Progress of Biogas Upgrading Based on Citespace Visual Analysis

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In this paper, CiteSpace (6.1.R6) software was used to visualize and analyze the number of papers, terminology, countries, institutions, and collaborative networks of authors in the field of biogas upgrading in the years 2009 to 2023 to provide theoretical references for related researchers and departments. The progress of biogas upgrading research is divided into three phases: 2009 to 2011 (Phase I), 2012 to 2015 (Phase II), and 2016 to 2022 (Phase III). The first phase laid the theoretical foundation for the later phase, which included the fields of biomethanation and variable pressure adsorption. The physical/chemical biogas upgrading technology in the second phase gradually matured. The number of research papers published in the third phase increased rapidly, and with the rapid increase in the number of published research papers, which accounted for 73.2% of the total amount of research and statistical literature, this phase of biogas upgrading technology research is even more mature. In addition, the field of biogas upgrading has a small network of cooperation among researchers, and no close cooperation exists among the countries and research institutions involved. Under the trend of carbon emission reduction, "life cycle assessment" and "biomethanation" clusters have become the current research frontiers in the field of biogas upgrading.

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INTRODUCTION

China is striving to achieve carbon peak by 2030 and carbon neutrality by 2060 (Fu *et al.* 2021). This goal provides much room for the development of China's biomass energy industry and has a far-reaching impact on the transformation of China's energy structure. Biomass is an energy source that uses plant photosynthesis to convert solar energy into chemical energy stored in living organisms (Cao 2012). At present, biomass energy utilization technologies mainly include biomass power generation, biomass liquid fuels, biogas, and biomass solid briquettes (Ma *et al.* 2019). Biogas is a relatively mature biomass utilization technology and is one of the important ways to solve the current energy challenges facing the world (Kapoor *et al.* 2019).

The China Rural Statistics Yearbook 2022 indicates that by the end of 2020, China's biogas users will have reached 30.07 million households, with the number of large-scale biogas projects having reached 93,481 nationwide. As biogas industry policies and other guarantee systems have gradually improved, the biogas industry has undergone a shift from primarily domestic biogas to large-scale biogas projects (Luo *et al.* 2022). Crude biogas contains 40 to 75% methane, 15 to 60% carbon dioxide, and water vapor, nitrogen, ammonia, hydrogen sulfide, siloxane, and hydrocarbons (Muñoz *et al.* 2015; Dannesboe *et al.* 2019; Santos *et al.* 2020; Atelge *et al.* 2021; Benato and Macor 2021; Carranza-Abaid *et al.* 2021). When the methane content of the biogas is 70%, the calorific value is about 21.5 MJ/Nm^3 (Wobbe index) (Nguyen *et al.* 2021), which is significantly lower than the calorific value of natural gas (35 MJ/Nm^3). In addition, some of the biogas contains components such as hydrogen sulfide (the fermentation matrix contains sulfates) and siloxanes (the fermentation matrix contains cosmetic laundry detergents, etc.), which not only affect the calorific value of the biogas, but also contain combustion products that are harmful to pipelines, gas engines, compressors, and cogeneration plants (Muñoz *et al.* 2015; Angelidaki *et al.* 2018; Angelidaki *et al.* 2019; Stolecka and Rusin 2021). Biogas upgrading solves the problem of poor quality of crude biogas, which is of great significance in promoting the development of the biogas industry.

Biogas upgrading technologies mainly include physical/chemical and biological methods. Physical/chemical purification techniques mainly include physical/chemical absorption (Sethupathi *et al.* 2017; Augelletti *et al.* 2020), pressure swing adsorption (PSA) (Abd *et al.* 2021), and membrane (Miehle *et al.* 2021) and cryogenic separation (Miehle *et al.* 2021). At present, the physical/chemical purification technologies mentioned above have been commercialized (Kapoor *et al.* 2019). Studies have shown that PSA can selectively adsorb carbon dioxide by using solid adsorbents or molecular sieves (Ali Abd and Roslee Othman 2022). These can increase methane content of biogas up to 97 percent (Kim *et al.* 2015; Alvarez-Gutierrez *et al.* 2016). Membrane separation technology is a highly promising technology for biogas upgrading, with new plants for membrane separation units growing by 82% over the 2015 to 2019 period (Nguyen *et al.* 2021). The principle of the cryogenic separation method is that the liquefaction of biogas occurs under certain temperature and pressure conditions (Abd *et al.* 2021), and the purification of biogas is carried out by utilizing the difference in the condensation temperatures of CO_2 and CH_4, which has the advantages of high purification efficiency and CO_2 recovery rate (Hashemi *et al.* 2019). Typically, the distillation process occurs at 103 K (temperature) and 80 bar (pressure). At this point, methane is preferentially distilled out (Porpatham *et al.* 2018; Sun *et al.* 2015). Two main types of biogas upgrading technology based on biological methods exist: one is the use of microalgae or photosynthetic bacteria to fix impurity gases such as CO_2 in biogas. Chlorella can achieve a carbon dioxide removal rate of 69.34±6.62% (Wang *et al.* 2023). Microbial electrolysis cell systems can increase CO_2 fixation by 83.3% (Sun *et al.* 2023). The other is the use of hydrogenotrophic methanogens to convert CO_2 to CH_4, thereby boosting CH_4 content (Antukh *et al.* 2022; Spyridonidis *et al.* 2023).

