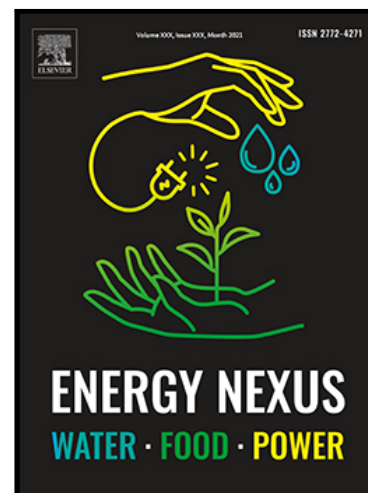


Journal Pre-proof

Research trends and perspectives on hydrothermal gasification in producing biofuels

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PII: S2772-4271(23)00029-3
DOI: <https://doi.org/10.1016/j.nexus.2023.100199>
Reference: NEXUS 100199

To appear in: *Energy Nexus*

Received date: 30 January 2023
Revised date: 7 April 2023
Accepted date: 25 April 2023

Please cite this article as: Rubens Costa Nogueira , Francisco Simão Neto , Paulo Gonçalves de Sousa Junior , Roberta Bussons Rodrigues Valério , Juliana de França Serpa , Ana Michele da Silva Lima , Maria Cristiane Martins de Souza , Rita Karolinny Chaves de Lima , Ada Amélia Sanders Lopes , Artemis Pessoa Guimarães , Rafael Leandro Fernandes Melo , Maria Alexandra de Sousa Rios , José Cleiton Sousa dos Santos , Research trends and perspectives on hydrothermal gasification in producing biofuels, *Energy Nexus* (2023), doi: <https://doi.org/10.1016/j.nexus.2023.100199>

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Highlights

- The bibliometric method is used to evaluate hydrothermal gasification.
- There is a growing interest in hydrothermal gasification research.
- VOSviewer and CiteSpace are used to analyze 331 articles.
- Comprehensive guidelines are given for hydrothermal gasification research.
- Related technologies such as hydrothermal liquefaction and pyrolysis are evaluated.

Journal Pre-proof

Research trends and perspectives on hydrothermal gasification in producing biofuels

Rubens Costa Nogueira¹, Francisco Simão Neto², Paulo Gonçalves de Sousa Junior³, Roberta Bussons Rodrigues Valério³, Juliana de França Serpa¹, Ana Michele da Silva Lima⁴, Maria Cristiane Martins de Souza¹, Rita Karolinny Chaves de Lima¹, Ada Amélia Sanders Lopes¹, Artemis Pessoa Guimarães¹, Rafael Leandro Fernandes Melo⁵, Maria Alexsandra de Sousa Rios⁶ and José Cleiton Sousa dos Santos^{1,2*}

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Abstract

Several researchers around the world have been investigating the use of supercritical fluids incorporated into the process to create a method known as hydrothermal gasification (HTG). Thus, the present study aims to evaluate what has been produced regarding biofuels produced from HTG. A bibliometric analysis of the Web of Science (WoS) database was performed for articles published between 2006 and 2022. In the first analysis, 331 articles were identified, and refined analyses for 320 and 311 publications of the Web of Science Core Collection database (2006- 2022) were performed using VOSviewer, CiteSpace, and Microsoft Excel. The year 2022 had the highest number of articles, with 54 publications, followed by 2021 and 2015 with 45 and 31 publications, respectively. The three journals with the most significant impact were Bioresource Technology, Algal Research: Biomass Biofuels And Bioproducts, and Biomass & Bioenergy, with TPs of 26, 18, and 17, respectively. China, the USA, and Canada represented 11.48%, 10.89%, and 7.52% of the total publications. Investments in research on supercritical fluids and carbonization should be more significant in countries that publish more. Much research still needs to be done for the advancement of the area, as evidenced by the low number of publications. Future studies should focus on related technologies such as liquefaction and pyrolysis

Keywords: supercritical fluids, hydrothermal gasification, Web of Science, citeSpace, VOSviewer, biofuel.

