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Review

Research on Modified Carbon Nanotubes in Wastewater Treatment

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Abstract: The application of modified carbon nanotubes (MCNTs) in the field of wastewater treatment has received much attention in recent years. To better understand the research progress and trends of MCNTs in wastewater treatment, this paper presents a bibliometric analysis of the scientific literature on the application of MCNTs in wastewater, highlight how it has evolved over the years, and identify the sectors for which the study could be beneficial. The CiteSpace software was used to perform the analysis of the data in which specific maps were used to represent the network of the relationships among countries, journals, organizations, authors, and keywords related to the investigated topic and subtopics. The results show that in the time frame of the study, most attention has been focused on the functional modification of carbon nanotubes, the study of their application in water treatment, and the study of the patterns and mechanisms of pollutant removal. The recycling of MCNTs and their environmental risks have not been sufficiently studied. In conclusion, the recycling and regeneration methods of MCNTs, as well as the potential ecological, environmental and health risks will be the focus of future research and more research should be invested.

Keywords: modified carbon nanotubes; wastewater treatment; bibliometric; CiteSpace

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1. Introduction

Carbon nanotubes (CNTs) are isomers of carbon and an excellent reinforcing material with high specific surface area, high porosity, high mechanical strength, excellent chemical inertness, and high hydrophobicity [1–4]. Single-walled carbon nanotubes and multi-walled carbon nanotubes are the two most common types of CNTs [5,6]. CNTs were commonly used in thermal conductors, ceramics, energy storage materials, biological applications, and air and water filtration due to it is a one-dimensional quan-tum material with dimensions in the nanometer range [7–11]. It was found that modified carbon nanotubes (MCNTs) i have better anti-pollution properties as well as excellent separation performance, and the modified and functionalized CNTs can improve the hydrophilicity, mechanical properties and bacterial inhibition properties of membranes [12–16]. However, the limited compatibility of CNTs with organic solvents and polymeric matri-ces has limited their application in novel materials. For example, poor dispersion in water, surface inertness, low adsorption efficiency and difficult separation, which can easily cause new environmental risks [17–21].

At present, MCNTs have received widespread attention in the field of water treatment and have been extensively studied in desalination, removal of heavy metal ions, and oil-water separation [1,22–24]. MCNTs are one of the research hotspots in the field of water treatment, mainly focusing on the properties of CNTs themselves and the removal of pollutants [25–29]. However, there is a lack of comprehensive quantitative research on the application of MCNTs in wastewater treatment, as well as the characteristics and

evolution characteristics of the research. Bibliometric analysis is a quantitative analysis of scientific papers in a field using statistical methods to visualize the history and status of research development and to predict emerging trends and explore future research hotspots and directions. CiteSpace is a visual analysis software [30], which is one of the most used bibliometric tools. The information regarding literature journals, authors, institutions, countries, etc., can be visualized and analyzed by CiteSpace, especially for in-depth diversified analysis of keywords, which can provide a more comprehensive understanding of a research field and discover research hotspots and research trends in the field [31,32].

In view of this, the milestones of the research on wastewater treatment with MCNTs were totaled using the bibliometric CiteSpace software, and their research lineage was sorted out and the research hotspots and evolutionary trends were revealed. The main research contents are as follows: (1) Publication trends, sources, journals, discipline distribution as well as main authors, countries, and institutions of research on modified CNTs. (2) Cooperation of main countries and institutions in the field of research on modified CNTs. (3) Summarizing existing hot issues and exploring research prospects and trends based on keyword clustering analysis. The research hotspots and research trends, major authors, institutions, journals, and their cooperation in the field can visually be reflected and it helps to clearly grasp the content and development trend of MCNTs research, which is of great significance to comprehensively understand and promote the research on MCNTs wastewater treatment.

2. Results and Discussion

2.1. Analysis of the Basic Characteristics of the Issued Articles

2.1.1. Analysis of the Number of Articles

The temporal distribution of the number of literatures reflects the development trend of MCNT wastewater treatment research. In the last 20 years, the total number of publications in the field of research on wastewater treatment of MCNTs has increased significantly in general (Figure 1). This indicates that the research on MCNTs in wastewater treatment in general is developing, progressing, and gaining attention. The results of the average annual number of publications showed that the average annual number of publications in the field of MCNTs in wastewater treatment research reaches 48, and the total number of publications was gradually increasing. From 2004 to 2009, the initial stage of MCNTs use in wastewater treatment, the total number of studies was small, averaging 3.5 articles per year. From 2010 to 2015, the research on MCNTs in wastewater treatment started to take off and the number of literatures began to grow steadily, averaging 33.2 articles per year. From 2016 to 2021, the average number of articles exceeded 100 per year and showed a significant growth trend, with a gradual increase in researchers and represented the boom in research on MCNTs in wastewater treatment. However, from 2017 to 2020, with an average of 115 publications per year, its growth trend slowed. This may be due to the recent application of new materials such as gra-phene, biochar, ceramic membrane and metallic glass in wastewater treatment [33–36]. In general, the research literature related to the use of MCNTs for wastewater treatment is gradually increasing. This is because MCNTs, as new wastewater treatment materials, have features such as high permeability, anti-pollution, excellent electrical and adsorption properties, and achieve better separation in water treatment than traditional industrial materials (silica gel, activated carbon, etc.).

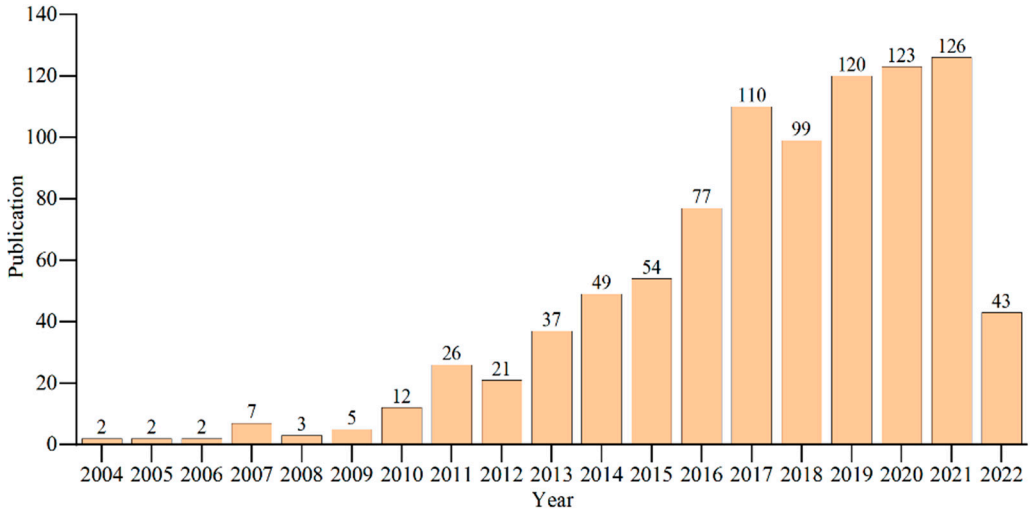

Figure 1. Annual publications on MCNTs.