Data visualization software allows quantitative analysis of trends, research themes, and collaborative networks in a particular research area. CiteSpace software was developed by Prof. Chaomei Chen. The lower version of CiteSpace software requires the user to additionally configure the JAVA environment on the computer operating system. However, according to the software introduction, higher versions of CiteSpace, such as version 6.1.R6, allow the user to not configure the JAVA environment. CiteSpace can use the knowledge graph of the subject's history to display important publications, research hotspots, and future frontiers of the research subject (Chen 2006). In addition, CiteSpace allows users to visualize the evolution of a research area over time using timeline diagrams compared to other software.

Currently, there are many studies on biogas upgrading and reforming, but they lack systematic exploration and analysis, which is not conducive to the sustainable development of the biogas industry. Therefore, this paper uses CiteSpace software to visually analyze the relevant literature in the field of biogas upgrading from 2009 to 2023. Analyzing the annual publication volume of papers in the field of biogas upgrading, the annual new output of terms, the collaborative network of authors/countries/institutions, the keyword clustering elucidates the research hotspots in the field of biogas upgrading with a view to providing references for researchers in the field of biogas upgrading

EXPERIMENTAL

Data Sources and Processing

The data in this paper were obtained from the Web of Science core collection database, and the advanced search function was used to retrieve data from 2009 to 2023 with the subject Topic (TS)=(biogas upgrading) OR TS=(biogas purification methods) OR TS=(biogas purification) OR TS=(biogas impurity removal) OR TS=(removal of carbon dioxide from biogas) as the subject to retrieve data from 2009 to 2023. A total of 2,779 documents were retrieved. The category refinement function was used to filter out areas relevant to this paper, including energy fuels, environmental sciences, biotechnology and applied microbiology, green sustainable technologies, multidisciplinary chemistry, chemical physics, and literature related to the field of engineering chemistry. Papers that met one of the following criteria were retained as the base data for this paper: the abstract clearly states that the paper's research is on the purification of biogas by physical, chemical, or biological methods; the research is on the separation of impurity gases, such as carbon dioxide, from methane/carbon dioxide mixtures or simulated biogas; and the study of the biogas or simulated biogas upgrading process is done by methods such as modeling and process simulation (Aspen Plus software). After a rigorous screening process, a total of 1084 papers were retained

Data Analysis Methods And Parameter Settings

According to the research content in the node type interface, select "Author", for author collaborative network analysis; select "Country" and "Institution" for collaborative networks in different countries; select "References" for cluster analysis of cited literature using keywords as the source of clustering labels to analyze the research frontiers in the field of biogas upgrading; and select "Term" for nominal terms analysis. In addition, according to the running results of Citespace use Excel software to draw annual publication volume of papers and new terms emerge every year in the field of biogas upgrading. The parameter settings of Citespace software are as follows: the "Time Slicing" is set to 1 year, the two parameters of "Text processing" and "Links" are kept at the default settings, and the "Selection Criteria" adopts the g-index algorithm (k=25).

RESULTS AND DISCUSSION

Annual Number of Published Papers

After a rigorous selection process, a total of 1,084 papers in the field of biogas upgrading were collected in the Web of Science Core Collection between 2009 and 2023.

