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REVIEW

Upcycling of plastic waste to carbon nanomaterials: a bibliometric analysis (2000–2019)

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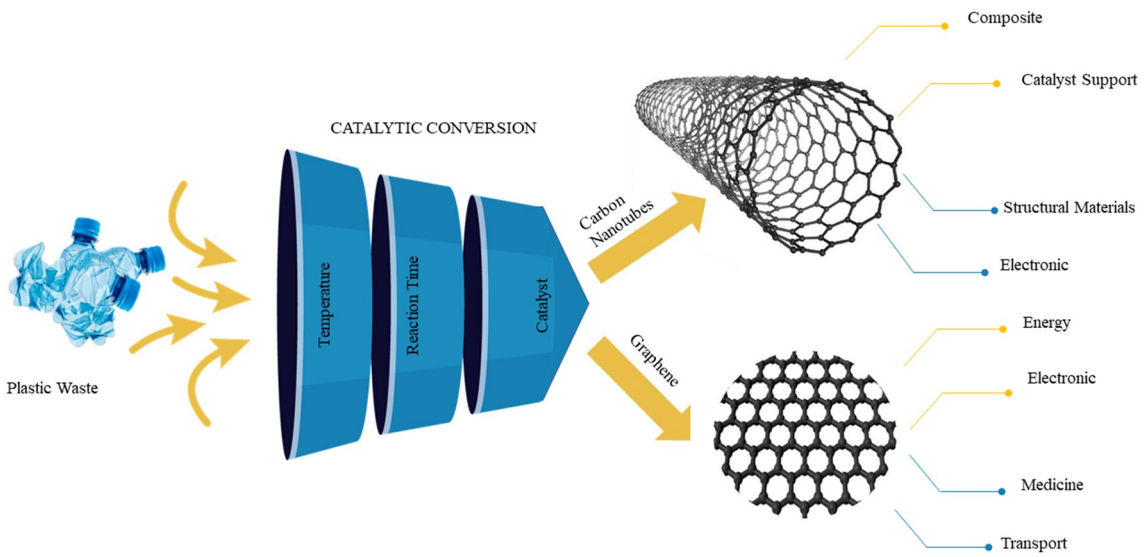
Abstract

Due to the rapidly escalating generation of plastic wastes, the development of an effective management strategy is vital to reduce their adverse effects on the environment. The conversion of plastic waste into carbon nanomaterials is a highly viable waste management solution. The growing number of scientific works depicts the potential of the technology for the low-cost synthesis of carbon nanomaterials. However, a detailed examination of developments on the topic is lacking in the literature. Therefore, this paper aims to provide an overview of the publication landscape and knowledge structure for research on plastic conversion into carbon nanomaterials. A bibliometric analysis was performed on 120 publications extracted from the Web of Science. Analysis revealed an increase in the number of publications from 2000 to 2019 with China identified as the most significant contributor. Pyrolysis coupled with chemical vapour deposition is the most widely adopted strategy for plastic waste conversion into carbon nanomaterials, due to the ease in catalyst and reaction conditions control. Most research teams reported successful synthesis of multi-walled carbon nanotubes, graphene, nanoparticles, nanowires and nanoplatelets. Reproducible synthesis of carbon nanomaterials, especially tunable carbon nanotubes, remains a major challenge. This paper provides important guidelines for researchers, funders and policymakers who are key players to develop ambitious projects on the synthesis of carbon nanomaterials from plastics as low-cost carbon precursors.

Graphical abstract



Keywords Pyrolysis · Epitaxial growth · Template-assisted growth · Carbon nanospheres · Carbon nanowires · Carbon nanostructures

Extended author information available on the last page of the article

Introduction

Humanity is now facing an unprecedented plastic waste crisis following the excessive use of single-use plastics, especially food packaging and biomedical waste. After short-term use, such plastics are discarded, thereby occupying significant space in landfills. The improper management of solid waste also leads to leakage of plastic wastes in large quantities into the environment. In the marine environment, such waste degrades into micro-/nanoplastics, which cause detrimental impacts to the terrestrial and marine ecosystems (Wong et al. 2020). Due to the omnipresence of micro-/nanoplastics and the irreversible impacts these materials exert on the global ecosystem, scientists now consider marine plastic pollution as a planetary boundary threat (Villarrubia-Gómez et al. 2018). To tackle such menace, various actions from the governments worldwide were put in place to combat the plastic waste crisis. The European Commission proposes transition from a linear economy (where the plastic products are produced, used and discarded) to a circular economy, where disposed plastic products are re-used or recycled, retaining the functional values of the products (Crippa et al. 2019). In such an economy, the end-of-life plastics are regarded as resource materials, which can be recovered, processed and fed back to the economy. This approach could greatly reduce the consumption of non-renewable feedstock by the plastic industry.

Various waste recycling technologies, including mechanical and chemical recycling, are well established at industrial scale to convert cleaned post-consumer plastic waste into value-added products. On the other hand, pyrolysis has been identified as a key solution for the thermochemical conversion of non-recyclable plastic waste into liquid fuels and chemical feedstocks, which can be utilised to produce new plastics (Kremer et al. 2020). Pyrolysis plays an important part in decoupling plastic production from fossil feedstock consumption as envisioned by Ellen Macarthur Foundation (2017) in the New Plastics Economy. Numerous scientific studies demonstrate the technical viability of plastic waste pyrolysis at laboratory and pilot scales (Armenise et al. 2021; Kizza et al. 2021). Nevertheless, the economic viability of large-scale plastic recycling via pyrolysis is questionable, due to the competition of the pyrolysis products with the fossil fuels. It is predicted that legislative supports and financial incentives from governments are necessary to enhance the role of the pyrolysis technology in the transition to the new plastic economy (Horvath et al. 2018).

Recently, there has been a growing interest in the conversion of plastic waste into carbon nanomaterials (CNM) (especially graphene and carbon nanotubes) and high-purity hydrogen gas (Liu et al. 2021). Graphene is a carbon allotrope where a single layer of carbon atoms is arranged in a two-dimensional honeycomb lattice, while CNT refers to tubes made of carbon atoms with diameters typically measured in nanometres. Due to the exceptional chemical, mechanical, tensile, textural, optical and electronic properties, CNM are being used in material reinforcement, electronics and sensors, energy storage, as well as environmental remediation (Mashkoor et al. 2020), and the list is expanding. The global graphene and CNT markets are valued at USD 87.5 million and USD 2.6 billion in 2019 and are expected to reach USD 876.8 million and USD 5.8 billion by 2027. Such increases translate to compound annual growth rates of 40.2% and 10.7% for these materials (Choudhary and Prasad 2020; Narune et al. 2021). As CNMs are mainly synthesized from fossil-based feedstock (mainly methane and ethylene), the increasing production of these materials at a large scale will lead to significant increase of carbon footprints. Besides, the high material cost and energy consumption during the CNM synthesis are the main reason for the high market price (from $100 to $100,000 per kilogram). Thus, scientists are searching for alternative low-cost feedstocks which enables a greener CNM synthesis process. There are a growing number of studies which demonstrate successful conversion of various agricultural by-products and industrial waste into graphene and CNT (Williams 2021). Plastic waste is regarded as the ideal synthesis feedstock, as the clean waste contains mostly carbon and hydrogen atoms, with only trace amount of other impurities. With proper pre-treatment, plastic waste can be converted to CNM and hydrogen gas, which is another valuable feedstock for chemical processes. As the real plastic waste is often heterogeneous and highly contaminated, significant costs in cleaning, sorting and size reduction of such plastics are vital to ensure high quality of the products. The high market CNM prices guarantee the profitability of the plastic recycling technology even when the pre-treatment cost is included. This is considered a significant advantage of the new recycling technology compared to the existing one, where plastic waste is converted into liquid fuels. The increasing amount of plastic waste generation worldwide also makes it an ideal material for long-term replacement of the traditional feedstock in CNM synthesis, without geographical area limitation. Such a conversion process is also considered an upcycling process, which retains the material value of the plastic waste in the economy. In view of these benefits, research and development on conversion of plastic into carbon nanomaterials (termed PtCNM in this paper) will become an important strategy to achieve a circular economy of plastics.

Despite a growing number of scientific publications that demonstrate the feasibility of CNM synthesis from plastic

waste, the publication landscape of PtCNM research has not been investigated. Therefore, the researchers, policymakers and funders who wish to contribute to the future development of this research topic face difficulties in identifying the current state of the technology development, especially the significant contributors (country/organisation/individual), the publication outlets, funding supports, etc. To address this knowledge gap, a bibliometric analysis is performed in this study based on 120 publications extracted from the Web of Science (WoS) to describe the important features of PtCNM research in 2000–2019. Bibliometric analysis is a popular technique for revealing the publication landscapes of well-defined research topics. In this study, the bibliometric analysis is performed to answer the following questions: (1) what are the temporal publication and citation trends in the field? (2) what are the core journals in this field? (3) what are the characteristics of the top-cited publications in this field? (4) what/who are the top productive countries, organisations or authors in this field? The scientific contents of the publications extracted from WoS were also analysed to provide readers with more details on the PtCNM research development.

The findings in this paper contribute to the PtCNM research in multiple ways. Firstly, this study provides a visualisation of the current research landscape with research hot spots that demand further investigations by the field experts. Secondly, scientists interested in the further development of PtCNM research could identify the top productive researchers for collaborations. The analysis results in this paper also provide directions for policymaking and funding decisions that could influence the pace of transition into a circular economy of plastics.

Methodology

Retrieval of publications

WoS is one of the most comprehensive scientific databases, citation indexing services and search engines when compared to Scopus, Science Direct and Google Scholar. WoS contains over 34,000 journals, conference proceedings, books, patents and data set records that span different disciplines such as science, engineering, social sciences and humanities. The published articles in WoS are indexed specifically by date, location, institution, document type and discipline, which enable researchers to trace the flow of research ideas development. Therefore, WoS was used as an important information source in bibliometric analyses on various research topics. The publication landscape of PtCNM research was also performed using WoS in this study.