2.1.2. Analysis of Countries

Visualization of cooperating countries in MCNTs studies (Figure 2) can not only identify the core countries in the research field of wastewater treatment of MCNTs but also reflect the academic exchange and cooperation between research countries in this field. In this study, country was chosen as the object of analysis in CiteSpace, the time slicing was set to "2004–2022", the years per slice was 1, and a threshold of top 50. Finally, the country analysis mapping was obtained (39 network nodes and 41 connections, density of 0.0553). The number of occurrences of the country were represented by the frequency, the position of the state in the field is represented by centrality. The more countries are connected, the more central the country is, the richer the study of the country is, and the more important the location of the country is. China was the country with the highest number of publications in this field (a frequency of 422), which is significantly larger than other countries, which indicates that China contributes significantly to the field of MCNT wastewater treatment, and its development status is in a leading position. Iran was ranked second (a frequency of 156) and India was ranked third (a frequency of 71) (Table 1). In terms of centrality, Saudi Arabia was the highest with 1.04, which indicates its central position within the field of MCNT wastewater treatment and its close relationship with other countries. It also indicates that researchers in Saudi Arabia tend to cooperate with other countries to complete research in the field of MCNT wastewater treatment. In terms of year, China appeared in 2004, which is currently the earliest appearance. China is a large country at the forefront of current economic development, which indicates that research on MCNT wastewater treatment is an important research hotspot at present and even in the future.

2.1.3. Analysis of Institution and Authorship

By using the institutions in CiteSpace as the analysis objects, an institutional analysis graph was obtained (101 network nodes and 67 connections with a density of 0.0133). The nodes in the graph are relatively dense and most of the data are available, while the total number of connections is only 67, indicating that institutional cooperation in the field of MCNT wastewater treatment is relatively frequent and some of them have obvious geographical characteristics (Figure 3a). Cross-institutional research and cooperation in the field of MCNT wastewater treatment need to continue to maintain transnational cooperation and strengthen academic exchanges. Meanwhile, the top ten publishing institutions in the field of wastewater treatment by MCNTs were compiled (Table 2), of which a total of nine institutions published more than 10 papers. Relatively speaking, these institutions are the most numerous institutions in the research field of MCNT wastewater treatment, and six of them are from China, which indicates that the depth and breadth

of research in the field of modified CNTs wastewater treatment by Chinese institutions are more prominent compared with other countries. In terms of publication year, the top 10 universities were all first published after 2010, indicating the rapid progress of MCNT wastewater treatment technology and the relative novelty of the topics. In terms of centrality, the University of Chinese Academy of Sciences (UCAS) has the highest centrality of 0.05, indicating that UCAS is an important bridge to promote cooperation among universities and is a university that has made great contributions in the field of CNTs wastewater treatment and promoted the development and progress of MCNT wastewater treatment. Furthermore, the half-life of the University of Chinese Academy of Sciences is seven, ranking first, which indicates that the research results of the University of Chinese Academy of Sciences have high theoretical value and still have great research prospects in the future.

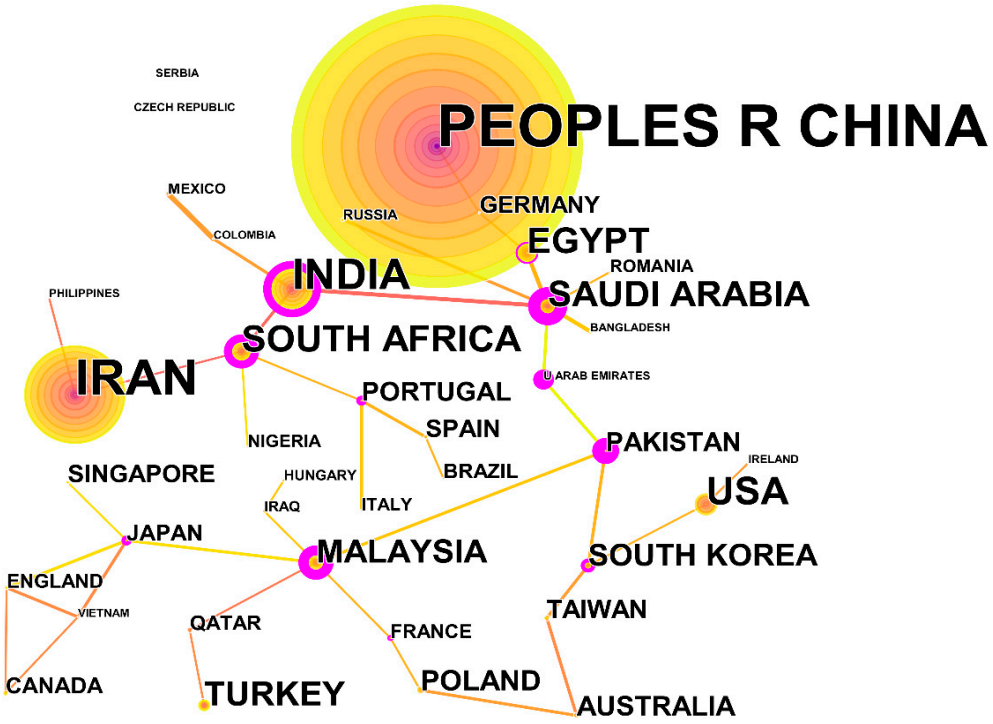

Figure 2. Visualization of cooperating countries in MCNTs studies.

Table 1. Top 10 countries with the most publications in MCNTs studies.