During this period, the number of papers published generally showed three phases: Phase I (2009 to 2011), during which the annual number of published papers was essentially flat. At this stage, the number of papers published was low at about 14 per year, and the research content of the literature covered most of the keywords or research topics obtained from the cluster analysis, such as life cycle assessment, fuel cell, Aspen Plus simulation, pressure swing adsorption, biomethanation, siloxane, economic analysis, and pilot scale. The papers published during this period laid the theoretical foundation for later research on biogas upgrading. The second period is 2012 to 2015, during which the annual number of papers published exhibited a clear upward trend. The papers published in this phase represent 20.79% of the total number of publications (2009 to 2022). The study shows that the physical/chemical biogas upgrading technology reached a mature stage during this period (Muñoz *et al.* 2015). For example, Serejo's research showed that the microalgae-bacteria bioreactor removes 80% of the CO_2 and 100% of the H_2S from the biogas when the ratio of external recirculated liquid/biogas is 10. (Serejo *et al.* 2015). In the third phase, from 2016 to 2022, 794 papers were published, accounting for 73.25% of the counted literature. According to Fig. 1, the number of annual papers published in this period shows a small cycle of a rising-declining-again riding trend every two years. However, the number of papers published in 2020 was notably lower than in 2021, but it is still higher than that of the second stage.

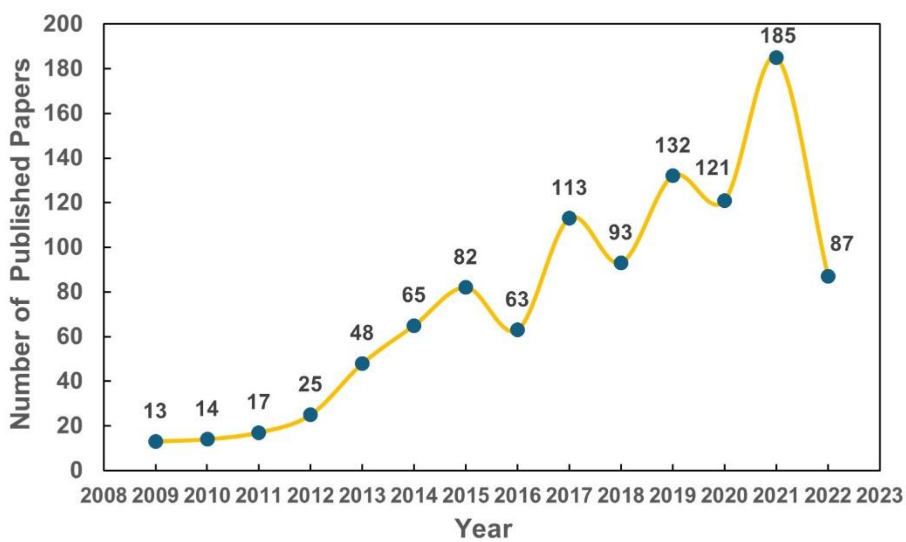

Fig. 1. Annual publication volume of papers

Analysis of Nominal Terms

The term represents a particular research direction in the field of biogas upgrading, and the term co-occurrence indicates a situation where two terms appear in multiple papers at the same time. The analysis of the noun term co-occurrence network can clearly grasp the frontiers and hot spots in the field of biogas upgrading during the research period. In this paper, CiteSpace software was used to count the noun terms in 1,084 papers, and a total of 629 noun terms were captured. By analyzing the number of occurrences of these terms, this paper found that 380 terms were used 3 times or less, accounting for 60.41% of the total number of terms. This finding indicates that most of the terms in the terminology co-occurrence network are less applicable and not widely recognized by peers. Of these

380 terms, 77 terms appeared between 2021 and 2023, and 28 new terms appeared in the first quarter of 2023. The relatively recent appearance of these terms has not yet demonstrated their importance, and researchers should continue to pay attention to these terms, especially those that appeared in the first quarter of 2023.

In addition, the relationship between the annual new output of terms (Fig. 2) and the annual publication volume of papers shows that the trend between the two is different, and the number of new terms per year is relatively stable. This finding indicates that the current trend of research in the field of biogas upgrading is relatively stable. A total of 9 clusters were obtained in this study (Fig. 3), and clusters were found by studying these 9 terms. Except for "add H_2" (which appeared in 2012), all term clusters appeared between 2009 and 2011, which is consistent with the conclusions drawn in the previous section: the papers published between 2009 and 2011 laid the theoretical foundation for the field of biogas upgrading. The study also found that terms used more than 100 times (Table 1) were all concentrated between 2009 and 2010, which is consistent with the previous conclusions. In addition, the results of the study showed four turning points (betweenness centrality ≥ 0.1) in the nominal term co-occurrence network.