On 20 August 2020, an advanced search was conducted in WoS using a carefully designed search string shown in Fig. 1 to retrieve all the related publications in 2000–2019. The search string constitutes two components: (1) all the

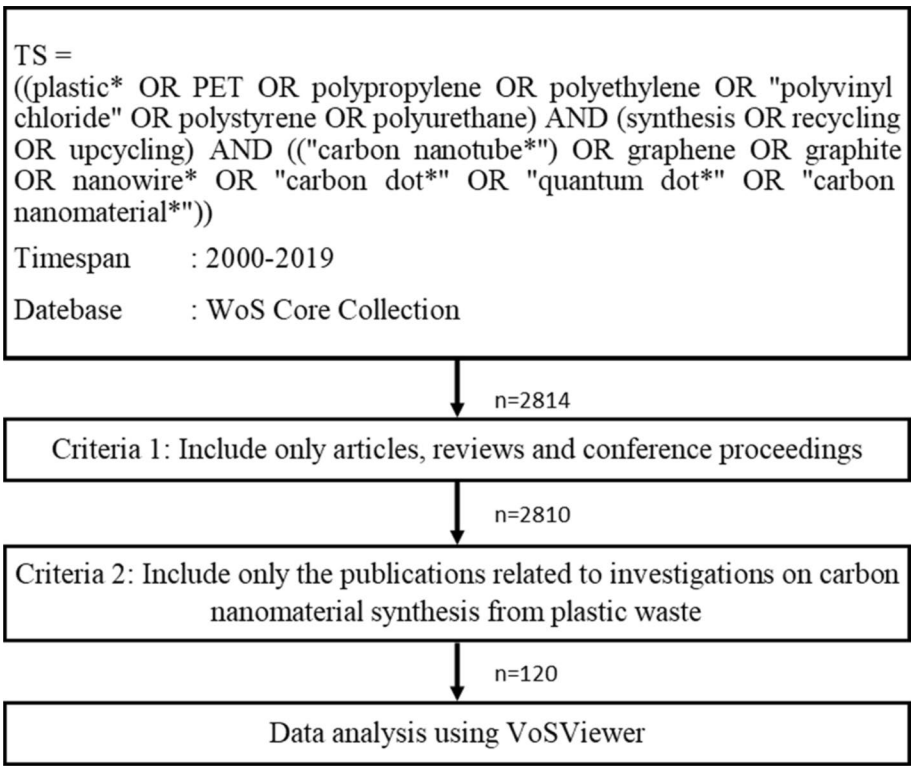


Fig. 1 Search strategy implemented to obtain relevant publications for bibliometric analysis (n denotes the number of publications at the different filtering stages)

plastic types used in the conversion process, and (2) the type of CNMs that could be formed from the conversion process. The asterisk symbols were used in the search string to include all terms with the same root. The timespan (2000–2019) was used to allow the comparisons on productivities from different countries/organisations/authors in four five-year periods (2000–2004, 2005–2009, 2010–2014, 2015–2019), which is a commonly used technique in other bibliometric analyses (Meseguer-Sánchez et al. 2021). A total of 2814 results were identified by the WoS database based on the given search string. The titles and abstracts of these results were further examined, and the publications that did not fulfil the eligibility criteria described in Fig. 1 were eliminated from the dataset. Finally, a total of 120 publications that provide information on the following aspects were selected for further analysis:

- Plastic waste categories such as polyethylene terephthalate (PET), polypropylene (PP), polyethylene (PE), poly(vinyl chloride) (PVC), polystyrene (PS) and polyurethane (PU).
- Synthesis techniques for CNMs from plastic waste such as chemical vapour deposition (CVD), arc discharge, combustion and pyrolysis.
- Assessment of the technical feasibility and/or economic potential of the technologies/conversion methods.
- CNMs including but not limited to graphene, carbon nanotubes (CNT), carbon dots, carbon nanowires, carbon spheres, carbon nanosheets, carbon nanocomposites, carbon nanoplatelets, microdiamonds, carbon nanocapsule, nanocrystals and carbon nanoparticles.

Data analysis

The related information or data from the 120 publications were exported from the WoS into a Microsoft Excel file. The downloaded information for each publication includes title, document type, source title, publication date, keywords, DOI, total citation (TC) together with the names and affiliations of the authors. The Journal Citation Report (JCR) for 2019 was retrieved to obtain relevant data of the journals where the articles were published. The impact factors (IF) and quartiles (Q) of the journals were also extracted and analysed. The IF is the ratio of the collective sum of citations obtained by a journal for the preceding two years to the entire number of citable materials published during a similar period (Wong et al. 2021). The Hirsch index (h-index) is another important metric that shows the impact of publications by any author, institution or nation. An author with an h-index of N has N publications each having at least N citations, which is a measure of the impact research work in a particular field.

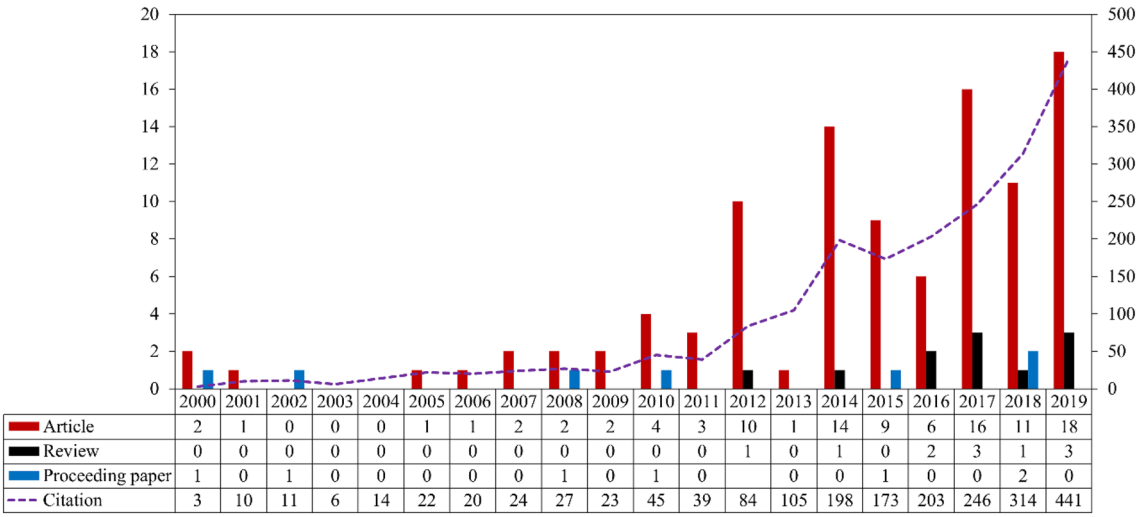
The VOSviewer software is a visualisation tool used to develop diagrammatic representations of the similarities between different objects including journals, countries, organisations and authors. In this study, VOSviewer was used to conduct the social network and keyword co-occurrence analyses based on the data exported from WoS. Specifically, the data were used to generate visualisation maps that illustrate the co-authorship, journal co-citation and keyword co-occurrence relationships in PtCNM research. The size of a node in the map demonstrates the importance of the representative objects in the map, while the thickness of a line connecting two nodes represents the strength of interactions between the two objects.

Results and discussion

Publication and citation trend

The publication trend of PtCNM research is reviewed from 2000 to 2019, which covers 2 decades. The publications on this topic comprise 103 articles (85.83%), 10 reviews (8.33%) and 7 proceedings papers (5.83%). While researchers from many non-Anglophone countries like China, India, Japan and Egypt are involved in this research topic, English is the most preferred language used in the retrieved dataset from WoS.

As shown in Fig. 2, the number of publications that report the conversion of plastic waste to CNM was relatively consistent from 2000 to 2009 averaging at least 1 to 2 publications every year. The time from 2003 to 2004 is considered the lag phase of the development of the research topic as no publications were indexed in WoS. From 2009 to 2010, the number of publications and total citations surged by about 100% with the numbers increasing from 2 to 4 and 23 to 45, respectively. Similarly, for the next three years (2010–2012), the publications increased from 4 to 10, while the citations increased from 45 to 84. Such observation could be related to the global realisation of the plastic waste problem and its potential as a carbon source for CNMs synthesis. Furthermore, the crucial work of Bazargan and McKay (2012) entitled, 'A review—Synthesis of carbon nanotubes from plastic wastes' could be the turning point of global researchers' attention to plastic waste utilisation for CNMs synthesis. However, only one article was published in 2013, probably due to ongoing experiments and write-up delays, which affected the publication period. After the year 2013, the term 'plastic waste' became more commonly used to highlight the potential routes for plastic waste management in publications. The publication dataset received a total of 2008 citations (average number of citations per publication: 16.7), indicating the interest of the scientific community on this topic.



	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	2011	2012	2013	2014	2015	2016	2017	2018	2019
Article	2	1	0	0	0	1	1	2	2	2	4	3	10	1	14	9	6	16	11	18
Review	0	0	0	0	0	0	0	0	0	0	0	0	1	0	1	0	2	3	1	3
Proceeding paper	1	0	1	0	0	0	0	0	1	0	1	0		0	0	1	0	0	2	0
Citation	3	10	11	6	14	22	20	24	27	23	45	39	84	105	198	173	203	246	314	441

Fig. 2 Publication and citation trends for research on CNMs synthesis from plastic waste

Table 1 Major research areas for CNMs from plastic waste from 2000 to 2019

Research areas	Record count	% of 120
Chemistry	59	49.17
Materials science	45	37.50
Engineering	36	30.00
Physics	23	19.17
Science technology other topics	23	19.17
Energy fuels	12	10.00
Environmental sciences Ecology	9	7.50
Polymer science	9	7.50
Electrochemistry	4	3.33
Biotechnology applied microbiology	2	1.67
Crystallography	1	0.83
Geology	1	0.83
Meteorology atmospheric sciences	1	0.83
Pharmacology pharmacy	1	0.83
Toxicology	1	0.83

As shown by the WoS database, the development in PtCNM research covers 15 major research areas. Most of the publications are categorised under chemistry (59, 49.17%), followed by Material Science (45, 37.50%) and Engineering (36. 30.00%). The strong focus on chemistry-related area is due to the formation and/or production of CNMs which relies heavily on chemical reactions and interaction between materials. The knowledge in material science and engineering applications of CNM materials also plays important parts in this research area. Other research areas involved in the CNMs production from plastic waste, shown in Table 1, indicate the requirement of cross-discipline collaborations in this research field.