Serial Number	Country	Frequency	Centrality	Year
1	China	422	0	2004
2	Iran	156	0.1	2011
3	India	71	0.79	2012
4	USA	43	0.1	2007
5	SOUTH AFRICA	36	0.62	2014
6	SAUDI ARABIA	33	1.14	2015
7	EGYPT	32	0.19	2017
8	TURKEY	26	0	2015
9	MALAYSIA	23	0.88	2016
10	SOUTH KOREA	17	0.34	2010

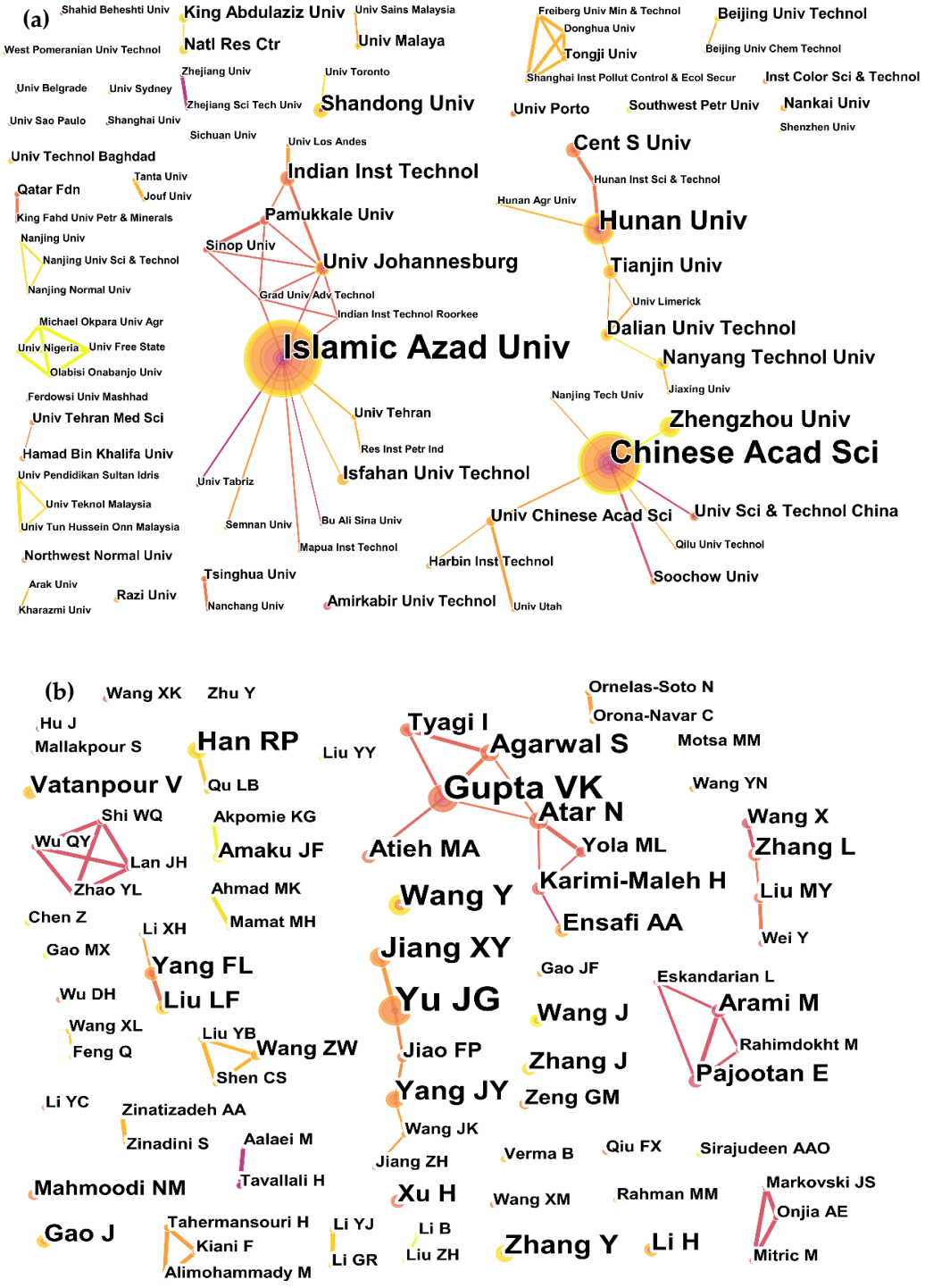

Figure 3. (**a**) MCNT research institute visualization. (**b**) MCNT article authors network visualization.

The core authors in the field of wastewater treatment with MCNTs can be identified by the visual analysis of publication authors, which reflects the academic communication and collaboration of researchers in the field. The top 10 authors in the field of modified CNTs wastewater treatment are shown (Table 3), among which Yu, Yi Wang, and Li Zhang are the most published authors in the field of MCNT wastewater treatment. Although there are many nodes of authors, most of them are independent individuals, and the strength of the connection between authors is generally low (Figure 3b). According to Price's law, when the number of papers published by the core authors of a field reaches more than 50% of the total number of papers in the field, the field can be considered to have formed a stable core

group of authors. In addition, half of the papers in the same field are authored by a group of highly productive authors, and the number of authors in this group is approximately equal to the square root of the total number of all authors [37]. Half of the papers in the same field were written by a group of highly productive authors, and this set of authors is equal to the square root of the total number of authors. The total number of authors engaged in research on wastewater treatment of MCNTs from 2004 to 2022 was 3649 (square root of 60.4), and the number of said core authors was 61. According to statistics, the number of papers by these core authors is 344, accounting for about 35.8% of the total number of papers and has not yet reached 50%, indicating that a stable core author group has not yet been formed in the field of MCNT wastewater treatment. Although the relevant research in the field of MCNT wastewater treatment has continued for nearly a decade, a stable core group of authors has not yet been formed. Therefore, the problem of wastewater treatment with MCNTs needs to be kept in focus and the academic frontier needs to be followed from time to time to obtain more abundant and high-quality research results.

Table 2. Top 10 organizations with the most publications in MCNTs studies.

Serial Number	Institution	Frequency	Centrality	Year	Half-Value Period
1	Islamic Azad Univ	42	0.01	2011	6
2	Chinese Acad Sci	37	0.05	2011	7
3	Hunan Univ	19	0.02	2010	7
4	Univ Johannesburg	13	0	2014	2
5	Zhengzhou Univ	11	0.01	2019	1
6	Dalian Univ Technol	11	0.01	2013	5
7	Isfahan Univ Technol	11	0	2013	5
8	Tianjin Univ	11	0.03	2013	5
9	Beijing Univ Technol	11	0	2015	4
10	Cent S Univ	10	0	2013	4

Table 3. Top 10 authors with the most publications in MCNTs studies.