Table 1. Terms Used More than 100 Times

Ranking	Counted	Year	Term
1	332	2010	Biogas Upgrading
2	250	2009	Biogas
3	206	2009	Carbon Dioxide
4	168	2009	Anaerobic Digestion
5	135	2009	Biogas Purification
6	133	2010	Biomethane
7	118	2010	Degrees C
8	111	2010	Upgrading Process
9	101	2009	Hydrogen Sulfide

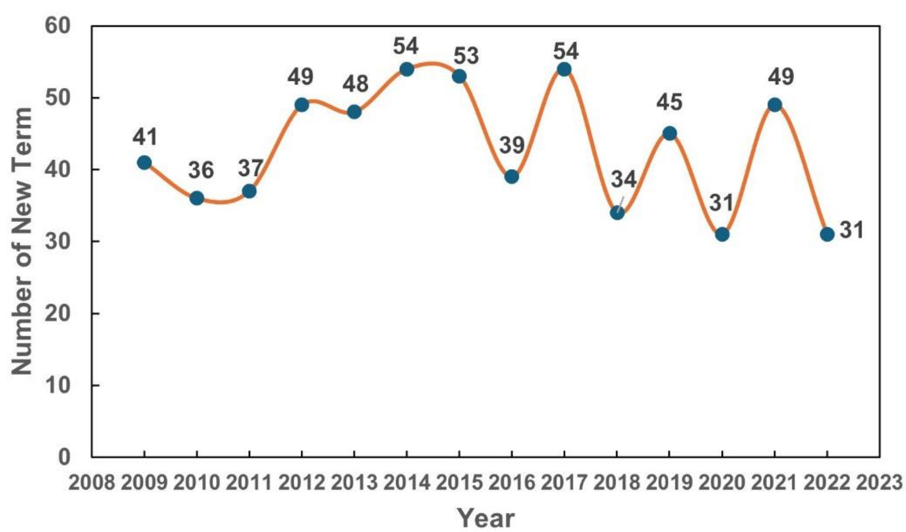

Fig. 2. Number of new terms

CiteSpace software uses betweenness centrality to measure the importance of nodes in the overall network. Turning points are nodes with high betweenness centrality, which are hubs that connect different groups in the network. Carbon dioxide had the largest betweenness centrality (0.18), and the betweenness centrality of anaerobic fermentation, chemical adsorption, and biogas purification were 0.16, 0.12, and 0.11, respectively. These terms may be highly influential in the field of biogas upgrading. The most used term was biogas upgrading, which was used 332 times in the 1,084 documents of this study and 73 times during 2021.