Abbreviation full term

HTG – Hydrothermal gasification

WoS – Web of Science

PRISMA – Preferred Reporting Items for Systematic Reviews and Meta-Analyses

TPs – Total Publications

JC – Journal Citation

TC – Total Citations

ACY – Average Citations by Year

IF – Impact Factor

APPY – Average Publications Per Year

CC – Citation by Country

RIO+20 – United Nations Conference on Sustainable Development

MPa – Mega Pascal

1 INTRODUCTION

Since ancient times, humans have created and modified their environment to improve their safety and quality of life [1]. For a long time, simple fuels were used to generate fire to meet their energy needs; however, the discovery of fossil fuels, processes, and scientific advances led to the Industrial Revolution and new ways to harness this energy [2-4].

Among the characteristics of fossil fuels, their high energy density and versatility attract considerable attention worldwide. However, these resources are exhaustible and have associated environmental problems [5-7]. The increase in the concentration of carbon dioxide gas in the atmosphere due to combustion, one of the main reasons for the increase in global temperature through the greenhouse effect, is one of the main problems associated with it [8-11].

In this sense, new ways are needed to explore more renewable energy, although the investment is more significant and financially less attractive [8]. To produce them, a suitable feedstock must be selected based on intrinsic factors, which are the situation of the environment in which it is intended to be produced [9]. When used to produce some form of energy, this feedstock is called biomass [10-13].

There are two basic types of energy—the first is traditional biomass, which differs from modern biomass (the second type of energy) in that it uses primitive, unsustainable methods standard in some isolated communities in Africa and Asia, and regions Latin America [14]. Modern biomass is considered sustainable because it is one of the products used to produce electricity or charcoal [15]. In addition, the proportion of biomass used in different regions of the world, whether traditional or modern, can vary greatly depending on their level of development—from 2% in OECD countries to 60% in some areas of Africa

[16].

The possibilities of biomasses are many, among which plant biomass stands out, which is an indirect way of producing energy from the sun by converting chemical energy through photosynthesis [17]. Plant biomass can be classified into two classes based on its constitution: lignocellulosic materials (e.g., lignin, cellulose, and hemicellulose) or low-stability organic components (e.g., lipids, proteins, simple polysaccharides) [18-20]. Additionally, noteworthy are the groups resulting from the use of organic waste, which, although not strictly natural, have the potential to produce biofuels [21].

Biofuels have been a promising alternative source to replace fossil fuels. Their first generation produced these from various consumable crops such as corn, ethanol, and other vegetable oils [12-15]. After field development, biofuels were produced from lignocellulosic and other non-edible oils and crops [19,20]. They were making biofuels even more favorable to environmental and social development. Although some countries still need to adapt the use of second-generation biofuels better [21,22].

In Brazil, sugarcane for ethanol production has been a reality for several decades, despite the obstacles of working with a food-based raw material, first-generation biofuel production. In addition to this problem, due to the high energy potential of sugarcane, it is common for mills to generate excess electricity, which, although sold to regional electricity companies, is a condition that must be better controlled [22]. A parallel solution is to control the energy cycle so that it does not act wastefully in any way [23].

The application of the gasification process can make possible the co-production of several products, including electricity [24, 25], liquid fuels [26], chemicals [27], and the removal or minimization of potentially polluting products [28].