Journal analysis

Journal analysis is an important element in the bibliometric analysis that provides readers with an overview of the diversity of publication outlets for a specific research topic. The publications examined in this study were published in 78 journals. Table S1 (Supplementary Materials) shows the top journals that published more than two publications in PtCNM research. The 12 journals in Table S1 account for a cumulative percentage (CP) of 38.33% of the total publications, which designates these titles as the core journals in the field of CNMs synthesis from plastic waste according to the Bradford's Law of Scattering (Wong et al. 2021). Six of the core journals are first quartile journals, while five are second quartile journals. Such observation reflect the high quality of the PtCNM research, while led to the publications of the study results in top-tier journals. *Carbon* and *RSC Advances* are identified as the top journals, each with six articles. This is followed by four journals, each with four publications, namely *ACS Sustainable Chemistry & Engineering*, *Industrial & Engineering Chemistry Research*, *Journal of Analytical and Applied Pyrolysis*, and *Journal of Applied Polymer Science*. These observations indicate that plastic waste conversion to CNM is an emerging multidisciplinary research topic which covers a wide range of categories, from waste management to material science and chemical engineering. It is worth noting that *Applied Catalysis B-Environmental* has the highest IF among the core journals, although it is ranked in the third tier with only three publications related to this study. This observation could indicate the prominent role of catalysts in CNM synthesis since this journal focuses on catalyst utilisation for green production.

As PtCNM is a multidisciplinary research topic, an understanding on the relationship among the core journals, as well

as their influence in this field, is important to scientists when deciding the suitable publication outlets for their research works. The co-citation network depicted in Fig. S1 (Supplementary Materials) reveals 3 clusters represented by different colours (red, green and blue). The network map revealed *Carbon* and *Applied Catalysis B-Environmental* both have a stronger influence on the research of PtCNMs as shown by the larger node size with total link strengths of 31 and 24, respectively. In general, the journals in the 'red' cluster publish articles related to the catalytic pyrolysis or degradation of plastic/polymer to generate carbon nanostructures. The underlying mechanism, formation pathway and kinetics are the major focus. On the other hand, the journals in the green cluster emphasise the applicability of the generated carbon nanostructures, specific industrial potentials and detailed analysis of product properties. This research area aims to bridge the gap between laboratory-scale synthesis and industrial-scale production. *Carbon* has the highest link strength because it is one of the earliest journals that published works on PtCNM research by Chernozatonskii et al. (1998), which is widely cited by other authors in the literature. The strongest co-citation relationship in Fig. S1 is observed between *Carbon* and *Journal of Applied Polymer Science* due to the significance of material science (especially related to the characterisation aspects) in CNM synthesis from plastic waste. *ACS Sustainable Chemistry & Engineering* is the only journal in a blue cluster with no co-citation relationship with other journals in Fig. S1. This observation may be because some of the published works integrate unconventional feedstocks such as e-wastes (Maroufi et al. 2018) to produce CNMs. Although novel, current technologies pose as a limiting factor in the expansion of this research scope. Figure S1 provides an important reference for scientists to decide the target journal to publish their works, based on the central themes of these works, to ensure maximum impacts for the research community.

Top-cited publications

The top-cited publications serve as important foundation in the knowledge structure of a specific research discipline. Therefore, an examination on the contents of these influential publications provides readers with an initial guide on the progress in the research discipline. The top 10 publications (comprising 3 reviews and 7 research articles) with the most citations in PtCNM research retrieved from WoS are shown in Table S2 (Supplementary Materials). Based on the WoS data, about 20.83% or 25 documents each have at least 30 citations. The successful synthesis of graphene crystals from plastic waste using the CVD method provides a breakthrough in terms of lower material cost, which have greatly improved the economic sustainability of the entire process (Sharma et al. 2014). Other techniques for producing high-quality CNMs such as combustion, pyrolysis and hydrothermal conversion are still under various stages of development. Tang et al. (2005) proposed a simple yet novel method for the combustion of PP in the presence of clay and nickel (Ni), which highlighted the research potential of CNMs synthesis. This article is the most cited in the field with 142 citations. Gogotsi et al. (2011) discovered another unique method that involves the hydrothermal treatment and transformation of PS into CNTs in an aqueous medium with a Ni catalyst. Likewise, Zhuo et al. (2010) proposed another approach whereby the gaseous products generated from the pyrolysis of waste PE were combusted over a catalyst filled stainless-steel wire mesh to produce multi-walled carbon nanotubes (MWCNT). The synthesis of CNMs through these unique methods generally received high citation counts due to the various potentials.

Analysis of country, organisation, authors and collaborations

This section presents the research contributions towards PtCNM research at country, organisation and individual levels. The temporal evolutions of collaboration networks at different levels are also visualised in different sections. These information can be used by researchers to identify potential project supervisors and/or collaborators with specific research expertise/experience for upcoming research projects. Universities and research organisations can also identify productive research staffs which deserve further supports. In addition, funders could identify the current progress in PtCNM at county/region level and provide specific funding supports that strengthen the national research capacity and international collaborations in strategy ways.

Countries and regions

Countries with significant contributions to PtCNM research are presented in Table 2. In total, 36 countries have explored this research topic, while eight have published six or more research articles from 2000 to 2019. The earliest investigators (starting from the year 2000) of the potential of PtCNM research were from the USA and Japan, although investigations in these nations ceased momentarily only to re-emerge after 2010. China was the largest contributor to the research field with 44 publications or 36.67% of the total publications over the past two decades. Subsequently, the research output from China remained consistent for the next few years but gradually increased after 2012.

The enormous quantities of plastic waste generated by China have created significant environmental pollution problems (Yao et al. 2017). The quest for solutions to address these urgent challenges has prompted scientists to explore alternative routes for plastic waste management in China.

Table 2 Top productive countries on PtCNM research

Countries/ regions	TP	%TP	Year				TC	*h*
			2000–2004	2005–2009	2010–2014	2015–2019		
China	44	36.67	0	7	17	20	1044	17
India	18	15	0	0	3	15	272	9
USA	12	10	2	0	9	3	391	7
Australia	9	7.5	0	0	2	7	198	7
Japan	8	6.67	1	0	5	2	234	6
Poland	8	6.67	0	0	5	3	137	6
England	7	5.83	0	0	1	6	129	5
South Korea	6	5	0	0	2	4	51	4

From 2010 to 2014, various countries have become involved in this research field following the advancement made by Chinese scientists in this field. Based on the gradual increment of research outputs and participation of more countries in this research field, it is envisaged that the publications will continue to rise in the coming years.

Figure 3 displays the network visualisation map on the level of international collaboration for eleven countries with at least one research collaboration. As observed, China had the most international collaborations with a total link strength of seventeen, followed by Poland, which had collaboration with thirteen countries. The thick line connecting China and Poland indicates the close collaboration in this research field between the two countries. Although Egypt and Russia had five publications from the last two decades, none had international collaborations. However, the USA and Australia had a similar level of international collaboration (each with eight countries). This observation is due to the role of the USA as one of the earliest countries in PtCNM research, which is evident in the nation's overall total citation counts. In recent years, researchers from Singapore have begun investigating the synthesis of CNMs from plastic waste. This change could be related to the implementation of Sustainable Singapore Blueprint 2015, which aims to achieve an overall 70% recycling rate by the year 2030 (Ministry of the Environment and Water Resources & Ministry of National Development 2015). The concept of transforming plastic waste into highly valuable CNMs with extensive applications is a sustainable solution. Hence, it is expected that more countries will foster or

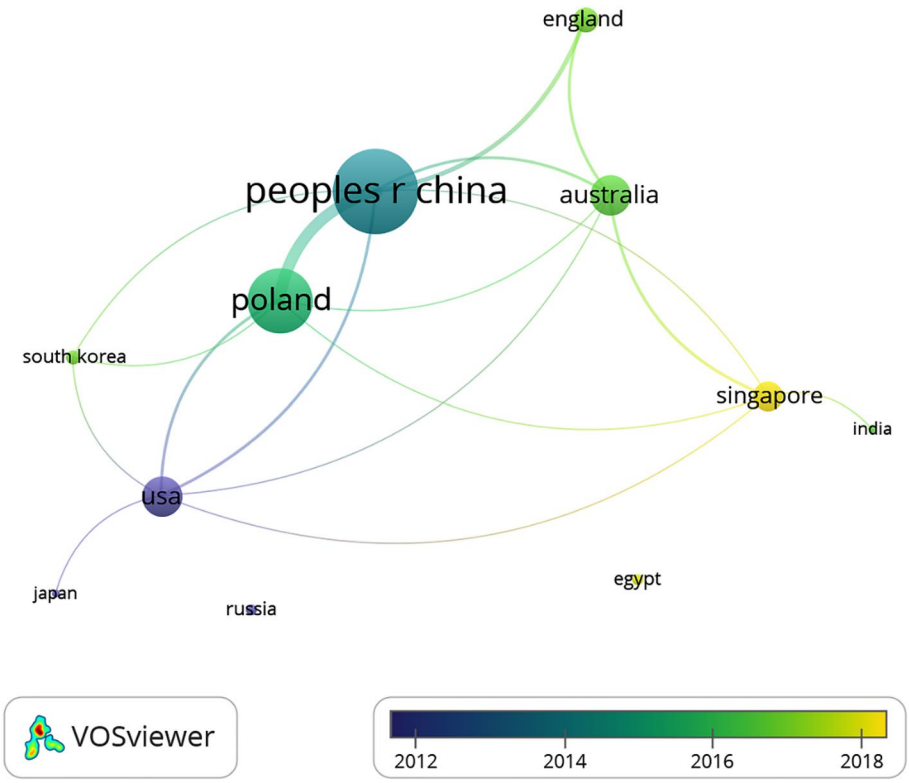


Fig. 3 Overlap visualisation of the co-authorship network between nations on CNMs synthesis from plastic waste research (minimum number of publications: 5, the minimum number of citations: 43, node size denotes total link strength)

adopt international collaborations in the development of this 'waste-to-gold' management route.

Organisations

The key publications on PtCNM research from 2000 to 2019 are attributed to 154 organisations and only 18 (11.7%) producing more than two publications. The organisations that have produced at least four publications from the year 2000 to 2019 are listed in Table 3. The Chinese Academy of Sciences is the most prolific organisation that contributed to this field with thirteen publications (10.83%), followed by the West Pomeranian University of Technology (Poland) (8, 6.67%). This is due to the significant international collaborations between the two institutions. Although the Pomeranian University of Technology came in second place in terms of publications, the Northeastern University (USA) was more influential with a higher citation count of 282. This is attributed to the Northeastern University's early start on PtCNM research in the year 2000. With more institutions becoming involved in this research field, it is expected that new research and citable papers are readily available in the near future.