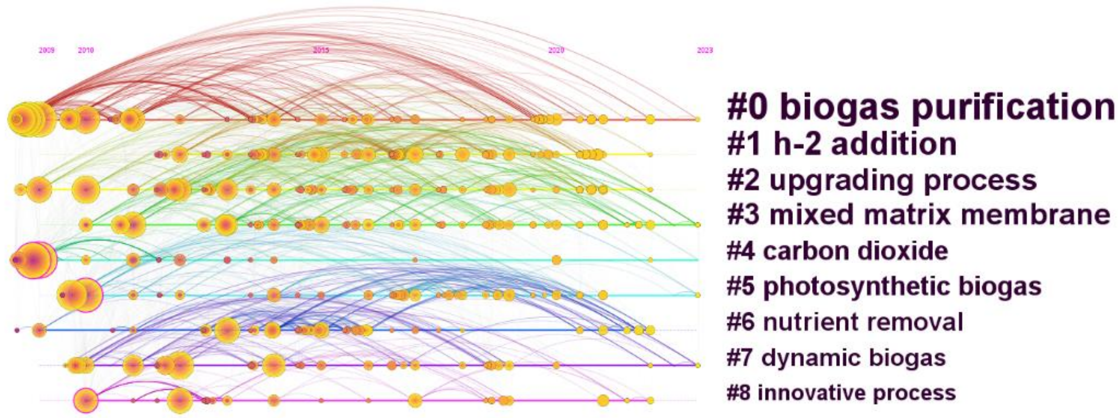

Fig. 3. Term cluster timeline

Analysis of Author Collaboration Networks

The analysis of the authors' cooperation network shows that the authors of relevant papers in the field of biogas upgrading do not form a large cooperation network but instead form many small cooperation networks. As shown in Fig. 4, the graph shows the names of 105 authors who have published three or more papers. The 8 authors who have published the most papers are shown in Table 2, and the number of papers published by these authors is more than 10 (including 10). The three authors with the highest number of publications are Munoz Raul, Angelidaki Irini, and Yongjun Zhao, who have published 34, 32, and 31 papers, respectively. These three authors form the three largest collaborative networks. The largest network of authors, led by Munoz Raul, includes 22 authors; Zhao Yongjun's network includes 18 authors; and Angelidaki Irini's network includes 14 authors. An analysis of these three collaborative networks shows that Zhao Yongjun's collaborative network had the closest cooperation between authors and was the most important one. Its importance is mainly manifested by the fact that 4 of the top 8 authors by the number of papers published are in the military cooperation network. These authors are Zhao Yongjun (31 papers, third in the number of papers), Sun Shiqing (24 papers, fourth in the number of papers), Liu Juan (13 papers, sixth in the number of papers), and Zhao Chunzhi (11 papers, seventh in the number of papers).

Burst detection finds nodes of sudden events or research hotspots (burst nodes) that have attracted academic attention over a period, and it helps researchers track dynamic changes and trends in the academic field. The more burst nodes in an author's collaborative network, the more it indicates that the author has led the field over a certain period. By studying these collaborative networks, we can get an initial idea of the trends in the field. The results of burst detection showed that the Munoz Raul-dominated collaboration network contained 22 authors and was the largest author collaboration network, but no burst points were detected. In addition, none of the five authors with the largest number of published papers were detected as burst nodes. Six nodes were detected in Zhang's cooperative network, 4 of which were burst nodes. Zhao Yongjun's cooperative network detected 3 burst nodes. This finding shows that the researchers in Zhang Xiangping and Zhao Yongjun's network are highly innovative and have made significant contributions to the development of the field of biogas upgrading.

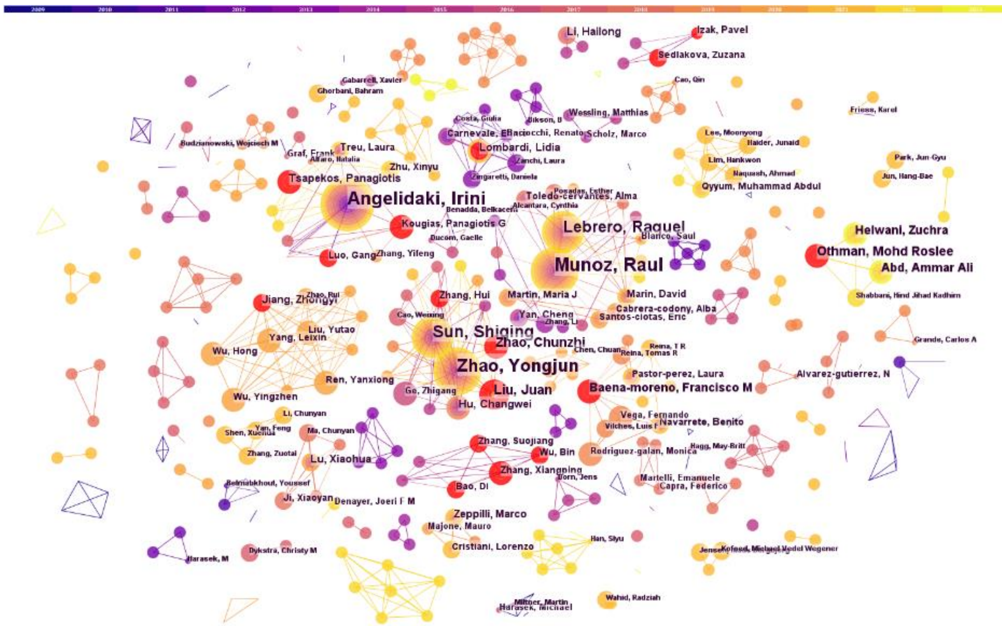

Fig. 4. Author cooperation network

Table 2. Author of 10 or More Published Papers

Ranking	Number of papers	Author
1	34	Munoz, Raul
2	32	Angelidaki, Irini
3	31	Zhao, Yongjun
4	24	Sun, Shiqing
5	22	Lebrero, Raquel
6	13	Liu, Juan
7	11	Zhao, Chunzhi
8	10	Othman, Mohd Roslee