Serial Number	Author	Frequency	Year	Half-Value Period
1	Yu JG	15	2013	4
2	Wang Y	11	2012	5
3	Zhang L	10	2014	4
4	Jiang XY	10	2015	2
5	Gupta VK	10	2015	1
6	Zhang Y	9	2013	7
7	Agarwal S	7	2015	1
8	Jiao FP	7	2013	4
9	Han RP	7	2019	1
10	Li H	7	2013	4

2.1.4. Analysis of the Distribution of Disciplines and Journals

Discipline association analysis is performed by linking the cited literature to the discipline of the cited literature, whose citations form the research frontier, and the cited literature forms the knowledge base. The JCR journal map option was selected in the overlay map option, and then the information service data were overlaid onto the original citation literature discipline base map by the add overlay feature of CiteSpace to obtain the dual-map overlay map of the discipline distribution involved in the research field of MCNT wastewater treatment (Figure 4a). The left half takes the discipline distribution of the cited literature as the research status of modified CNT wastewater treatment; the right half takes the discipline to which the cited literature belongs as the research basis of modified CNT wastewater treatment. The curves connect the relationship between the research status and the research base, and the inner numbers of the ellipses indicate the number of papers in each discipline. As shown in Figure 4a, the left side of the figure

shows the main journal distribution groups of MCNT wastewater treatment in Web of Science database, and the right side of the figure shows the main cited journal groups. It can be concluded that the research in the field of MCNT wastewater treatment was mainly concentrated in the journal groups of physics, chemistry, material science, medicine, molecular science, and neurology. Citations in the field of MCNT wastewater treatment were mainly concentrated in the journal groups of molecular science, biology, chemistry, physics, mathematics, environmental science, computers, and systematics. Among them, four outward citation paths exist in veterinary medicine and physics in the citation domain on the left side of the figure, and this cluster was the most dominant citation cluster. Additionally, the corresponding chemistry, materials science, and chemistry clusters have the highest number of citations with a z-value of 5.127 when physical science, materials science, and chemistry are used as source journals.

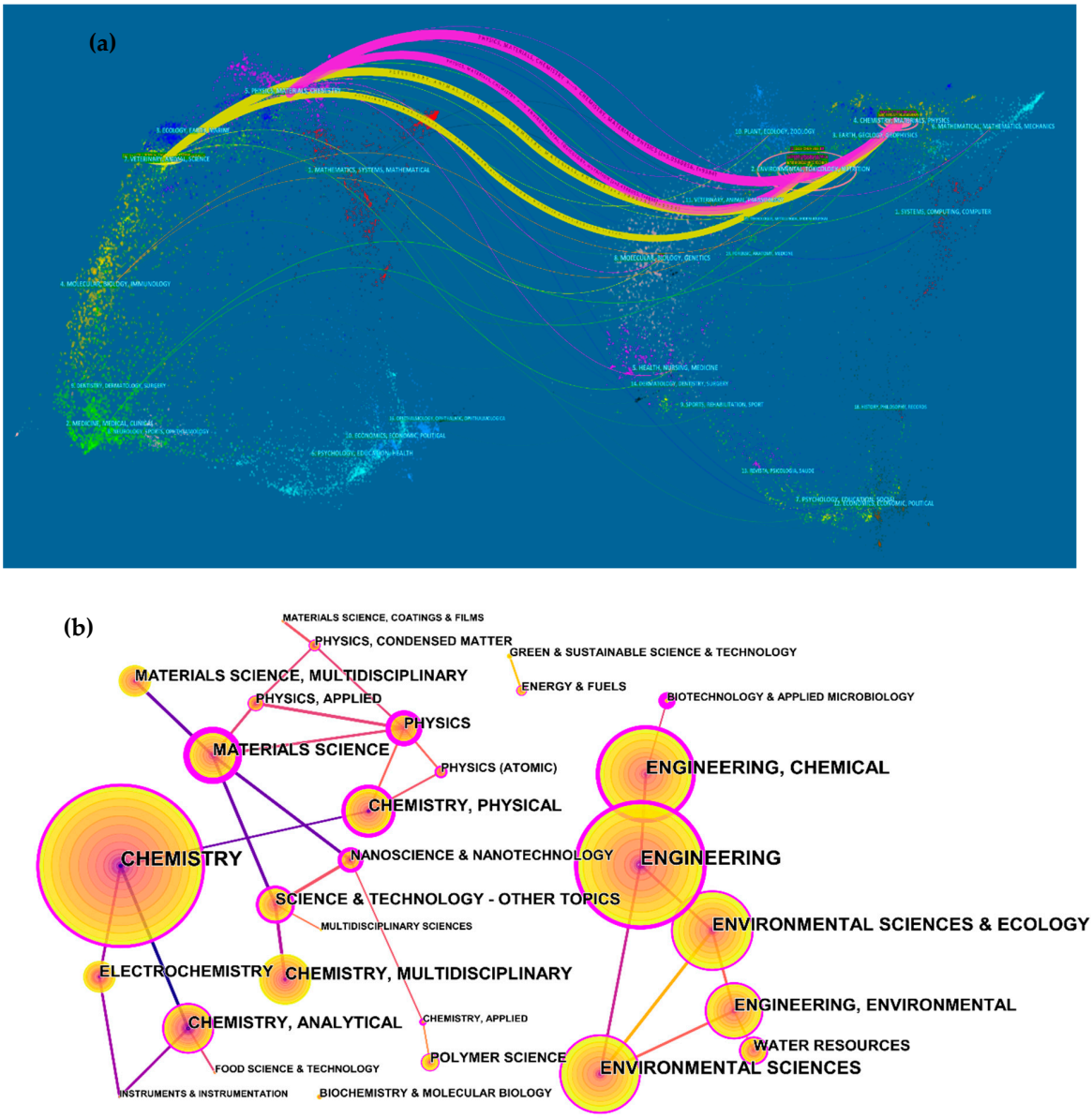

Figure 4. (**a**) The dual map overlay of MCNTs. (**b**) Disciplinary distribution of MCNTs.