Figure 4 shows the co-authorship network among the organisations with a minimum of four publications and eight citations. There are five clusters in total with the largest one comprising organisations that have collaborations with the Chinese Academy of Sciences. The other four clusters consist of intra-institutional collaborations (e.g. Nanyang Technological University, University of Hull and University of Science and Technology of China) or inter-institutional collaborations within countries (e.g. Indian Institutes of Technology and Visvesvaraya National Institute of Technology). One of the reasons why China is the leading country in this research field is due to the independent efforts and research publications of two teams at the Chinese Academy of Sciences and University of Science and Technology of China. However, inter-institutional collaboration is still low, which may be the reason for the slow development of plastic waste management process. It is worth noting that the pathways for plastic waste conversion to CNMs are still under development although the participation of other institutions is predicted to rise in the near future.

Authors

Table 4 shows the list of authors with at least 5 publications comprising 11 top authors or 72.73% from China. Overall, there are 458 authors involved in PtCNM research, but only 22 authors (4.8%) have produced more than 2 publications within the studied period. Figure 5 displays the co-authorship network for authors with a minimum of five publications and 53 citations. Chen Xuecheng and Tang Tao are the two most prolific researchers from China with ten publications each. Tang Tao has a slightly higher citation count of 371 when compared to 300 for Chen Xuecheng. Both researchers co-authored the most cited paper titled 'Synthesis of multiwalled carbon nanotubes by catalytic combustion of polypropylene' in 2005. The inter-institutional collaboration by Tang Tao from the Chinese Academy of Sciences and Chen Xuecheng from Shanghai University has made the duo the most productive authors. The duo had also co-authored various publications related to the conversion of plastic waste such as polyolefins and PET into CNMs for dye absorption (Feng et al. 2014) and supercapacitor applications (Wen et al. 2019). Figure 5 also reveals that the authors from China are closely linked to each other, which explains why the majority of the highly prolific authors and cited publications are from China. Furthermore, Zhang Junhao led another independent research group at the University of Science and Technology of China.

The author Ewa Mijowska from the West Pomeranian University of Technology in Poland is linked to all the six top publishing authors in China (Fig. 5), which further

Table 3 Top productive organisations on PtCNM research

Organisations	Country	TP	% TP	Year				TC	*h*
				2000–2004	2005–2009	2010–2014	2015–2019		
Chinese Academy of Sciences	China	13	10.83	0	3	4	6	384	9
West Pomeranian University of Technology	Poland	8	6.67	0	0	5	3	137	6
Indian Institutes of Technology	India	5	4.17	0	0	2	3	116	5
Northeastern University	USA	5	4.17	0	1	3	1	282	5
University of Science and Technology of China	China	5	4.17	0	2	3	0	119	5
Visvesvaraya National Institute of Technology	India	5	4.17	0	0	0	5	53	4
Nanyang Technological University	Singapore	4	3.33	0	0	0	4	36	3
Northeast Forestry University	China	4	3.33	0	0	0	4	8	2
University of the Chinese Academy of Sciences	China	4	3.33	0	0	3	1	82	4
University of Hull	England	4	3.33	0	0	0	4	81	3

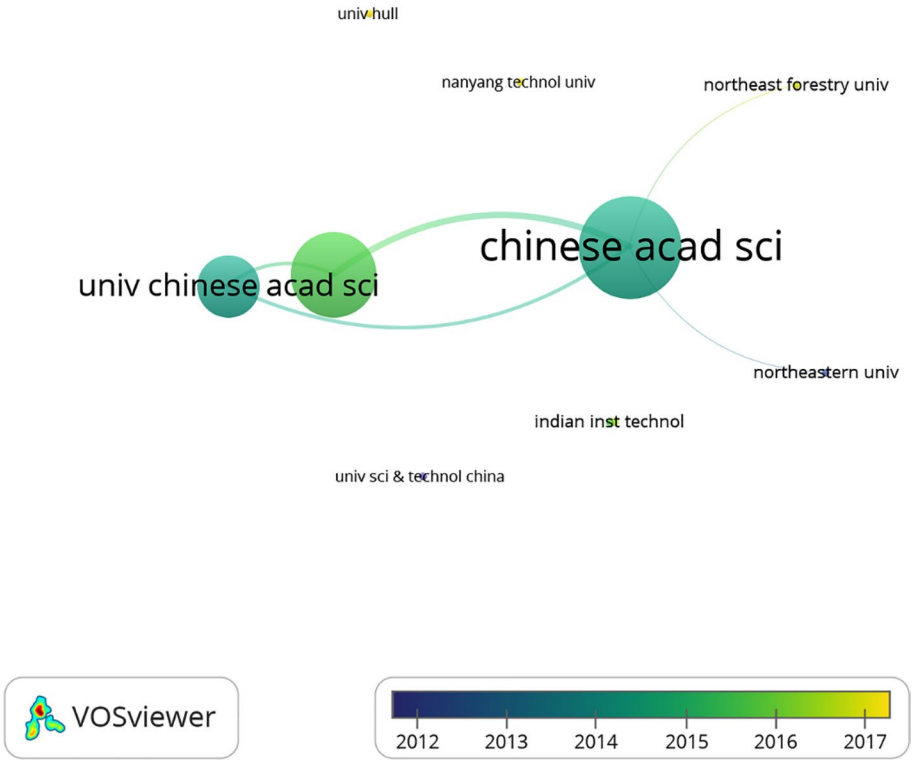


Fig. 4 Collaboration network at institutional-level CNM synthesis from plastic waste in research (minimum number of publications: 4, minimum number of citations: 8, node size denotes total link strength)

Table 4 Top productive authors on carbon nanomaterials synthesis from plastic waste

Authors	Country	TP	% TP	Year				TC	*h*
				2000–2004	2005–2009	2010–2014	2015–2019		
Chen, Xuecheng	China	10	8.33	0	2	5	3	300	7
Tang, Tao	China	10	8.33	0	2	5	3	371	7
Gong, Jiang	China	8	6.67	0	0	5	3	137	6
Song Rongjun	China	8	6.67	0	2	2	4	127	4
Mijowska, Ewa	Poland	7	5.83	0	0	4	3	106	5
Wen, Xin	China	7	5.83	0	0	5	2	125	5
Liu, Jie	China	6	5.00	0	0	4	2	125	5
Jiang, Zhiwei	China	5	4.17	0	1	3	1	174	5
Vijayakumar, R. P	India	5	4.17	0	0	0	5	53	4
Wu, Chunfei	England	5	4.17	0	0	1	4	126	4
Zhang, Junhao	China	5	4.17	0	2	2	1	123	5

affirms the research collaboration established between China and Poland. The research collaboration with Ewa Mijowska mainly focuses on the synthesis of CNM for water treatment (Feng et al. 2014) and electronic devices (Wen et al. 2019). The findings indicate that the establishment of a productive authorship profile and high research impact in certain fields require both inter-institution and intra-institution research collaborations.

Funding agencies

Funding is an indispensable factor which moves the research frontier forward. Therefore, an investigation of the available funding agencies in PtCNM research explains the productivities at different levels and provides researchers with essential information on potential funding supports for upcoming projects. Table S3 (Supplementary Materials) displays

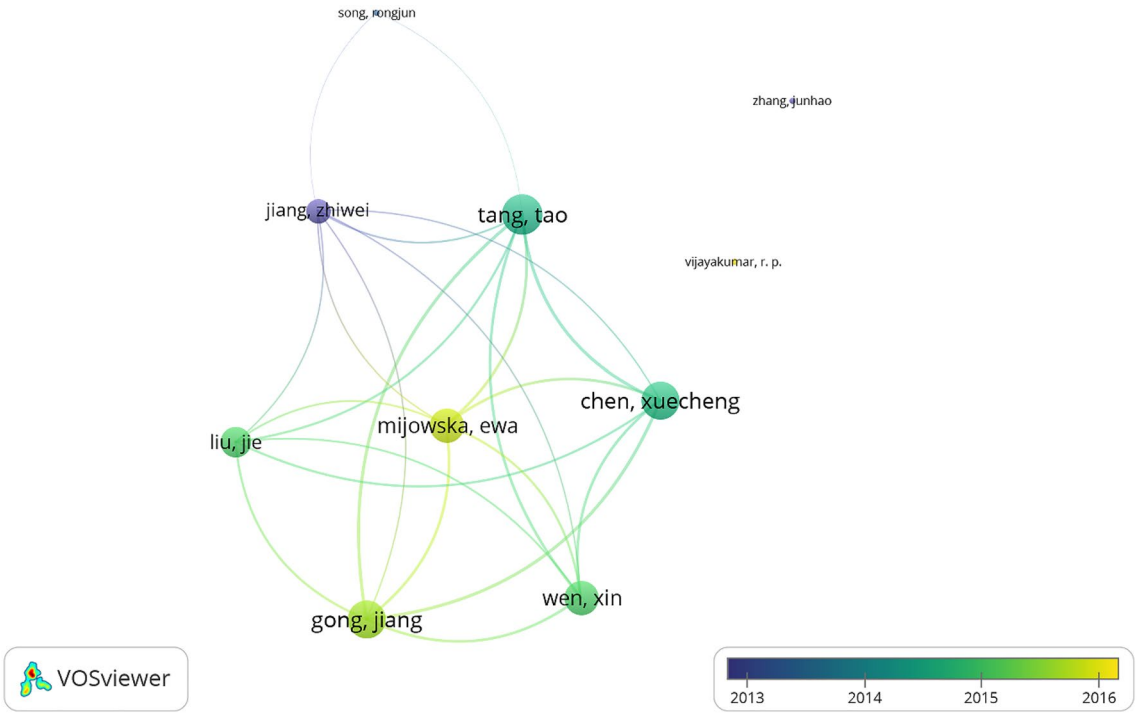


Fig. 5 Overlap visualisation of the co-authorship network of authors in CNMs synthesis from plastic waste (minimum number of publications: 5, minimum number of citations: 53, node size denotes total link strength)

the funding agencies with at least 3 publications within the studied period. From the extracted WoS data, 89 agencies have funded PtCNM research in 2000–2019. The funding by the National Natural Science Foundation of China and the National Basic Research Program of China has propelled the research publications of China with a record of 44 publications from 2000 to 2019. The funding agencies in Table S3 were mostly located in the Asia–Pacific region (e.g. India, Japan, Singapore, Australia), which highlights the greater concerns about plastic waste problems in the region. In particular, Japan, Singapore and Australia have high plastic recycling rates backed by effective waste plastic recollection systems (UNCRD 2019). The transformation of plastic waste into valuable materials such as CNM will attract further funding and investments, which could foster progress in this area of research. The analysis also revealed that 28 (or 23.33%) publications received no funding for the research work, whereas 4 (14.29%) received some form of supports. This observation highlights the concern that funding is one of the most important issues for progress in this research field. The common approach used to acquire funds for research work was through national grants as some nations have funds allocated to research development. Smagulova et al. (2019) successfully obtained research funds from the Ministry of Education and Science of the Republic of Kazakhstan for CNT synthesis from HDPE. Besides, the Thai Government Research Fund supported the work on carbon dots synthesis from PU reported in Dela Cruz et al. (2019).