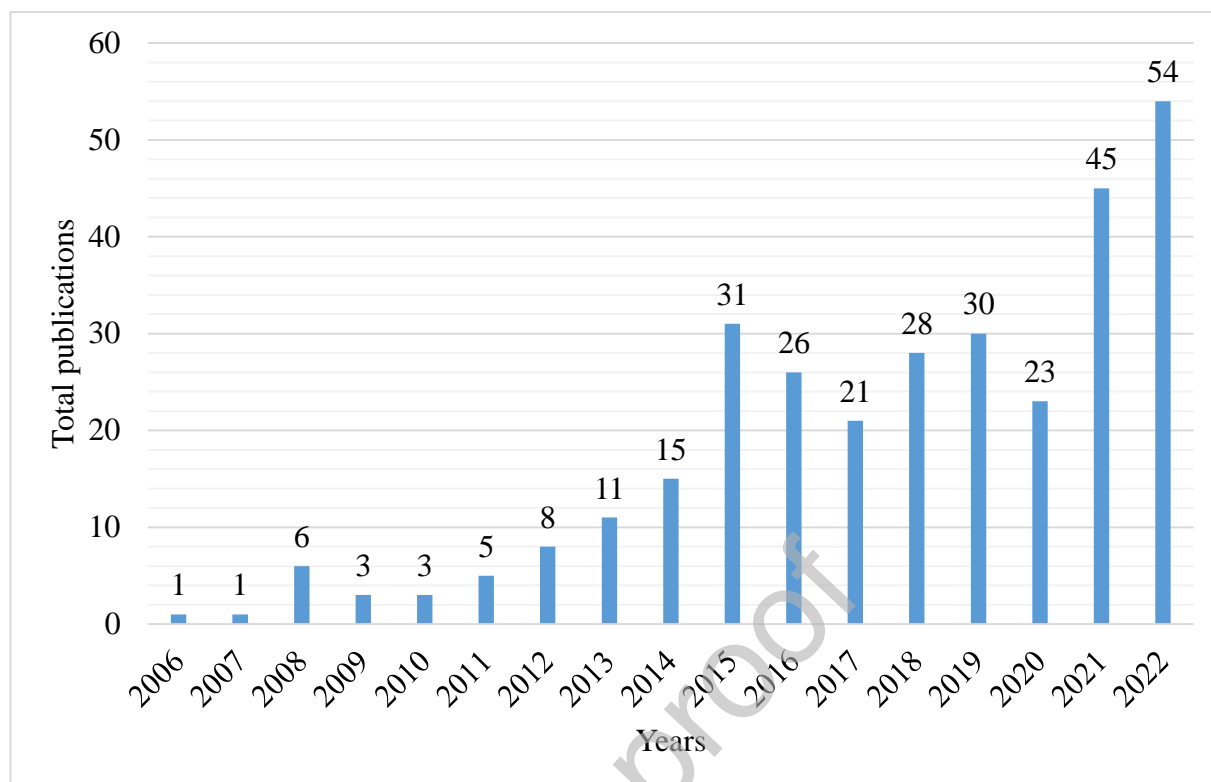
Gasification aims to produce synthetic gas for use in high-efficiency gas turbines. Many authors emphasize that since research is still early, exploring gasification may be advantageous, especially for small producers in remote areas where there is still a need to produce energy as heat and electricity [16,29-31]. Several techniques have been implemented to improve gasification yields, thus drawing attention to the incorporation of gasification into supercritical fluid systems and transforming it into a new strategy for extractive processes called hydrothermal gasification or high-pressure gasification (HTG) [32-36].

The present study aims to evaluate, by means of bibliometric analysis of the Web of Science (WoS) database, from 2006 to 2022, the development processes and prospects for future biofuel production research by hydrothermal gasification. In doing so, we aim to understand how HTG influences literary production regarding biofuels. We also intend to answer the following questions RQs:

- RQ1: How has scientific production been in research on HTG-produced biofuels from biomass and organic waste?
- RQ2: What are the main research hotspots (keywords) used in this research?
- RQ3: Who are the seminal founders (historical emergence of different perspectives) in HTG biofuel production research?
- RQ4: What are the main subfields and emerging themes in research on HTG biofuel production?

In an attempt to understand the process of approaching the issue, the studies produced over the years were quantified (Figure 1).

Figure 1 – Distribution of publications on biofuel production from hydrothermal gasification between 2006 and 2022.



There has been an increase in research since 2012 (Figure 1). This may be because RIO+20 promoted more meaningful investments in the green economy, intending to contribute to poverty eradication and opportunities for sustainable development and economic growth. In this way, RIO+20 has expanded pathways that allow for real social inclusion, improve the well-being of communities, create new jobs, and contribute to preserving the Earth's ecosystems. [37, 38].

The bibliometric analysis allows the reader to understand the overview of the studies carried out worldwide, the institutions promoting research, and the authors focusing on the content. It also allows the analysis of the total number of publications, citations, and impact factors, among other parameters [39]. To the authors' knowledge, this is the first research in the literature on the bibliometric analysis of biofuels through hydrothermal gasification, providing the scientific community with a systematic mapping analysis on the subject.