Analysis of National And Institutional Cooperation Networks

In this paper, 430 organizations from 72 countries or regions were screened. As shown in Fig. 5, the labeled nodes in the outer circle represent countries or institutions with 30 or more articles, and the labeled nodes in the inner circle are the institutions involved in our analysis. Institutions belonging to China published the highest number of papers (240), followed by institutions belonging to Italy with 125 papers. Table 3 shows the top 12 countries and institutions in terms of the number of published papers. According to the CiteSpace analysis results, nodes with betweenness centrality ≥ 0.1 are turning points, and this type of node is an important "bridge" that connects networks without direct collaboration. Turning points are marked with purple outer circles in the collaborative network. As shown in Fig. 5 and Table 3, the top 5 countries or institutions in terms of the number of published papers are all turning points and have made significant contributions to the technology exchange in the field of biogas upgrading.

An analysis of the connections between the nodes in Fig. 5 shows that the connections between countries or institutions with more than 20 papers are not close, except for Denmark and the Technical University of Denmark, and China and the Chinese Academy of Sciences, with no direct cooperation between Austria and the Czech Republic. A total of 7 institutions or regions in China have more than 10 or more published papers, namely, Chinese Academy of Sciences with 33 papers, Jiaxing University with 32 papers, University of Chinese Academy of Sciences with 16 papers, Tianjin University with 13 papers, Nanjing University of Technology with 11 papers, Shanghai University of Applied Sciences with 11 papers and Taiwan with 11 papers, the rest of the institutions or regions having fewer than 10 published papers. These seven institutions or regions accounted for 52.92% of the total number of publications in China. The Chinese Academy of Sciences, Shanghai Institute of Technology, and Jiaxing University cooperated extensively, but no cooperation occurred between the institutions affiliated with Taiwan Province of China and the other six institutions. This finding shows that the cooperation among the institutions that publish papers related to the field of biogas upgrading needs to be strengthened.

Fig. 5. National and Institutional cooperation network

Table 3. Top 12 Countries and Institutions in Terms of Number of Papers Published

Ranking	Counted	Betweenness centrality	Country/institution
1	240	0.64	China
2	125	0.42	Italy
3	123	0.28	Spain
4	70	0.18	USA
5	67	0.24	Germany
6	62	0.09	Denmark
7	50	0.16	France
8	49	0.08	England
9	43	0.09	South Korea
10	40	0.04	Technical University of Denmark
11	39	0.04	University of Valladolid
12	33	0.12	Australia

Cited-Reference Cluster Analysis

Cited-reference were clustered using keywords as the source of clustering labels, and a total of 15 clusters were analyzed, and 8 of the main clusters were analyzed. According to CiteSpace, the cluster "accelerated carbonation" contains the most citations. The cluster covers the period 2008 to 2017, and one of the keywords is "Aspen Plus," a large-scale general-purpose process simulation system for production plants design, steady-state simulation, and optimization. The "accelerated carbonation" cluster includes some literature on the feasibility of simulating biogas upgrading systems using Aspen Plus. In addition, "greenness" and "chlorella" were mentioned more frequently in the cluster. The two most recent clusters to emerge are the "life cycle assessment" (formed in 2014) and the "pressure swing adsorption" (formed in 2013) clusters. The "life cycle assessment" cluster mainly covers the economics of biogas upgrading technologies, with the main keywords being "biomethanation," "microalgae," "chemisorption," and "economic evaluation". The "pressure swing adsorption" cluster focuses on increasing the methane content in biogas through carbon dioxide adsorption, and the main keywords are "methane enrichment," "heat adsorption" and "carbon dioxide capture".

The "biomethanation" cluster has lasted for 13 years and is the longest lasting cluster. "Hydrophilic methanogens", "power-to-gas" and "gas-liquid mass transfer" are the three key words of the cluster, whose main research focus is the conversion of carbon dioxide and hydrogen from biogas to methane using biochemical reactions of hydrophilic methanogens. The "siloxanes" cluster focuses on the removal of volatile methyl siloxanes, sulfur compounds, and octamethylcyclotetrasiloxane from biogas. The "microalgae" cluster was started in 2011, with nutrient removal and algal-bacterial photobioreactors as the keywords for the cluster, which mainly utilizes biogas to cultivate microalgae for carbon dioxide removal. The "fuel cells" cluster was formed in 2004 and is the earliest cluster formed in this study, with the keywords "activated carbon fiber cloth" and "fuel upgrade". The "Acidithiobacillus ferrooxidans" cluster has the shortest duration (6 years) in this study, and its keywords were "pilot scale," "adsorption selectivity," "system integration," and "carbon dioxide removal". In summary, under the trend of carbon emission reduction, the "life cycle assessment" and "biomethanation" clusters have become a hot research topic in the field of biogas upgrading.