In the scenario where national funds are scarce or low, institutional funds were the next available option. The institutional support from King Saud University in Saudi Arabia enabled the development of biocompatible carbon nanocapsule for cancer therapy at the Queen Elizabeth Hospital, Australia (Mezni et al. 2017). This type of international collaboration was the backbone of advanced research as foreign funds can be implemented to achieve a common goal. Funding from the private sector was also vital to research progress. For instance, the work by Sharon et al. (2015) on transformation of PE into graphene, CNT and carbon dots was supported by the Society of Instrument and Control Engineers—an independent platform for international conferences. Moreover, Wuhan Optics Valley Environmental Technology Co. Ltd. supported the work of Liu et al. (2018) that reported CNT synthesis from the thermal conversion of plastic waste. The funds from various grants enabled the hiring of manpower and purchasing of materials, equipment and services necessary for research work.

Keyword co-occurrence analysis

Examination of the keywords co-occurrence relationship in a specific research topic is an important step in the bibliometric analysis that reveals the research hot spots. In this

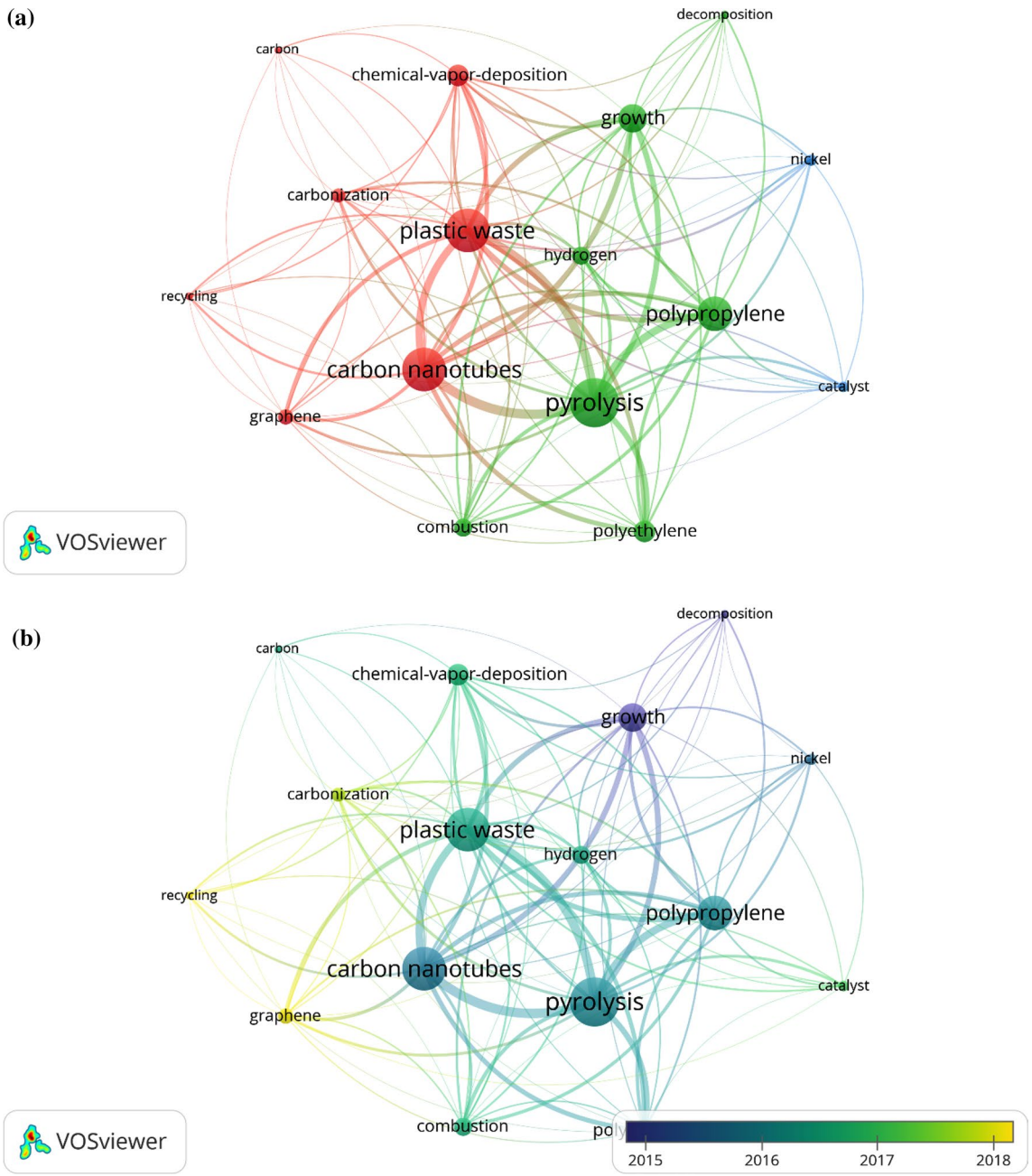


Fig. 6 **a** Network and **b** overlap visualisation maps of keyword co-occurrence in PtCNM research (minimum occurrence: 6)

study, network and overlay visualisation maps (Fig. 6) that illustrate keyword occurrence in PtCNM research were produced using VOSviewer software based on the keywords extracted from the publication dataset. A thesaurus file (Supplementary Materials, Table S4) was used to merge different keywords with similar meanings. A total of 622 keywords were identified from the selected literature. To simplify the co-occurrence analysis, only keywords with a minimum occurrence of five were used to generate the visualisation maps. Figure 6 shows the network and overlay visualisation maps that consist of 32 keywords. 'Carbon nanotubes' (231), 'pyrolysis' (217) and 'plastic waste' (156) are the three keywords with the highest total link strengths, as the terms represent the core idea of PtCNM research. This is followed by 'polypropylene' (149), 'growth' (149), 'chemical vapour deposition' (95), 'hydrogen' (90), 'carbonisation' (87), 'combustion' (78) and 'nickel' (62).

The keywords in Fig. 6 are grouped into four clusters, each marked with a distinctive colour. Each cluster represents a research hot spot in PtCNM research. Cluster 1 (in

red colour) contains nine keywords, while three possess the highest total link strengths in the keyword co-occurrence map. This cluster represents the synthesis of CNT and nanofibres from plastic waste, with nickel being an important catalyst in the process (Bazargan and McKay 2012). Cluster 2 (in blue colour) depicts a growing interest in the feasibility of graphene synthesis from plastic waste. Cluster 3 (in yellow colour) illustrates the scientists' interest in hydrogen production from the plastic waste in addition to the carbon nanostructures. Cluster 4 (in green colour) describes the potential of other thermochemical processes, including combustion, flame synthesis and carbonisation, as a potential solution of low-cost CNM synthesis and plastic waste valorisation.

Discussions on PtCNM research

Plastics are excellent low-cost carbon precursors for CNM synthesis due to the high-carbon content. Thus, the synthesis of graphene and CNTs from plastic via a bottom-up approach is an interesting option in plastic waste management. Table 5 displays the key information extracted from the publications on the plastic-to-CNT conversion, while Table 6 summarises the information extracted from the reports on plastic-to-graphene conversion. The information in the tables includes the types and origins of plastics, catalyst/substrate, reaction conditions, yields and quality of CNTs/graphene products. In general, the publications are separated according to the technologies used (pyrolysis-CVD in two stages, one-step synthesis, template-assisted growth and combustion). As illustrated in Table 5, a great number of research teams explored the CNT synthesis. This observation agrees with Cluster 1 in the keyword co-occurrence map (Fig. 6), which indicates the importance of pyrolysis in plastic waste conversion to CNT. On the other hand, limited studies have reported on graphene synthesis from plastic waste (Table 6), which corresponds to Cluster 2.

The most widely used strategy in plastic-to-CNM synthesis is the adoption of two-stage reactors, where the plastic pyrolysis and CNM synthesis occur in two separate yet connected chambers. The reaction parameters in each chamber can be controlled independently for maximum performance (Backes et al. 2020). The successful synthesis of high-quality CNTs (indicated by low I_D/I_G ratio from Raman spectrometry analysis) from waste PE based on the pyrolysis-CVD reactor setup has been reported. However, the intermediates produced during plastic pyrolysis are highly reactive and thus are also prone to recombination forming liquid fuels (Aboul-Enein et al. 2018). The CNT synthesis thus occurs in parallel with liquid fuel formation, as shown in Table 5. To prevent the catalyst deactivation by the liquid product, the condensation of the heavy hydrocarbons is necessary before the carbon precursor enters the synthesis reactor (Borsodi et al. 2016). Nevertheless, the formation of a liquid product inevitably leads to a lower hydrocarbon gas yield for CNT growth. The most practical approach is to allow re-cracking of condensable vapours from plastic pyrolysis into non-condensable gases, as demonstrated by Aboul-Enein and Awadallah (2019). Thus, the formation of liquid (fuel) products can be avoided, while the CNT yield can be maximised. Due to the high-hydrogen content of plastic waste, the decomposition of the carbon precursor always accompanies the release of hydrogen radicals, which combine to form hydrogen gas. Thus, a research goal in this field lies in the conversion of plastic waste to CNM and hydrogen gas by suitable reactor and process design.