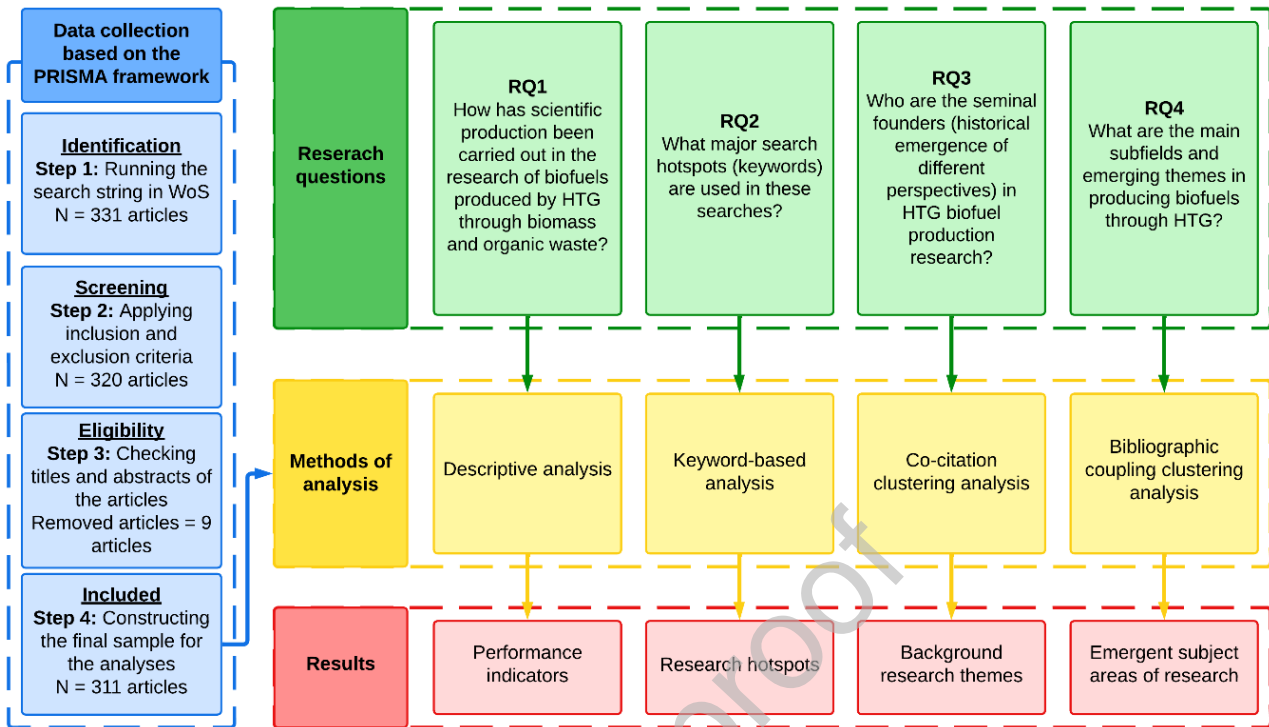
2 METHODOLOGY

A systematic analysis of scientific mapping was adopted according to Catumba et al. [40] and Sales et al. [41]. The survey obtained according to the questioning of the RQs presented studies from different parts of the world, institutions, and authors, based on citations, publications, impact factors, and other parameters. In addition, to provide an image of the trends and perspectives of how the topic of biofuels through hydrothermal gasification evolves in the scientific community, a grouping by clusters is shown, according to Ranjbari et al. [42].

2.1 Data source

In this study, data for bibliometric analysis were obtained from the Web of Science (WoS) database because many authors cite it as an essential research database in various fields of knowledge. [40, 41]. The search criteria are shown in Figure 2.

Figure 2 – Structure representing the search and analysis criteria.



The keywords “hydrothermal,” “gasification,” and “biofuel” were searched on November 10, 2022. The initial search retrieved articles published between 2006 and 2022. Articles published in English were screened according to the Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) statement [42-44]. Papers that did not include all the information on the platform, were presented at conferences, and were still in preprint format were excluded.