In this study, category was selected as the analysis node by CiteSpace software with time slicing "2004–2022", years per slice of 1, a threshold of top 50, and finally, 49 nodes, 70 lines, and a density of 0.0595 were obtained (Figure 4). The top 10 disciplinary postings

are shown with larger nodes and fonts for chemistry and engineering, indicating that the research on wastewater treatment of MCNTs is more prominent in the disciplines of chemistry and engineering, which is related to their own characteristics and roles. There are different connecting lines between different disciplines, indicating that the existing research is not single-disciplinary longitudinal research, but a multidisciplinary cross-fertilization of integrated research topics (Figure 4b), for example, materials science and chemistry, chemistry, multidisciplinary and chemistry, etc. The disciplines ranked above 150 in frequency were chemistry, engineering, engineering, chemistry, environmental science and ecology, and environmental science, which indicates that the research on MCNTs for wastewater treatment is mainly concentrated in the fields of energy, environment, and chemistry (Table 4). The centrality of materials science is 1.09 and ranks first, which indicates that in the study of wastewater treatment of modified CNTs, most of the literature covers the contents related to the discipline of materials science, such as the organization, nature, production process and use efficiency of MCNTs. Secondly, the centrality of chemical engineering ranked second with a value of 0.67, indicating that the basis of the literature research is based on the advantages of MCNTs in environmental protection. As shown by the year, the disciplines of chemistry and analytical chemistry have an earlier year of 2004, indicating that the principles of wastewater treatment of MCNTs were the main research direction at the beginning of study.

Table 4. Top 10 disciplines with the most publications in MCNT studies.

Serial Number	Discipline	Frequency	Centrality	Year
1	Chemistry	396	0.57	2004
2	Engineering	295	0.62	2010
3	Engineering, chemical	226	0.67	2010
4	Environmental sciences and ecology	192	0.26	2011
5	Environmental sciences	190	0.26	2011
6	Engineering, environmental	143	0.31	2010
7	Chemistry, multidisciplinary	133	0	2010
8	Chemistry, analytical	123	0.39	2004
9	Materials science	123	1.09	2007
10	Chemistry, physical	119	0.62	2007

The distribution of the top 10 journals in the field of MCNT wastewater treatment in this study is shown in Table 5. The total number of articles published in the top 10 journals was 24.48%, which did not exceed 25%, indicating that the distribution of the literature was relatively scattered and the research results were not concentrated in a few journals. The average impact factor of the top 10 journals was 6.0525, and the impact factor of some journals even exceeded 10. For example, the impact factors of the Chemical Engineering Journal and the Journal of hazardous materials were 13.273 and 10.588 respectively, which indicates that the research on wastewater treatment with MCNTs is valued and favored by high-level journals.

Table 5. Top 10 journals with the most publications in MCNTs studies.

Serial Number	Journal	Number of Published Papers	Proportion	IF
1	Chemical Engineering Journal	32	3.33%	13.273
2	RSC Advances	27	2.81%	3.361
3	Environmental Science and Pollution Research	26	2.71%	4.223
4	Desalination and Water Treatment	25	2.60%	1.254
5	Journal of Environmental Chemical Engineering	25	2.60%	5.909
6	Journal of Molecular Liquids	22	2.29%	6.165
7	Chemosphere	21	2.19%	7.086
8	Journal of Hazardous Materials	21	2.19%	10.588
9	Electrochimica ACTA	18	1.88%	6.901
10	International Journal of Electrochemical Science	18	1.88%	1.765

2.2. Analysis of Research Hotspots

2.2.1. Analysis of Keyword Network

In this study, keyword analysis and visualization tools of bibliometrics were used to analyze the research topic [38,39]. The keywords in the field of modified CNTs wastewater treatment were analyzed, and keyword co-occurrence mapping was generated with 145 network nodes and 171 connected lines with a density of 0.0164 (Figure 5). The keyword nodes are composed of different colors, the studies of the keywords at different time periods are represented by the color change from inside to outside, and the closer the color of the outermost circle is to purple, the closest to the current study is indicated. The size of the keyword node circle and the size of the label represent the frequency of the keyword, i.e., the higher the frequency of the keyword, the larger the node circle and label of the keyword. The thicker the line, the higher the frequency of co-occurrence between the keywords, and the color of the line represents the year of co-occurrence between the keywords. The top 20 keywords in terms of frequency are listed (Table 6). The number of occurrences of the keyword are represented by the frequency, and the position of the keywords in the field are represented by the centrality. The description of centrality can be seen in the figure and reflects the keyword concatenation, i.e., the more keywords are concatenated, the higher the centrality of the keywords, the richer the keyword research is, and the more important the position of the keywords.

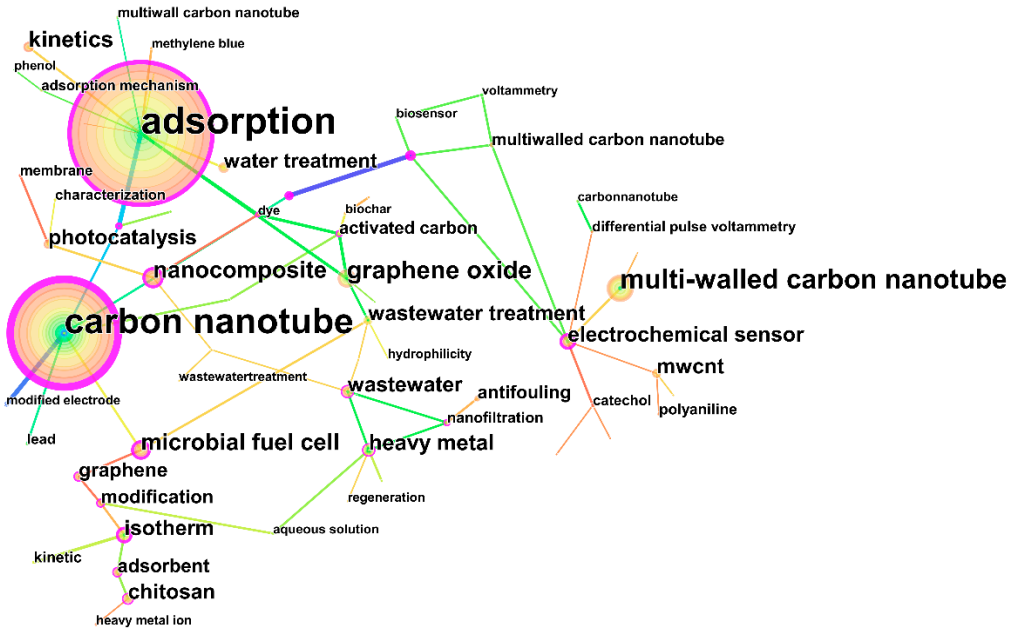

Figure 5. Keywords network co-occurrence of MCNTs.

