]. CaO also effectively promotes deoxygenation reactions in pyrolysis of PET [73] and biomass [68]. In a multistep co-pyrolysis of a PVC/cellulose mixture performed by Sophorant et al. [74], CaO exhibited excellent performance in dehydrohalogenation from PVC degradation and reforming of biomass pyrolysis products.

Such observations validate the versatility of CaO in plastic and biomass pyrolysis, as well as co-pyrolysis of the mixture. Lim et al. [75] attempted the use of crushed concrete waste (which is rich in CaO) in PET pyrolysis. The authors reported negligible catalytic effect on the maximum decomposition temperature; however, effective neutralization of decarboxylated compounds led to increased aromatic content in the liquid product. Another study performed by Hussain et al. [76] demonstrated the marginal improvement of liquid and gaseous products yields and qualities in PS pyrolysis over burnt brick powder, due to the high Al₂O₃, SiO₂ and CaO contents in these catalysts. These findings could mark the potential of CFA and concrete/cement waste as sources of low-cost catalyst.

Due to its textural, physical, and chemical properties, activated carbon [77] and biochar [78] derived from lignocellulosic materials can be used for various applications, especially in environmental remediation. The catalytic effects of activated carbon and biochar (abbreviated as AC/B) in plastic pyrolysis are recently reported. For example, Li et al. [79] reported the pyrolysis of LDPE/HDPE mixtures over biochar synthesized via poplar wood gasification. The biochar was able to promote cracking of LDPE degradation intermediates to form more gaseous product. However, such effect was not observed in HDPE pyrolysis, evidenced by the wax formation. Conversion of waste plastics to jet fuels (C₈-C₁₆ hydrocarbons) over commercial activated carbons was also reported [80]. Such an excellent pyrolysis performance was related to the high catalyst acidity (due to the phosphoric acid activation), high BET surface area and well-defined pore distribution. Based on these research findings, these authors proposed the use of catalysts derived from lignocellulosic waste as low-cost and renewable catalysts in plastics pyrolysis.

However, it is possible that these authors did not consider the possible roles of AC/B as sources of cellulose and lignin, as pointed out by Xue et al. [81]. In this view, the plastic pyrolysis in the presence of AC/B could have similar process mechanism to that of co-pyrolysis of plastic/bio-waste mixture (which is discussed in Cluster 3), where AC/B also act as reactant. It remains necessary to examine the structural integrity of used AC/B after plastic pyrolysis. According to Li et al. [79], used biochar possessed higher crystallinity, and was less susceptible to temperature-programmed oxidation. The oxidation process for the used biochar samples also occurred at higher temperatures when compared to the fresh sample. TGA analysis results indicated lower mass loss percentages for all the used catalysts when compared to the fresh catalyst. The evolution of CO₂ gas from fresh and used biochars indicates oxidation of it together with the coke. All these results could indicate the partial degradation of the biochar (amorphous) structure due to the participation in plastic pyrolysis mechanism. If this is the case, it will be necessary to develop methods that can accurately quantify the mass loss due to the structural degradation and mass gain due to the coke formation on the catalysts. As coke and the AC/B are all carbonaceous in nature, thermal regeneration of used catalysts could be challenging. More research data on the structural integrity of used AC/B is needed to provide further insights on the plastic pyrolysis mechanism over AC/B, and the regeneration/reuse of the AC/B.

4.3. Research hotspot 3

The third research hotspot (indicated by Cluster 3 in blue colour)

represents the researchers' explorations on co-pyrolysis of biomass and plastics. Following the increasing attentions on the climate change mitigation, conversion of biomass to renewable biofuels is regarded as an effective strategy to reduce the carbon footprint related to the use of fossil fuels. Thermochemical conversion, especially pyrolysis, is a technically feasible route for such conversion. However, this process suffers from low yield of liquid product with high oxygenated compound content, and hence high acidity. Catalytic pyrolysis improves the liquid yield, but the tendency of catalyst coking presents another challenge [82]. These disadvantages are related to the high carbon content and low hydrogen content in the biomass. This challenge could be solved by co-feeding plastics (with high hydrogen content) and lignocellulosic biomass in the pyrolysis [83]. Such practice increases the effective carbon to hydrogen ratio, leading to reduced coke formation and increased aromatic hydrocarbon yield. Despite the viability of co-pyrolysis in the absence of catalysts [84], catalytic co-pyrolysis is more advantageous due to the ability of zeolites (especially ZSM-5) in catalyzing degradation of plastics and biomass. The distinctive degradation mechanisms of these components give rise to the synergistic effects on aromatics formation. The participation of the volatile intermediates from the plastic and biomass degradation resulted in complicated reaction pathways in series and also in parallel. Also, catalyst properties influence the reactions of these intermediates on the active sites. Therefore, the reaction mechanism for co-pyrolysis could be unique for each plastic/biomass/catalyst system.