2.2 Data analysis

The free software CiteSpace (version 6.1. R4 Basic) was used to create the bibliometric maps. CiteSpace is software that supports the visualization and construction of bibliometric maps. The data obtained from WoS and compiled in CiteSpace allowed the construction of maps of journals, countries, institutions, authors, and keywords based on the correlation data. In addition, Microsoft Excel (Microsoft Office 365[®]) spreadsheets were used for data analysis and plotting.

3. RESULTS AND DISCUSSION

To respond more clearly to the RQs of the study, the results obtained are presented in the sections: Distribution of Scientific Journals; Distribution by Country and Institution; Quantitative analysis of cited references [Ranjbari et al. 2022].

3.1 Distribution of Scientific Journals

Through the 311 published papers [32-36, 45-350], 38 different journals were found (an average of 8.18 articles per journal and 19.44 articles per year). From these numbers, one can see a scientific interest in HTG-produced biodiesel. For a more detailed analysis, please see Table 1.

Table 1 – Ranking of the 12 journals that have published the most on biofuel production by HTG between 2006 and 2022.

N	JOURNAL TITLE	TPs	%	JC	TC	ACY	COUNTRY	IF	APPY
1	Bioresource Technology	26	8,36	59,35	1543	96,438	England	11.889	1,625
2	Algal Research: Biomass Biofuels And Bioproducts	18	5,79	42,00	756	47,250	Netherlands	5,267	1,125
3	Biomass & Bioenergy	17	5,47	83,18	1414	88,375	England	5,610	1,062
4	Renewable & Sustainable Energy Reviews	17	5,47	151,59	2577	161,063	England	14.982	1,062
5	Biofuels Bioproducts & Biorefining	14	4,50	72,07	1009	63,063	England	4.102	0,875
6	Fuel	14	4,50	74,50	1043	65,188	England	6,609	0,875
7	Energies	14	4,50	102,00	1428	89,250	Switzerland	3,004	0,875
8	Journal Of Supercritical	12	3,86	55,00	660	41,250	Netherlands	4.577	0,750

Fluids									
9	Energy & Fuels	11	3,54	53,00	583	36,438	USA	3.605	0,687
10	Science Of The Total Environment	8	2,57	150,50	1204	75,250	Netherlands	7.963	0,500
11	Energy	7	2,25	51,57	361	22,563	England	7,147	0,437
12	Fuel Processing Technology	6	1,93	64,83	389	24,313	Netherlands	7,033	0,375

TPs = Total Publications; % = Proportion; JC = Journal Citation; TC = Total Citations; ACY = Average Citations by Year; IF = Impact Factor in 2020; APPY= Average Publications Per Year.

Through the quantitative analysis of the publications of these 12 journals, we observed that 164 of the 311 articles were analyzed (approximately 53% of the total articles). The journal *Bioresource Technology* has the most significant number of articles published on the subject (28 publications), representing slightly over 8% of the total publications analyzed and reaching 1,543 citations.

The journal has the second-highest impact factor (surpassed only by *Renewable & Sustainable Energy Reviews*, which ranks 4th in total publications). *Science of the Total Environment*, *Energy*, and *Fuel Processing Technology* ranked 10th, 11th, and 12th, respectively, and have higher impact factors than the other journals not mentioned. Characteristics such as these underscore the difficulty of producing articles in higher-impact journals, even if the results are promising.

3.2 Distribution by Country and Institution

Understanding the importance of the number of countries for a bibliometric analysis relates to the relevance of the topic for that location; thus, it can serve as an indication of possible trends in the number of production locations [39]. Thus, the 12 countries with the most publications were identified (Table 2).

Table 2 – The 12 most productive countries regarding HTG-produced biodiesel between 2006 and 2022.