Several research teams have also successfully converted plastic waste to CNT or graphene using different synthesis routes, which correspond to Cluster 4. One example of such route is the one-step carbonisation, which is realised due to the dual function of metal particles in plastic pyrolysis and CNT/graphene growth (Cui et al. 2017a). The template-assisted growth of CNT from plastic waste has also been reported in the literature (Liu et al. 2017). This strategy involves the growth of CNT from the carbon precursors in a well-defined volume provided by the template. The CNTs formed possess uniform length, diameter and quality. The template can be removed easily through acid or alkaline washing to obtain pure CNTs. However, the yield of CNTs synthesized with Ni supported on anodic aluminium oxide, as reported by Liu et al. (2017), is sensitive to the temperature. Steam injection increases the CNT quality at the expense of its yield. Nevertheless, more in-depth investigations are required to provide definitive evidence on the occurrence of CNM synthesis via template-assisted growth.

Most of the scientific investigations listed in Tables 5 and 6 focused on the effects of several key factors on the CNM formation, namely choice of catalyst/substrate, temperature, plastic type and reaction environment. The effects of these factors are discussed below.

Choice of catalyst/substrate

As shown in Table 5, Ni-based catalysts are used in almost all plastic-to-CNM conversion through the CVD route. This observation is related to the significant role of Ni metals in promoting the dehydrogenation of carbon precursors (Bajad et al. 2016). Similarly, Yang et al. (2018) observed the positive effects of Ni in the dry reforming of methane and hydrocarbons. Recently, Aboul-Enein and Awadallah (2019) investigated the conversion of PP waste over Ni and Ni-Cu catalysts supported on La_2O_3. The support effectively suppressed catalyst agglomeration, while the addition of Cu enhanced the catalytic decomposition of carbon precursors. The growth of high-quality CNTs with fewer defects on Ni

Table 5 Summary of key publications on plastics-to-CNT conversion. The numbers and alphabets in brackets represent the different plastic type/substrate/catalyst/reaction conditions

Ref (Year)	Type of plastics reaction steps and conditions	Conversion, yield, properties
Pyrolysis-CVD (separate placements for plastics and substrate/catalyst)		
Panahi et al. (2019)	Waste plastics: PE (1), PP (2), PET (3), PS (4) and mixture (5) Substrate: stainless steel wire cloths (SS-304(a), SS-316 (b), SS-316L(c)), acid (HCl) washed for 10 min followed by heat treatment in air at 800 °C for 1 min, followed by rapid air quenching Pyrolysis: 800 °C, N_2, 2 L/min Synthesis: 800 °C, N_2, 2 L/min	Product: MWCNT forest Yield (wt%): 1.6 (1a), 4.1 (1b), 3.7 (1c), 1.7 (2a), 6.0 (2b), 4.5 (2c), 0 (3a), 1.6 (3b), 0.8 (3c), 1.3 (4a), 3.7 (4b), 2.4 (4c), 1.7 (5a), 3.9 (5b), 3.6 (5c) Diameter (nm): 50 (1), 40 (2), 60 (3), 75 (4), 60 (5) For PE-derived CNTs: I_D/I_G: 0.44 (1a), 0.36 (1b), 0.37 (1c) I_{2D}/I_G: 0.75 (1a), 0.79 (1b), 0.74 (1c) I_{2D}/I_D:1.71 (1a), 2.22 (1b), 1.98 (1c)
Moo et al. (2019)	Virgin LDPE (1), PP (2), mixed plastics (3) Catalyst: NiO on $CaCO_3$ Pyrolysis: 600 °C, N_2, 50 mL/min, 30 min Synthesis: 500 °C (a) or 800 °C (b), N_2, 50 mL/min, 30 min	Product: MWCNT Yield ($wt_{product}/wt_{catalyst}$) (%): 21 (1a), 32 (2a), 3 (3a), 31 (1b), 22 (2b), 22 (3b) I_D/I_G = 1.49 (1a), 1.56 (2a), 1.47 (3a), 0.38 (1b), 0.38 (2b), 0.39 (3b) Oxygen reduction reaction potential vs Ag/AgCl/KCl (sat) electrode (V): − 0.132 (1a), − 0.135 (2a), − 0.108 (3a), − 0.179 (1b), − 0.176 (2b), − 0.212 (3b)
Aboul-Enein and Awadallah (2019)	Waste PP cups Catalyst: 50% Ni/La_2O_3 (1), 40% Ni-10% Cu/La_2O_3 (2) Pyrolysis: N_2, 50 mL/min, 500 °C (re-condensation of liquid product) Synthesis: N_2, 50 mL/min, 700 °C	Product: MWCNT and carbon nanofibres Gas yield (wt%): 55.2 (1), 38.1 (2) Carbon yield (wt%): 34.8% (1), 51.9 (2)
Aboul-Enein et al. (2018)	Waste LDPE (1), PP (2), HDPE (3), PET (4), PS (5) Catalyst: 10% Ni-9.5% Mo/Al_2O_3 Pyrolysis: N_2, 30 ml/min, 650–700 °C, 20 C/min Synthesis: N_2, 30 ml/min, 650 °C	Product: MWCNT In step 1: Residue + char yields (wt%): 24.2 (1), 21.5 (2), 33.0 (3), 61.8 (4), 55.1 (5) Gas yields (wt%): 72.3 (1), 70.4 (2), 62.0 (3), 35.4 (4), 22.4 (5) Liquid yields (wt%): 2.8 (1), 7.4 (2), 4.5 (3), 2.2 22.1 (4), (5) In step 2: Gas yield (wt%): 72.2 (1), 70.5 (2), 61.9 (3), 35.5 (4), 22.5 (5) Carbon yield (g/g_{cat}): 66.3 (1), 57.1 (2), 44.2 (3), 6.0 (4), 2.4 (5) I_D/I_G: 1.26 (1), 1.31 (2), 1.68 (3), 1.43 (4), 1.74 (5)
Aboul-Enein et al. (2017)	Waste LDPE Catalyst: 5% Ni-Mo/Al_2O_3 (1) and 10% Ni-Mo/Al_2O_3 (2) Pyrolysis: N_2, 50 mL/min, 500–800 °C (re-condensation of liquid product) Synthesis: N_2, 50 mL/min, 600–800 °C	Product: MWCNT Product yields (wt%) during pyrolysis: Liquid: 0.4 (500), 1.8 (600), 3.0 (700), 0.4 (800) CNT: 14.3 (500), 18.9 (600), 27.6 (700), 27.8 (800) Gases: 29.7 (500), 55.9 (600), 72.5 (700), 85.7 (800) Residue + wax: 68.8 (500), 41.4 (600), 23.9 (700), 12.8 (800) At different pyrolysis temperatures: I_D/I_G: 2.02 (500 C), 2.11 (600 C), 1.93 (700 C), 1.60 (800 C) At different CVD temperatures: I_D/I_G: 2.21 (600 C), 1.93 (650 C), 1.76 (700 C), 1.48 (750 C), 1.63 (800 C)
Liu et al. (2018)	Waste HDPE Catalyst: ceramic membrane (Ni/Al_2O_3), with Ni content of 0.1 mol/L (1) or 0.5 mol/L (2) or 1.0 mol/L (3) or 2.0 mol/L (4) Pyrolysis: N_2, 100 ml/min, 500 °C Synthesis: N_2, 100 ml/min, 600 °C (a) or 700 °C (b) or 800 °C (c)	Product: CNT Yield (wt%): 3.1 (1b), 6.0 (2b), 9.4 (3b), 8.0 (4b), 7.2 (2a), 1.2 (2c) Diameter (nm): 15.7 (1b), 16.9 (2b), 20.8 (3b),24.9 (4b), 21.2 (2a),—(2c)

Table 5 (continued)

Ref (Year)	Type of plastics reaction steps and conditions	Conversion, yield, properties
Yang et al. (2018)	Virgin PP mixed with waste PE Catalyst: Ni/Al_2O_3 Gasification: feeding rate of 19.8 g/min, air, equivalence ratio (ER) of 0.1 (1), 0.15 (2) and 0.2 (3), temperature: 500 °C (a), 600 °C (b), 700 °C(c) Synthesis: 680 °C	Product: CNT At different ER: Yield ($wt_{plastic}/wt_{cat}$, %): 22.5(1), 21 (2), 16 (3) I_G/I_D: 0.71 (1), 0.5 (2), 0.58 (3) At different temperature: Yield ($wt_{plastic}/wt_{cat}$, %): 16 (a), 22 (b), 26 (c) I_G/I_D: 0.65 (a), 0.71 (b), 0.86 (c)
Yang et al. (2015)	Virgin PP mixed with waste PE Catalyst: H-Ni/Al_2O_3 (1) or A-Ni/Al_2O_3 (2) or N-Ni/Al_2O_3 (3) Temperature: 600 °C (a) or 680 °C (b) or 750° C (c)	Product: CNT CNT yield ($wt_{plastic}/wt_{cat}$, %):: 8.4 (1a), 16.6 (2a), 22.0 (3a), 20.0 (1a), 24.3 (1b), 30.5 (1c) H_2 yield (mmol/g plastic): 29.5 (1a), 30.9 (2a), 32.4 (3a), 25.5 (1a), 27.4 (1b), 28.6 (1c) I_G/I_D: 0.71 (1), 0.66 (2), 0.43 (3)
Tripathi et al. (2017)	Waste PP Catalyst: stainless steel tube reactor oxidised at 900 °C Pyrolysis: 500 °C, Ar, 100 ml/min Synthesis: Ar, 100 ml/min, 600 °C(1) or 700 °C (2), or 800 °C (3) or 900 °C (4) or 1000 °C (5) or 1100 °C (6)	Product: MWCNT Yield (wt%): 0.6 (1), 15 (2), 22.5 (3), 42.4 (4), 31.4 (5), 35 (6) I_D/I_G: 0.87 (1), 0.75 (2), 0.56 (3), 0.48 (4), 0.63 (5), 0.58 (6) I_{2D}/I_G: 0.35 (1), 0.55 (2), 0.62 (3), 0.60 (4), 0.59 (5), 0.53 (6)
One-step CNT formation (mixed polymer and catalyst)/carbonisation		
Cui et al. (2017a)	Virgin PP (1), PE (2), PS (3) Catalyst: Ni–Mo–Mg (a) and Ni–Mo–Mg/5% carbon black (b) Conditions: Mixed catalyst and polymer, N_2, 850 °C, 15 min	Product: MWCNT Yield (wt%): 26.24 (1a), 53.56 (1b), 28.36 (2a), 65.95 (2b), 18.39 (3a), 41.85 (3b) I_G/I_D: 0.79 (1a), 1.07 (1b), 0.9 (2a), 0.96 (2b), 0.99 (3a), 1.1 (3b)
Bajad et al. (2016)	Waste PP Catalyst: Ni/Mo/MgO Condition: 800 °C, vacuum, 30 min	Product: CNT Yield (wt%): 53.3 I_G/I_D: 0.93 (as-synthesized), 0.75 (purified)
Bajad et al. (2017)	Mixed plastics (source not specified) Catalyst: Ni/Mo/MgO Pyrolysis: 700 °C Synthesis: 800 °C, reaction time: 80 min	Product: CNT CNT Yield (wt%): 11.1 H_2 yield (wt%): 78.8
	Virgin PP (mixed with silicone microflakes) Catalyst: Ni–Mo–Mg	Product: CNT-based Si–C nanocomposites Electrochemical performance (300 mA/g, 0–2.5 V): Initial discharge capacity: 1453 mAh/g, Coulombic efficiency: 66.2% After 50 charge/discharge cycles: Final discharge capacity: 1185 mAh/g, Coulombic efficiency: 98.8%
	Virgin PP Catalyst: Organically modified montmorillonite (OMMT) combined with NiO with a diameter of 18 nm (1) or 26 nm (2) or 40 nm (3) or 96 nm (4) or 128 nm (5) or 227 (6) nm Condition: Mixed catalyst and polymer, 700 °C, 5 min	Product: CNT Yield (%): 51.9 (1), 48.4 (2), 41.7 (3), 29.5 (4), 21.9 (5), 10.8 (6) Length (nm): 7.2 (1), 0.6 (2), 17.9 (3), 5.2 (4), 2.6 (5), 0.7 (6) Diameter (nm): 45.7 (1), 48.6 (2), 56.5 (3), 71.3 (4), 126.1 (5), 247.8 (6) I_G/I_D: 0.57 (1), 0.58 (2), 0.61 (3), 0.55 (4), 0.46 (5), 0.42 (6)