To achieve co-pyrolysis, co-feeding of plastic waste and biomass into the pyrolysis unit is a commonly used strategy [85]. However, this can contribute to a lack of synergistic effect in thermal co-pyrolysis of PE/cellulose mixture. The addition of HZSM-5 catalyst significantly promotes such effects, leading to high aromatic yield. Significant synergistic effects on the formation of aromatic hydrocarbons were also observed in catalytic co-pyrolysis of various biomass-plastic combination, including waste plastic films/*Quercus variabilis* [86], cellulose/LLDPE [87] and LDPE/walnut shell [88]. Yuan et al. [89] used MgO and MgCO₃ catalyst in co-pyrolysis for a PVC/rice husk mixture. Lower contents of acids (<2%), alcohols and phenols in the bio-oil were observed together with higher hydrocarbon contents, presumably due to the effects of basic catalysts during the conversion process. By combining a two-step catalytic pyrolysis process with a one-step catalytic co-pyrolysis process, it was possible to convert LDPE/walnut mixture to liquid product with high aromatic content (82.5 %) and low oxygenated compounds selectivity (<1%). The key lies in the prevention of secondary reactions by controlling the decomposition temperatures at different steps [88].

Many consumer goods are made of a plastic-wood combination as these materials often complement each other. Convenient valorization of these products at the end of their use presents an additional motivation to develop co-pyrolysis technology. Wood-plastic composite (WPC), for example, is a combination of wood fibers and thermoplastics, which have high moisture resistance and rot-resistance. Therefore, such material is widely used in outdoor deck floors and park benches. Park K. B et al. [90] reported efficient conversion of WPC over zeolites to aromatic hydrocarbons in a tandem micro-reactor-gas chromatography/mass spectrometry. H-BETA zeolite displayed higher catalytic efficiency in aromatics formation when compared with H-ZSM5 and H-Y zeolites. Similar observations were also made by Lin et al. [91] in catalytic co-pyrolysis of WPC using pyrolysis-gas chromatography/mass spectrometry. The author group also noted the effects of zeolite porosity and acidity on the product distribution. The research findings from these research groups demonstrate the feasibility of WPC waste valorization via co-pyrolysis.

Overall, the recent works on co-pyrolysis focus on quantifying the synergistic effects for different plastic/biomass mixture, as well as careful design of process/system conditions to maximize the yield and quality of bio-oil for further exploitations. The use of algal biomass [92], and carbonaceous consumer waste [93] in co-pyrolysis process open

more paths for renewable biofuels production in addition to innovative waste management practices.

4.4. Research hotspot 4

The fourth research hotspot (indicated by Cluster 4 in yellow) refers to studies of reactor designs and reaction parameters for plastic pyrolysis. Many different reactors systems have been utilized in plastic pyrolysis research, including, but not limited to: batch/semibatch reactor [94], fixed bed reactor [95], rotary kiln reactor (RKR) [96], conical spouted bed reactor [97], and multi-staged reactors [98]. The effects of reaction parameters (temperature, ratio of catalyst to polymer, residence time etc.) on plastic pyrolysis process behavior, as well as product yields and compositions in the aforementioned reactor systems have also been extensively investigated. Basically, the reactor design and the reaction parameters affect two fundamental criteria in plastic pyrolysis: 1) effective heat transfer from the reaction environment to the catalyst/plastic system, and 2) effective interactions between the plastic decomposition moieties with the catalyst particles. Readers are referred to the recent reviews produced by Anuar et al. [5] and Murthy et al. [99] on these topics.

The auger reactor, also known as screw reactor, has also shown great potential in the pyrolysis of biomass and waste materials. By varying the screw speed, it is possible to control the residence time of feed materials in the reactor, hence the type of pyrolysis (e.g. slow, intermediate, or fast). The ease in mass and heat transfer control in such reactor also makes it a popular choice in pyrolysis research [100]. An interesting concept proposed by Park et al. [101] is “activator-assisted pyrolysis,” during which plastics could be “activated” prior to pyrolysis. In their work, virgin PP powder was continuously fed into an auger reactor (25–300 °C) before entering a fluidized bed reactor connected in series (bubbling zone temperature: 600–730 °C). Polymer melting with a minimal degree of degradation was observed upon an increase of auger reactor temperature to 300 °C, and hence the plastic pyrolysis only took place in the fluidized bed reactor. The authors observed a significant increase in gas yield (from 68–80 wt.%, at the expense of liquid yield), together with a significant production of H₂ and methane (at the expense of ethene and propene yields) in the gas product upon the increase in auger reactor temperature. Kinetic assessment results revealed decreased activation energy of PP after being heated to 300 °C in the auger reactor. Based on such observations, the authors proposed that the heating step in the auger reactor weakened the C–C and C–H bond strengths in the plastic, leading to higher degree of bond scissions along the polymer chains during the pyrolysis process. Such an innovative work provides new evidence of the effects of heating pretreatment on the degradation behavior of plastic. Whether such an “activation” effect only takes place in an auger reactor or if it might be useful in other reactor types with controllable residence time should be further investigated.