The keyword "adsorption" is the most frequently occurring term in this field (162), which indicates that the function of adsorption is the main research basis and has received much attention from scholars (Table 6). CNTs were widely used for the removal of pollutants in water treatment because of their good adsorption properties [3,40]. Next, "carbon nanotube" ranked second in frequency (123). The studies have shown that the research on CNTs dates to a paper by Sumio Iijima [11] published in Nature in 1991. The frequency of "multi-walled carbon nanotube" ranked third, indicating that "multi-walled carbon nanotube" has become a hot spot for research. The term "multi-walled carbon nanotube" was widely used in the preparation of composite materials [40]. By centrality, "wastewater" is the highest (0.94), which indicates a central position within the field of MCNT wastewater treatment. In recent years, with the in-depth research, modified CNTs have been used in oil-containing wastewater treatment, printing and dyeing wastewater treatment, and pollutant removal from domestic wastewater [24]. The half-life represents the duration of

the keyword's research value, and the half-life of "carbon nanotube" is the highest (12), indicating that CNTs have great research space in the future.

Table 6. Top 20 keywords with the most publications in MCNTs studies.

Serial Number	Frequency	Centrality	Keyword	Year	Half-Value Period
1	162	0.78	adsorption	2010	8
2	123	0.33	carbon nanotube	2006	12
3	41	0.33	multi-walled carbon nanotube	2010	8
4	29	0.43	graphene oxide	2013	5
5	26	0.94	wastewater	2012	7
6	25	0	nanocomposite	2016	4
7	22	0.03	microbial fuel cell	2014	5
8	20	0.43	water treatment	2016	3
9	20	0.39	heavy metal	2012	4
10	20	0.18	electrochemical sensor	2015	4
11	19	0.13	kinetics	2016	4
12	18	0.13	photocatalysis	2012	7
13	18	0.03	wastewater treatment	2012	6
14	17	0	mwcnt	2016	3
15	17	0.27	isotherm	2013	4
16	16	0.25	graphene	2010	9
17	12	0	chitosan	2016	4
18	12	0.57	modification	2017	3
19	11	0.37	multiwalled carbon nanotube	2012	8
20	10	0.17	adsorbent	2018	3

2.2.2. Analysis of Keyword Clustering

The keywords of the research topics of MCNT wastewater treatment in the last 15 years were clustered by LLR algorithm and 11 clusters were derived, as shown in Figure 6, and the information of each cluster is shown in Table 7.

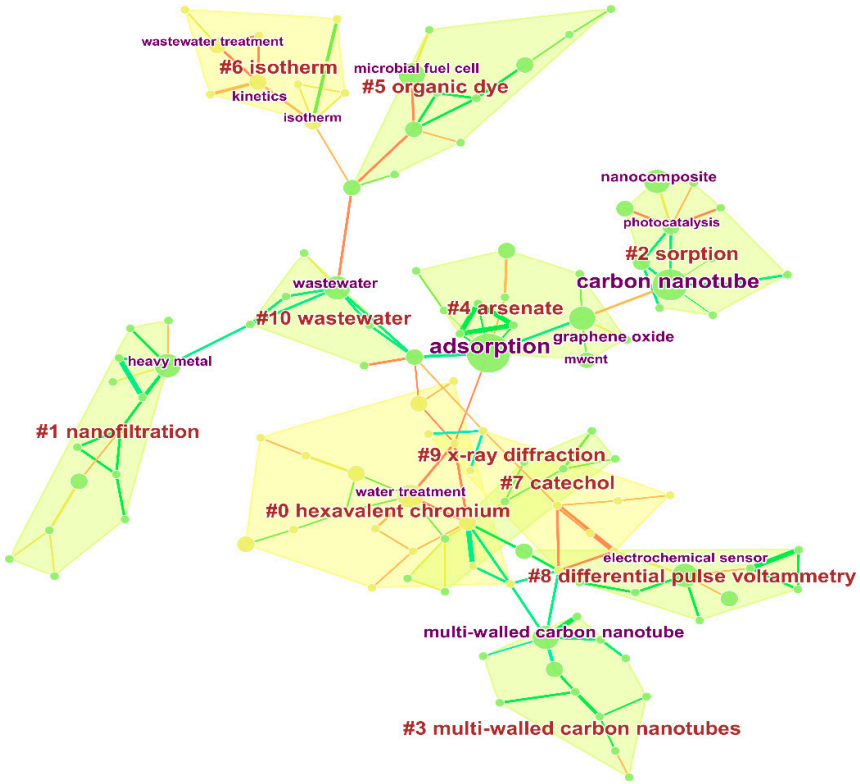

Figure 6. Analysis of MCNT keywords co-occurrence clustering.

Table 7. Keywords information in MCNTs studies.

Serial Number	Number of Nodes	Mean Contour Value	Average Year	Main Content		
0	16	0.813	2016	water treatment	chitosan	multiwalled carbon nanotube
1	14	0.971	2015	heavy metal	antifouling	nanofiltration
2	12	0.979	2014	carbon nanotube	nanocomposite	photocatalysis
3	12	0.982	2013	multiwalled carbon nanotube	sensor	phenol
4	12	0.931	2015	adsorption	graphene oxide	mwcnt
5	11	0.909	2015	microbial fuel cell	graphene	modification
6	10	0.979	2017	kinetics	wastewater treatment	isotherm
7	9	0.892	2015	modified electrode	multiwall carbon nanotube	catechol
8	9	0.915	2014	electrochemical sensor	polyaniline	voltammetry
9	8	0.978	2016	nanoparticle	characterization	Wastewater treatment
10	7	0.902	2013	wastewater	dye	desalination

The studies of MCNT wastewater treatment were grouped into 11 categories, namely #0 hexavalent chromium, #1 nanocomposite, #2 sorption, #3 multi-walled carbon nanotube, #4 arsenate, #5 organic dye, #6 isotherm, #7 catecho, #8 differential pulse voltammetry, #9 x-ray diffraction, and #10 wastewater (Figure 6). Based on the cluster location analysis, the 11 research clusters were classified into three types, among which the first type of clusters is not the mainstream area of wastewater treatment research for MCNTs because of their deviation from the main body and the small number of studies. This type is mainly a cluster with "#6 isotherm" as the clustering keyword, and this cluster gathers "kinetics", "wastewater treatment ", and "isotherm". The second type is relatively independent from the main study at a certain distance, but has a certain amount of research, this type includes the "#8 differential pulse voltammetry" keyword. The third type of taxa are those that overlap with each other and have close inheritance and derivation relationships. They include "#0 hexavalent chromium", "#1 nanocomposite", "#2 sorption", and "#3 multi-walled chromium". "#3 multi-walled carbon nanotube", "#4 arsenate", "#5 organic dye", and "#7 catechol", "#9 x-ray diffraction", and "#10 wastewater" as cluster labels, and the largest number of clusters is in the research direction of wastewater treatment. The research was about the application of MCNTs for the treatment of various wastewaters, such as heavy metal wastewater (chromium), printing and dyeing wastewater, phenolic wastewater, benzene o-diphenol, etc.