Table 5 (continued)

Ref (Year)	Type of plastics reaction steps and conditions	Conversion, yield, properties
Liquid Injection-CVD		
Hedayati et al. (2019)	Black packaging waste (1), white weigh boats (2), both PS Plastic was dissolved in toluene to form a plastic solution Catalyst: ferrocene dissolved in toluene (5 wt%) Pyrolysis: solution flow: 1–3 mL/h, H_2/Ar: 1 L/min, 225 °C Growth: H_2/Ar: 1 L/min, 780 °C	Product: MWCNT I_G/I_D = 2.1 (1), 2.1 (2)
Template-assisted CNT synthesis		
Liu et al. (2017)	Waste HDPE Template: Ni-based anodic aluminium oxide membrane, with Ni content of 0.1 mol/L (1) or 0.5 mol/L (2) or 1.0 mol/L (3) or 2.0 mol/L (4) Pyrolysis: N_2, 100 ml/min, 500 °C Synthesis: N_2, 100 ml/min, 600 °C (a) or 700 °C (b) or 800 °C (c)	Product: CNT Yield (wt%): 7.5 (1b), 5.3 (2b), 22.0 (3b), 20.5 (4b), 1.8 (1a), 4.7 (1b),—(1c) Diameter (nm): 26.7 (1b), 38.3 (2b), 40.1 (3b), 54.1 (4b), 26.2 (1a), 33.5 (1b), 27.2 (1c)
Flame synthesis/combustion		
Wulan et al. (2018)	Waste PP Substrate: stainless less steel 316 wire mesh, acid-etched using isopropanol followed by oxidative heat treatment at 800C, for 10 min Pyrolysis: 525 °C (1) or 550 °C (2) or 600 °C (3) at 30 min (a) or 45 min (b) or 60 min (c) Synthesis: Ar (100 ml/min), O_2 (33.3 ml/min), 1 h Condition: 525 °C (1) or 550 °C (2) or 600 °C (3), 30 min (a) or 45 min (b) or 60 (min) or 90 min (a)	Product: MWCNT CNT yield: 43.0 (1a), 54.0 (1b), 26.0 (1c), 0.69 (1d), 9.7 (2a), 2.1 (2b), 1.6 (2c), 0.9 (2d), 0.08 (3a)

Table 6 Summary of key publications on plastics-to-graphene conversion. The numbers and alphabets in brackets represent the different plastic type/substrate/catalyst/reaction conditions

Reference(s)	Type of plastics and reaction conditions	Conversion, yield
Pandey et al. (2019)	Waste PP + PE + PS Catalyst: bentonite nanoclay Pyrolysis: N_2, 400 °C, 5 °C/min Synthesis: N_2, 20 mL/min, 750 °C, 10 °C/min	Product: Graphene nanosheets Yield = 15% $I_G/I_D = 0.90$ Quantum capacitance, $C_Q = 18.85$ μF/cm^2 at − 1 V
El Essawy et al. (2017a); El Essawy et al. (2017b)	PET waste bottle Catalyst: ferrocene Stainless steel autoclave reactor, 800 °C, 8 °C/min, holding time: 1 h	Product: Graphene Yield (wt%): 36, $I_D/I_G = 1.13$, $S_{BET} = 721.7$ m^2/g, pore size = 2.1 nm Adsorption Capacity at 25 °C: 761.3 mg/g (for MB), 642.9 mg/g (AB25)
Cui et al. (2017b)	PP (lunch box) (1), PP (plastic bag) (2), PE (plastic bag) (3), PE (preservative film) (4), PE (clean agent bottle) (5), PE (valve bag) (6), PMMA (virgin) (7), PS (virgin) (8), PP (virgin) (9), PVC (virgin) (10), PE (virgin) (11), PET (virgin) (12) Catalyst: polycrystalline Ni foil Step 1: Annealing of Ni foil (Ar/H_2, 1050 °C, 30 min) Step 2: Graphene growth (Ar/H_2, 1050 °C, 120 min)	Product: Graphene foil Conversion (%) = 32.6 (1), 31.7 (2), 38.5 (3), 27.4 (4), 26.0 (5), 32.0 (6), 14.8 (7), 42.9 (8), 26.8 (9), 10.3 (10), 34.1 (11), 17.8 (12) I_D/I_G values: 0.024 (1), 0.029 (2), 0.050 (3), 0.034 (4), 0.055 (5), 0.068 (6), 0.649 (7), 0.013 (8), 0.098 (9), 0.030 (10), 0. 1 (11), 0.066 (12) Electrical conductivities (S/cm) = 3700 (1), 3600 (2), 3820 (3), 3100 (4), 3250 (5), 3010 (6), 1770 (7), 3480 (8), 2880 (9), 3710 (10), 2900 (11), 3080 (12)

catalyst was observed due to the homogenous size of the Ni catalyst particles. Nevertheless, the formation of carbon nanofibres and CNT with different diameters was observed on the Cu-Ni alloy, which was transformed into a quasi-liquid state forming metal particles with different sizes at high calcination temperature. This observation explains the significant influence of the catalyst particle size on the CNT diameter.

The catalyst particle size also plays an important role in the formation of graphene and CNT (such as crystalline foil graphene, metal particles CNTs) from carbonaceous materials. Yang et al. (2015) showed that H–Ni/Al_2O_3 with the smallest crystallite size of Ni produced the highest CNT yield. This observation is related to the increased bulk diffusion of carbon atoms from the metal–gas interface to the metal–nanotube interface. The diameter of the CNMs is influenced by the metal particle size on the catalyst support (Aboul-Enein and Awadallah 2019). The synthesis of small-sized CNT requires eliminating the aggregation of the metallic particles by controlling the reaction temperature (to prevent metal sintering) and the existence of strong metal–support interaction. According to the authors, carbon nanofibres synthesized over Ni–Cu alloy particles possessed a higher diameter than that over Ni particles. Since the lower melting point of Cu resulted in partial deformation of Ni–Cu, an increase in the particle size was observed. Liu et al. (2018) reported the application of ceramic membrane as unconventional support for Ni catalyst. The existence of catalyst particles with different sizes at high Ni loading led to the formation of MWCNTs with different diameters. Yang et al. (2015) also showed that H–Ni/Al_2O_3 with the smallest crystallite size of Ni produced the highest yield of CNT. This observation is related to the high bulk diffusion of carbon atoms from the metal–gas interface to the metal–nanotube interface. Based on the above findings, strict control of catalyst nanoparticle size is vital to produce CNT with the desired size.

The advantages of Ni-based catalyst in PtCNM conversion were also investigated by other research teams. Due to its rich content of Ni and Fe, a stainless steel reactor can be used as a catalyst in addition to its role as a reaction vessel. Tripathi et al. (2017) used an oxidised stainless-steel SS 316 tube to successfully convert PP waste into MWCNTs. The removal of the Cr protective layer on the stainless steel surface was necessary to fully exploit the catalytic properties of Ni and Fe. Cui et al. (2016) successfully performed the one-step synthesis of a CNT network from PP waste. The key to the formation of the special CNM was the addition of carbon black to the Ni-Mo-Mg catalyst. The nano-scaled carbon black particles were able to capture the free radicals produced during the plastic pyrolysis, which then participate in the growth process. Nevertheless, such a scavenger ability was not observed in the large carbon black particles. Due to the presence of the highly stable graphitised carbon structure, the carbon black-soldered-CNT network was thermally stable up to 500 °C.

Despite the excellent catalytic role of Ni metal, the deactivation of Ni-based catalysts is widely reported due to the formation of amorphous carbon that covers the catalyst surface. Cui et al. (2017a) proposed the addition of Mo to Ni as a solution to such limitation, as the bond formation between Co and the carbon atoms (during the growth stage) is more

stable than the bond formed between Ni and carbon. Thus, the addition of Mo to Ni is expected to increase the catalyst lifetime. Based on the use of MgO as excellent support for Ni and Co particles, Cui et al. (2017a) claimed the invention of Ni–Mo–MgO as a universal catalyst that results in high CNTs yields from PP, PE and PS blends.