Most plastic pyrolysis is conducted in the absence of oxygen to prevent combustion. N₂ or Ar are normally used to ensure an inert environment in the pyrolysis reactor. Nevertheless, scientists are now evaluating the effectiveness of biomass and plastic pyrolysis in CO₂ environment, as a strategy of CO₂ utilization [102]. According to Kwon et al. [103], formation of dangerous hydrocarbon compounds during waste feedstock pyrolysis can be effectively suppressed in a CO₂ environment, due to the elevated degree of volatile species cracking. When applied in thermal pyrolysis of PVC, the CO₂ environment restrained the formation of benzene derivatives [104]. In addition, CO₂ molecules also acts as carbon scavenger/oxidant during the pyrolysis process, leading to CO formation. Considering the appreciable H₂ production during plastic pyrolysis, Choi et al. [105] proposed the possibility of syngas production from plastic pyrolysis, where the H₂/CO ratio in the product gas can be controlled via manipulating CO₂ gas input. Another work by Jung et al. [106] showed that the presence of Ni-based catalyst increased the H₂ production further during PE pyrolysis in an N₂ environment,

nevertheless quick catalyst deactivation was observed due to the coke formation. Such issue can be tackled by performing the pyrolysis reaction in the CO₂ atmosphere, where the coke was oxidized to form CO and light hydrocarbons. In other words, in-situ catalyst regeneration occurred, leading to prolonged catalyst lifetime and hence H₂ production. These findings demonstrate the possibility of H₂ or syngas production from catalytic pyrolysis of plastic waste in a CO₂ atmosphere, and more studies are needed to verify such potential.

To illustrate the number of studies conducted using different reactor types, a density visualization map (Fig. 7) was generated using VOS-Viewer based on 601 of the articles in our dataset. It is noted that the fluidized bed reactors (represented by the intense red color) are the most studied reactor type with 45 % (270 documents), followed by batch or semi-batch reactors with 20 % (118 documents), and conical spouted bed reactors featured in 15 % of all analyzed documents. Other systems such as microwave [107] or plasma reactors [108] allow for process intensification and the possibility to improve heat and mass transfer phenomena, thus offering better control of kinetics and reducing side-product formation. Moreover, it is possible to highlight two important trends from the analyzed data. On one hand, the scientific production related to fluidized bed reactors is widely distributed among different authors. This could signify that knowledge related to fluidized bed technologies that were initially generated by the petrochemical industries has been recently adopted by scientists interested in plastic conversion. On the other hand, the advances in conical spouted bed reactor seem to be almost entirely made by Bilbao’s team, firstly driven by their experience on biomass pyrolysis and their efforts related to overcoming the limitation of fluidized bed reactors related to handling irregular solid feeding [109]. Finally, batch or semi-batch reactors have been commonly used to carry out proof-of-concept studies at laboratory scale. While such reactors can be especially dedicated to screening of catalysts and feedstocks, they offer modest progress for technological development.

Although pyrolysis is a well-investigated technology which can offer a strategic opportunity to tune-up process parameters to achieve different goals (waste-to-fuel, waste-to-chemicals, etc.), only few potential solutions have achieved a high level of readiness (TRLs) and meet the ambitious goals around plastic circularity. As noted in Table 3, the readiness level of technology are under development. In its current form, the batch reactor may be considered a mature technology, but its high operational cost limits its global proliferation. A newer generation of batch feed system could lower costs—for a more thorough analysis of batch reactor potentials readers are invited to follow [110].

Based on data analyzed in this work, pyrolysis using fluidized bed reactor offers the highest TRL, with most of the studies on this technology conducted at laboratory or pilot plant scales. However, plastic pyrolysis using fluidized bed reactors could be hampered by the high costs associated with the pre-treatment process, catalyst stability, and continuous feeding of solid plastic. Meanwhile, Solis [6] and by Punkkinen [111] suggest thermal cracking and catalytic cracking have a TRL of 8-9 and, according to a recent report of the European Technology Platform for Sustainable Chemistry [112], the overall TRLs of thermochemical routes for plastic conversion are currently at TRL 6-7. Therefore, we expect more studies and progress on the readiness level of fluidized bed and conical spouted bed reactors in the near future.