The different clusters cover different keywords (Table 7). The average profile value for each cluster is greater than 0.7, indicating that the effect of each cluster is consistent with the study. The average time to publication of keywords within clusters was represented by the average year, which characterized whether the topic was close to the frontier. For example, the average year (2017) for Cluster 6 indicated that the clusters were relatively close to cutting-edge research. The average years of Cluster 3 and Cluster 10 were 2013, indicating that the research themes of Cluster 3 and Cluster 10 are relatively more traditional. In addition, it is also possible to continue to subjectively classify and merge themes based on the similarity of research topics.

2.3. Analysis of the Evolution of Research Trends

2.3.1. Analysis of the Evolution of the Study

To understand the evolution of keywords in the field of wastewater treatment research on MCNTs, a temporal evolution was analyzed. The time zone was chosen as the analysis node with a time slicing of "2004–2022", a years per slice of 2, and a threshold of g-index = 5.

Keywords with smaller nodes were hidden to generate the current graph. The keywords with smaller nodes were hidden and the graphs were generated (107 nodes and 112 lines with a density of 0.0197). To further elucidate the evolution of the research themes over time, a timeline diagram of the co-citation network was plotted (Figure 7). These cluster labels were further analyzed and categorized into three areas of research: (1) research around the nature, function, and preparation of catalysts of CNTs themselves, containing "carbon nanotube", "adsorption"; (2) the application of modified CNTs in water treatment, which is reflected in the graph of "multi-walled carbon nanotube", "walled carbon nanotube", "heavy metal", "wastewater treatment", "wastewater", "water treatment", "wastewater", "nanocomposite", "kinetics", "membrane", and "modification", mainly focusing on the removal of contaminants from water by MCNTs and the composition of MCNTs composite membranes.

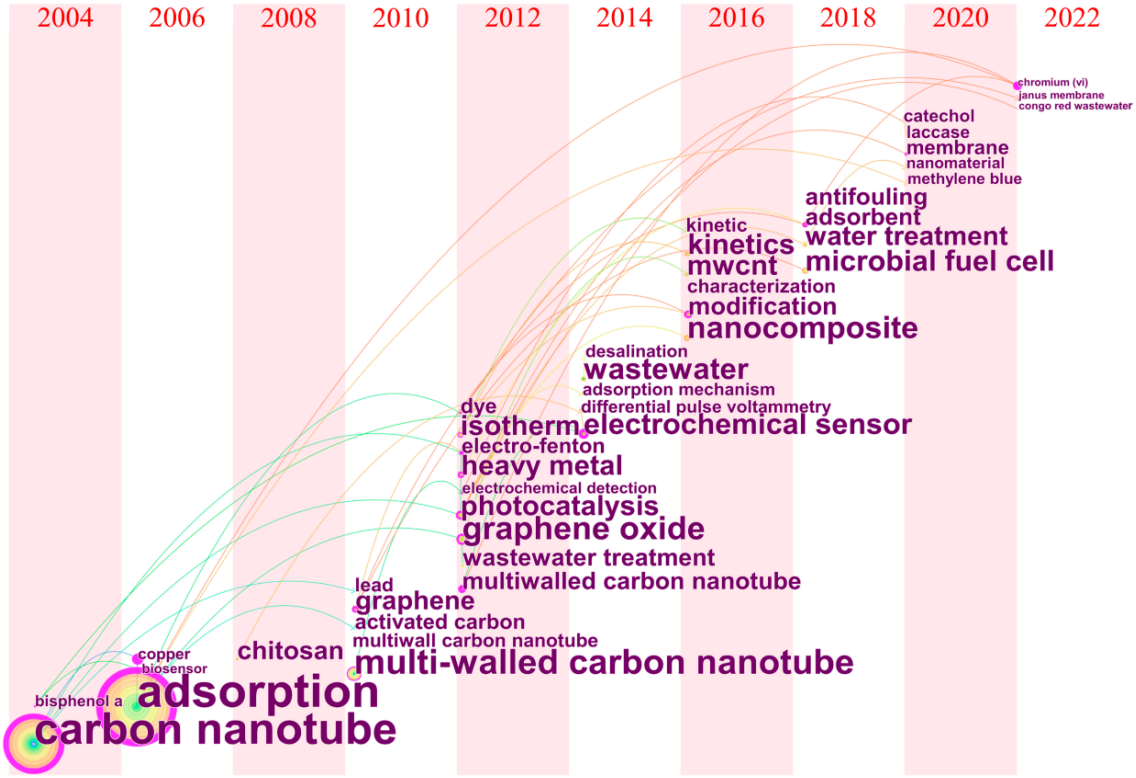

Figure 7. Keyword co-occurrence time zone view of MCNTs.

In terms of time, the overall evolution can be divided into four stages: the initial stage, the initial development stage, the rapid development stage, and the stable development stage. In the initial stage (2004–2009), the research themes were focused on the exploitation of the properties, composition, and functions of CNTs themselves, focusing on the adsorption mechanism [41,42] and the use of CNTs for water treatment by exploiting their adsorption properties [41,42]. After that, the research entered the early development phase (2010–2013), with research themes distributed into multi-walled carbon nanotubes, heavy metals, wastewater treatment, and photocatalysis, indicating that this phase focused on the use of modified multi-walled carbon nanotube to remove heavy metal from wastewater [1,43–45], and the focus on multi-walled carbon nanotubes continued to be high from 2010 to 2013. Over time, the research entered a rapid development phase (2014–2019), with research themes distributed into microbial fuel cells, electrochemical sensors, and nanocompounds. After 2020, the research entered into a stable development stage, with research themes covering membranes, methylene blue, nanomaterials, catechol, mechanisms, etc., reflecting the research themes covering technical and environmental fields in response to

the previous stage of water treatment. This stage mainly focuses on the application of MCNTs on membranes [46–48], methylene blue removal [49,50], etc.