Temperature

Temperature is another significant reaction parameter in plastic-to-CNM conversion after the choice of catalyst/substrate. Higher pyrolysis temperatures result in higher gas yield, which is vital to improving the yield of CNT (Aboul-Enein et al. 2017). In a pyrolysis–catalysis reactor, pyrolysis/degradation temperature has less influence on the CNTs quality (indicated by I_G/I_D value); however, the tube diameter and morphology varied greatly at different pyrolysis temperatures. In comparison, the catalysis temperature exerts a more significant influence on the yield and quality of the CNT product. A sufficiently high synthesis temperature is needed for the optimal growth of CNTs; however, increasing the temperatures above an optimum point could lead to defective CNT or amorphous carbon formation (Liu et al. 2018). The temperatures used during the catalyst calcination and CNT growth also affect the particle size on the metal catalyst. The agglomeration or sintering of metal particles at high reaction temperatures increases the diameter of CNT (Kumar and Ando 2010).

Plastic type

The unique molecular structures of different plastics exert different impacts on the CNT synthesis. Aboul-Enein et al. (2018) showed that PP and LDPE pyrolysis at 700 °C produced high gas yield as the branched-chain structure in these polymers is prone to decomposition. In contrast, the straight-chained structure of HDPE was more resistant to thermal decomposition at similar temperatures. The pyrolysis of PET resulted in oxygen-rich intermediates and aromatic compounds, while PS pyrolysis led to high liquid yield with mostly styrene and alkylated benzenes. The CNTs synthesized from PET and PS also had the lowest purities when compared to those synthesized from PP, HDPE and LDPE. Based on the findings, it is inferred that a higher temperature is needed to achieve high degradation of plastics that contain straight-chained and aromatic structures. Similar observations were also made by Panahi et al. (2019). When real plastic waste was used as the origin of carbon precursors, the feed for the synthesis of CNT was contaminated with Cl, S and N. The presence of PVC could also hinder the formation of CNT (Borsodi et al. 2016). Therefore, it is important to maximise the homogeneity, while minimising the presence of contaminants in municipal plastic waste.

Reaction environment

By varying the reaction conditions and/or additives used during the reaction, it is possible to generate CNM with different structures and compositions. Zhang et al. (2014) analysed the relationship between the reaction conditions and/or additives used with the composition and structure of the CNTs. The group investigated the synthesis of Fe/CNT composites from PP waste catalysed by ferrocene and NaN_3 in a tightly sealed autoclave. The resulting CNTs possessed bamboo-like structures with the Fe nanoparticles observed on the nanotube tips. However, when $NaNH_2$ (instead of NaN_3) was used with ferrocene, the formation of nanoflakes and nanoparticles rich in Fe was observed (Zhang et al. 2014). The replacement of NaN_3 with water yielded one-dimensional worm-like structures and carbon spheres. During the process, the Fe was oxidised to Fe_2O_3 by water, and the metal species experienced magnetic dipole interactions, which led to chain-like structure formation. Similarly, the replacement of NaN_3 by $CO(NH_2)_2$ entailed the conversion of Fe to Fe_3O_4 and subsequently the formation of flower-like structures. The replacement of NaN_3 by S caused the formation of FeS and subsequent formation of flower-like spheres, large flakes and polyhedron structures. Based on the observations, the CNM growth on the catalyst is prone to various factors including the oxidation states of Fe and their interactions.

Recent advances, challenges and future directions

As shown in Tables 5 and 6, the conversion of plastic waste into CNMs is a plausible upcycling method, as the products have higher economic value than the waste. Pyrolysis plays an important role in decomposing the long-chain polymeric structure before the synthesis step. The current emphasis in PtCNM research is placed on CNT synthesis, while the investigations on the potential of graphene synthesis from plastic waste are underway. The conversion of plastics to other CNMs has also been reported. For example, Tripathi et al. (2019) reported hollow carbon spheres synthesis bypassing the plastic-derived hydrocarbon gas over spherical silica template, followed by the dissolution of the template using hydrogen fluoride. The N-doping of the carbon spheres occurred by mixing the plastics feed with melamine before pyrolysis. The addition of potassium chloride resulted in carbon spheres with enhanced graphitisation degree and lower thickness (Yu et al. 2019).

Some researchers have also achieved outstanding results by synthesizing CNM with tuneable optical properties. For example, Song et al. (2019) successfully constructed light-emitting diodes based on carbon dots synthesized from PS waste with tunable photoluminescence. Chaudhary et al. (2021) reported a more environment-friendly synthesis of

photoluminescent carbon dots from local plastic waste with high biocompatibility. The chemo-sensor probe developed using the carbon dots showcased excellent performance in Cu detection in real water samples.

The PtCNM research is still at a very early stage and is still far from the large-scale synthesis of CNM. Recent studies continue to provide more information on different aspects of CNM synthesis from plastic waste. Yao et al. (2021) observed different MWCNT growth patterns during pyrolysis of mixed plastic waste over Ni–Fe metals supported on different porous materials. The bimetal supported on MCM-41 catalyst produced the highest CNT yield with superior quality, due to the high surface area and porosity. On the other hand, poor metal dispersion on BETA catalyst resulted in the lowest MWCNT quality among all the catalysts used. In other words, their work demonstrated the significance of metal–support interaction on the CNT yield and quality. Jia et al. (2020) also investigated the role of such interaction on the catalytic effects of Ni supported on oxides of different rare earth or alkaline earth metals (La, Mg, Sr), which were prepared using the in situ growth method. The moderate interaction between Ni and La resulted in balanced dispersion of Ni nanoparticles, which in turn produced optimum catalyst activity on the plastic-to-CNT conversion. On the other hand, weak metal–support interaction impeded Ni nanoparticles distribution on Sr, while the strong metal–support interaction retarded dissolution of Ni nanoparticles on Mg. These observations indicate the importance of catalyst design in producing CNT with desired morphology, size and yields.

It is interesting to note that some works in Table 5 report the successful synthesis of MWCNT, while the others mentioned the successful synthesis of CNT. In contrast, the concrete evidence of successful single-walled carbon nanotubes (SWCNT) synthesis from plastic waste has not been reported so far. A possible explanation for this observation could be due to the higher reaction temperature required for SWCNT when compared to MWCNT synthesis. The analysis of CNT formation from conventional carbon sources showed that SWCNT growth is more favourable at a higher temperature (900–1200 °C) when compared to MWCNT (600–900 °C). This phenomenon is mostly attributed to the high energy of formation for SWCNT. Besides, the metal nanoparticles must be small enough (below 10 nm) to enable SWCNT formation (Kumar and Ando 2010). As PtCNM research is still in its infancy, conversion of plastic waste to SWCNT will be a great yet rewarding challenge.

Although many studies demonstrate the technical feasibility of plastic waste conversion to CNM, the potential and impacts related to the inclusion of the process in the plastic waste recycling industry are yet to be fully explored. To answer this question, Ahamed et al. (2020) performed a grave-to-gate life cycle assessment on a hypothetical LDPE recycling process, which includes a plastic pyrolysis unit, a pyrolysis oil upgrading system and an MWCNT synthesis system. When compared to a scenario where only plastic pyrolysis oil is produced, another scenario that includes MSWNT synthesis from the pyrolysis gas produces more significant environmental benefits, especially in terms of climate change and fossil depletion. Such benefits are related to the replacement of fossil-based feedstock by plastic-based hydrocarbons in MWCNT synthesis. An MWCNT yield of 4% is sufficient to produce the significant positive effects that compensate for accompanying negative impact related to human toxicity. More life cycle assessments are needed to provide further evidence on the economic and environmental benefits of such a process under different scenarios of local waste generation, as well as parameters for plastic waste pyrolysis and MWCNT synthesis.

Limitations of this study

The bibliometric analysis reported in this study has several limitations. Firstly, WoS was the only source of data used for this analysis. The use of other scholar databases and/or search engines (such as Scopus, Google Scholar, PubMed or China National Knowledge Infrastructure) may lead to the retrieval of the different dataset, and hence, different interpretations of the knowledge structure of PtCNM research. The search strategy (i.e. search string and timespan) could also affect the search results if designed differently. Furthermore, the nodes with minimal occurrences are omitted in the co-authorships and co-occurrence analyses to enable clearer visualisations and simplify the analysis. The change of the minimum number of occurrences in the analyses could also lead to different visualisations and hence different interpretation outcomes. Despite these limitations, this article provides sufficient details of the publication landscape on PtCNM research required to stimulate other scientists' interests in plastic waste valorisation.

Conclusion

This study presented the publication landscape on PtCNM research from 2000 to 2019 based on a bibliometric analysis on publication extracted from WoS. During the study period, an increase in the number of annual publications and annual citations was observed for PtCNM research, which illustrates the enormous scientific interest in developing this research as a potential solution to plastic waste management. Twelve journals are identified as the core journals in this field. The co-citation analysis revealed strong relationships among these journals: Carbon, Journal of Applied Polymer Science, and Applied Catalysis B-Environmental. The early PtCNM research was mostly contributed by scientists from

the USA and Japan. Nevertheless, China is regarded as the largest contributor to PtCNM research from 2000 to 2019 due to the significant contributions of the Chinese Academy of Sciences. Besides, many PtCNM publications acknowledge the funding from the National Natural Science Foundation of China, making this agency the most significant sponsor of PtCNM research. According to the keyword co-occurrence analysis results, the conversion of plastic waste to CNTs over the nickel catalyst is the main focus in PtCNM research. Other important research hot spots in this research topic include graphene synthesis using the CVD method, hydrogen production, as well as the use of various synthesis methods for CNM synthesis from plastics. The influences of catalysts/substrate, temperature, plastic type and reaction environment on the CNM synthesis from plastic waste are explained based on a compilation of publications extracted from WoS. In general, pyrolysis plays an important role in converting plastic to incondensable hydrocarbon gas, which is an important feedstock in CNM formation. More studies are needed to verify the economic and environmental benefits related to the inclusion of CNM synthesis in the plastic waste management sector. Besides, the financial support from the funders and policymakers is vital to stimulate further development in PtCNM research, which could ascertain the role of this technology in regional or global plastic waste valorisation roadmaps.

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Declarations

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