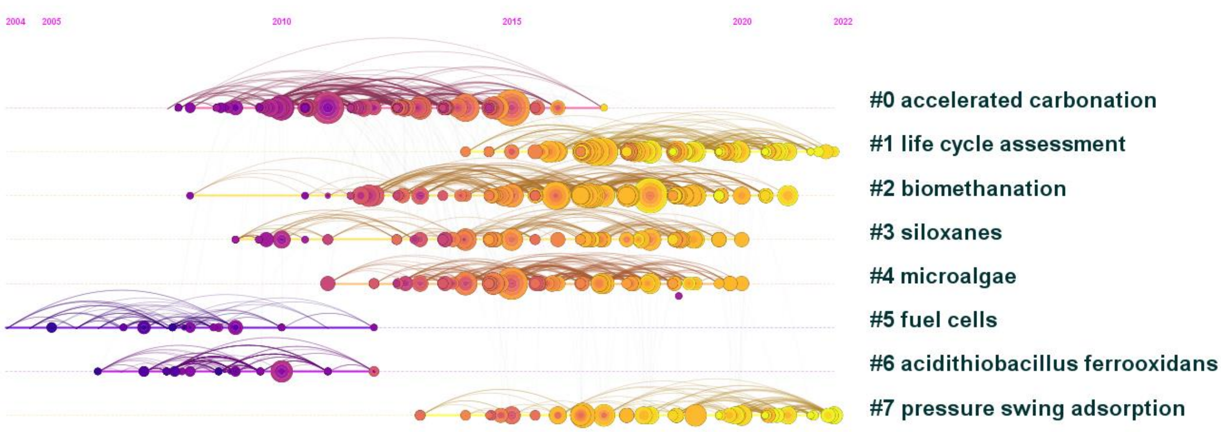

Fig. 6. Clustering results of cited-reference with keywords as the source of clustering labels

OUTLOOK

This research has found that the current frontiers in the field of biogas upgrading are life cycle assessment and biomethanation. Life Cycle Assessment (LCA) is dedicated to studying the energy consumption and greenhouse gas emissions of each step of the biogas upgrading process in order to target the reduction of energy consumption and greenhouse gas emissions, while biomethanization is dedicated to achieving the upgrading process in a simpler, less costly, and less energy-intensive way (by using the excess electricity generated by wind and photovoltaic power generation as an energy source). The goal of both types of research is to make the biogas upgrading process cleaner. Therefore, in the future, researchers can focus their attention on the combination of LCA and biomethanation, which is very important for sustainable development in the field of biogas upgrading. And researchers can use LCA to improve biogas upgrading based on physical/chemical methods, which may reduce greenhouse gas emissions from the factory and bring additional benefits to the factory.

CONCLUSIONS

1. On the basis of CiteSpace's visual analysis of 1084 papers published in the field of biogas upgrading between 2009 and 2023, combined with the publication trend of biogas upgrading-related papers, the progress of biogas upgrading research is divided into three phases: 2009 to 2011 (Phase I) has relatively few research on biogas upgrading, but the research scope in this phase is broad, including the fields of biomethanation, variable pressure adsorption, and so on, laying a theoretical foundation for later biogas upgrading-related research; 2012 to 2015 (Phase II), during which the physical/chemical biogas upgrading technology matures and H_2S removal reaches 100%; the papers published during 2016 to 2022 (Phase III) accounted for 73. 25% of the total research statistical literature, and the research on biomethane upgrading techniques has matured and the number of published papers is rapidly increasing.
2. The trend of the number of noun terms added over time is not consistent with the number of published papers. The term clustering results indicate that the most used terms are concentrated in 2009 to 2011 (Phase I). The further indicates that the 2009 to 2011 study established the theoretical foundation for biogas upgrading.
3. The co-occurrence network analysis results show that the collaborative network among researchers in the field of biogas upgrading is small, and the collaboration among the countries and research institutions involved is not close. Under the trend of carbon emission reduction, "life cycle assessment" and "biomethanation" clusters have become the current research frontiers in the field of biogas upgrading.

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