2.3.2. Analysis of Research Frontiers

The time series of key frequency words can be analyzed to clearly reveal the trend of the macroscopic view on research hotspots, to understand the evolutionary dynamics of MCNT wastewater treatment research hotspots, which is conducive to the judgment of the development trend and future directions (Table 8). The blue line indicates the time interval, the red line indicates the cycle of burst keywords, and the end points of the red line indicate the beginning and end of each burst time interval [51]. Strength represents the emergence intensity of the keywords, and the greater the emergence intensity of the keywords, the higher the research intensity and the more the output of the research in that period. The keywords "bisphenol a", "carbon nanotube" and "modified electrode" appeared from 2004 to 2011, with a highlighted intensity of 1.9081, 3.4487, and 1.9755, respectively (Table 8). According to the timeline mapping hotspots of research from 2004–2011, the removal of bisphenol using MCNTs [52,53] and the application of MCNTs in electrodes [54] started to be focused on in this period. The long duration of the emergent words indicates that these keywords were widely followed by researchers and active research activities during this period. The emergent words from 2012 to 2017 were "heavy metal", "phenol", "multi-walled carbon nanotube", and "kinetic", with emergent intensities of 3.758, 3.0731, 2.7801, and 2.7769 respectively. This showed that the research on MCNT wastewater treatment tends to be refined and intensified. It mainly focuses on the removal of heavy metals from wastewater [1,55], the removal of phenolic wastewater [56,57], the application of "multi-walled carbon nanotube" in water treatment [58–60], and the kinetic model of MCNTs [60,61].

Table 8. The burst word data for MCNTs studies keywords.

Keywords	Year	Strength	Begin	End	2004–2022
bisphenol a	2004	1.3316	2004	2015	
modified electrode	2004	2.5701	2005	2010	
biosensor	2004	1.8612	2007	2015	
carbon nanotube	2004	1.4233	2007	2009	
lead	2004	1.6444	2011	2017	
heavy metal	2004	4.5838	2012	2014	
wastewater treatment	2004	1.8985	2013	2019	
voltammetry	2004	2.2202	2013	2015	
carbonnanotube	2004	1.6725	2014	2017	
isotherm	2004	2.6205	2016	2018	
graphene oxide	2004	2.8097	2016	2018	
aqueous solution	2004	2.0775	2016	2017	
kinetic	2004	2.8563	2017	2018	
characterization	2004	2.3764	2017	2018	
multi-walled carbon nanotube	2004	4.2505	2017	2018	
photocatalysis	2004	3.1537	2018	2020	
water treatment	2004	3.0471	2018	2019	
microbial fuel cell	2004	2.9558	2018	2019	
polyaniline	2004	1.8794	2019	2022	
wastewater	2004	2.6343	2019	2022	
mwcnt	2004	1.6035	2019	2022	
antifouling	2004	2.8326	2019	2022	
membrane	2004	1.875	2020	2022	
nanocomposstte	2004	3.0564	2020	2022	
kinetics	2004	1.5081	2020	2022	

This indicates that the research on MCNTs in wastewater treatment has entered a period of deep development, and many comprehensive and cutting-edge studies have

emerged and received wide attention. With the passage of time, the focus on the use of modified CNTs in water treatment has begun. Focus on the application of modified CNTs for anti-pollution in water treatment [62,63], modified CNT composite membranes [47,62,63], modified CNT nanomaterials [64,65], and modified CNTs for oil-water separation [66,67].

3. Data and Methods

3.1. Data Collection

Web of Science (WOS) is a global academic information database platform containing more than 12,400 authoritative and highly influential international academic journals covering natural sciences, engineering, social sciences, arts, and humanities dating back to 1900 [68]. High quality bibliographic records are provided by WOS and are widely used for bibliometric studies [69]. Therefore, WOS was chosen as the bibliographic database to be searched in this study [68]. The search was conducted on 2022-6-18 with subject search topic terms = modified and ("CNT*" OR "carbon nanotube*") AND ((waste water) OR (sewage) OR (Wastewater)), Timespan = all year. Therefore, 1026 studies were initially identified and after filtering based on literature exclusion review, books, and conference proceedings, etc., the dataset for this study was reduced to 918 records.

3.2. Methods

The literature data were analyzed in terms of circulation, countries, institutions, journals, research directions, and research hotspots using Excel 2016, OriginPro 2022b Beta, and CiteSpace 5.8 software. Among them, the data records retrieved were counted using Excel 2016 software and Origin analyzed the trend of literature volume by year and country. The co-citation intensity of journals and institutions in this research area was analyzed using CiteSpace software, and keywords in different time periods were detected with the help of time series graph function; the research hotspots in different time periods were classified through the clustering of keywords, and finally, a visual analysis of the evolution of this research area was formed [70,71].

4. Conclusions

The bibliometric approach was used in this study, the articles published in the last 20 years on the research progress of wastewater treatment of MCNTs were summarized, and the trend of articles in this field in the last 20 years was analyzed. The following conclusions were obtained:

(1) The progress of modified CNTs wastewater treatment research is broadly divided into three stages: the starting research stage (2004–2009), the stable development stage (2010–2015), and the rapid development stage (2016–present). The University of Chinese Academy of Sciences has made great contributions to the field of MCNT wastewater treatment and promoted the development and progress of MCNT wastewater treatment. Yu JG, Wang Y, and Zhang L were the scholars with the largest number of publications in this field and a more complete system of cooperation has been formed between the institution and the authors. The disciplines of chemistry and analytical chemistry appeared the earliest, indicating that the principles of wastewater treatment of MCNTs were the initial research direction. The top three journals in the field of MCNT wastewater treatment research in terms of number of articles published were Chemical Engineering Journal, RSC Advances, and Environmental Science and Pollution Research.

(2) The term "adsorption" had the highest frequency in this field, which indicates that the function of adsorption is the main research basis and has received the attention of many scholars.

(3) The research in the field of MCNT wastewater treatment was mainly focused on the pollutant removal law and removal mechanism, the analysis of modified function, and its applications in water treatment, such as oil-water separation, printing and dyeing wastewater, removal of heavy metals, and organic pollutants, etc. In addition, the research

on the recovery and recycling of modified carbon nanotubes and the environmental risk should be further strengthened.

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