5. Limitations

All articles filtered and selected for this analysis and review were retrieved from WoS database, which could neglect some influential documents. Additionally, documents related to the pyrolysis of tire, rubber or urban solid waste and combinations are not considered in this study. The search query was designed with the intention to recover all the scientific documents closely related to the subject addressed; however, the authors know that is possible that several influential articles or journals could have been inadvertently ignored. Also, a deeper analysis

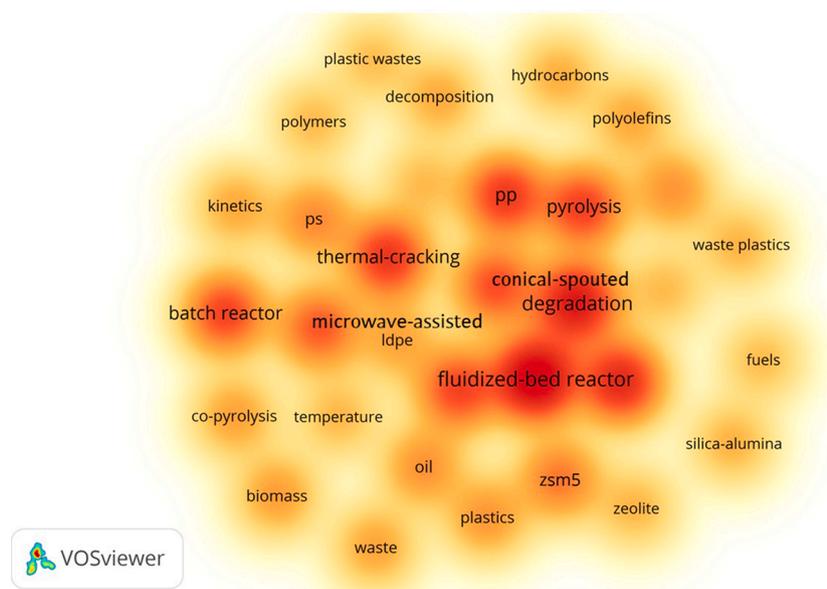


Fig. 7. Density visualization map of the most employed reactors for plastic pyrolysis during elaborated considering 601 research papers from 670 documents (from 951 matches only the first 33 co-occurrence keywords are displayed).

including other databases such as Scopus, Google Scholar could be useful to give a better compression and reinforce the findings highlighted in this paper.

6. Conclusions and coming challenge

Accelerating the circular economy around plastic materials will be critical in decreasing the impact of plastic waste into the environment. Despite the great promise of plastic pyrolysis, several bottlenecks still limit its industrial scaling and worldwide adoption. Bottlenecks include the high energy input needed to achieve complete plastic conversion, the relatively poor understanding of kinetic reactions and corresponding mechanisms, and limited results in terms of real waste experiments. Scientists and engineers will need to address these bottlenecks soon. Nevertheless, the analysis and review of twenty years of published research suggests that thermochemical routes will continue being a focal point or “hot topic” in coming years.

The landscape of core journals shows that Journal of Analytical and Applied Pyrolysis is the most influential journal related to plastic pyrolysis, publishing twice as many articles on this subject than other journals. The underlying network of the total publications shows only a small proportion of the plastic pyrolysis work is carried out by collaboration between international institutions. Researchers based in Spain have been the most productive in this field, but researchers based in China are making increasingly impactful contributions, possibly pushed by new policies related to plastic management. In the near future, the balance of research productivity may shift.

The analysis of keyword co-occurrence revealed four research hotspots in plastic pyrolysis research: thermal and kinetic analysis on plastic degradation, as well as a nascent strategy based on machine learning to analyze the kinetics and reaction mechanism for complex feedstock (hotspot 1), explorations on the effects of various catalysts on the pyrolysis performance (hotspot 2), co-pyrolysis of biomass and plastics (hotspot 3) and explorations on the effects of reactor designs and process parameters on pyrolysis performance (hotspot 4). Further investigations on the extracted publications revealed the emergence of several scientific interests under these hotspots, which will move the frontier of plastic pyrolysis research forward.

Finally, the authors consider that the transition from a linear to circular economy around plastic must resolve a central concern: How fast and how far can nations and industries adopt and scale new

pyrolysis technologies and techniques? The answer is no simple matter. Current infrastructure and technologies are limited in their ability to select and transform all of the diverse types of plastics into high-value feedstock's. Investment in applied research and the construction of new facilities is critical, but we must also develop policies that improve the current value chain for plastics and retrofit existing infrastructures to manage and separate plastic waste.

Author contributions statements

Sabino Armenise: Conceptualization, Writing - Original Draft.
 SyieLuing Wong: Methodology – Formal analysis-investigation.
 José M. Ramírez-Velásquez: Validation.
 Franck Launay: Writing - Review.
 Daniel Wuebben: Final Editing.
 Norzita Ngdi: Visualization.
 Joaquin Rams: Funding acquisition & Review.
 Marta Muñoz: Supervision.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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