N	COUNTRY	TPs	%	TC	CC	ACY	H-Index
1	CHINA	58	11,485	6.580	113,45	658,00	28
2	USA	55	10,891	3.269	59,44	326,90	32
3	CANADA	38	7,525	1.652	43,47	165,20	20
4	INDIA	29	5,743	981	33,83	98,10	15
5	GERMANY	22	4,356	737	33,50	73,70	11
6	ENGLAND	22	4,356	1.848	84,00	184,80	13
7	ITALY	18	3,564	828	46,00	82,80	11
8	MALAYSIA	18	3,564	731	40,61	73,10	11
9	BRAZIL	14	2,772	382	27,29	38,20	9
10	JAPAN	13	2,574	393	30,23	39,30	7
11	AUSTRALIA	12	2,376	484	40,33	48,40	8
12	FRANCE	12	2,376	546	45,50	54,60	8

TPs = Total Publications; % = Proportion; CC = Citation by Country; TC = Total Citations; ACY = Average Citations by Year.

The 12 most productive countries account for 61.60% of the total publications of the 66 countries analyzed. China, with 61 publications, concentrates slightly over 11.7% of the total, followed by the United States (60 publications and 11.5%). H-index is relevant and represents the qualitative evaluation of researchers, analyzing the impact of each researcher [351]. In this work, the impact factor of American researchers exceeds by a margin of 4 points, even with fewer researchers. Figure 3 illustrates the demographics of published papers through a map.

Figure 3 – Geocoding of the organizations that publish the 311 articles analyzed.

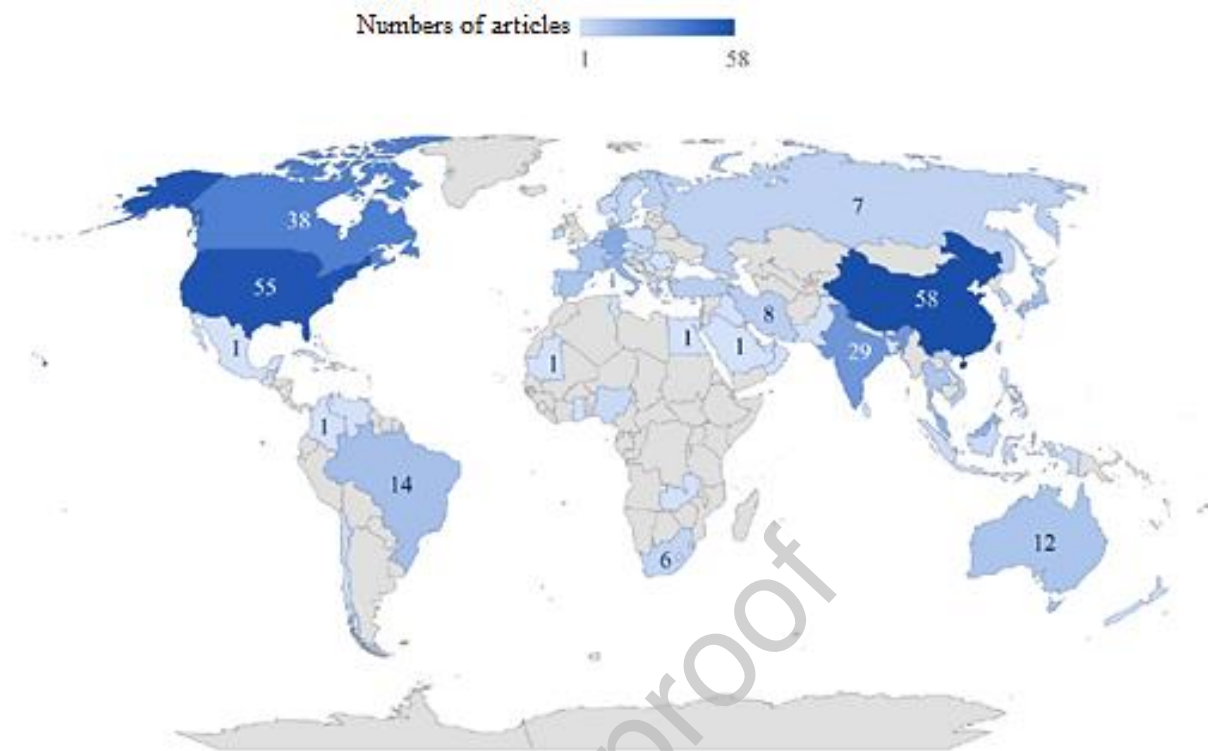
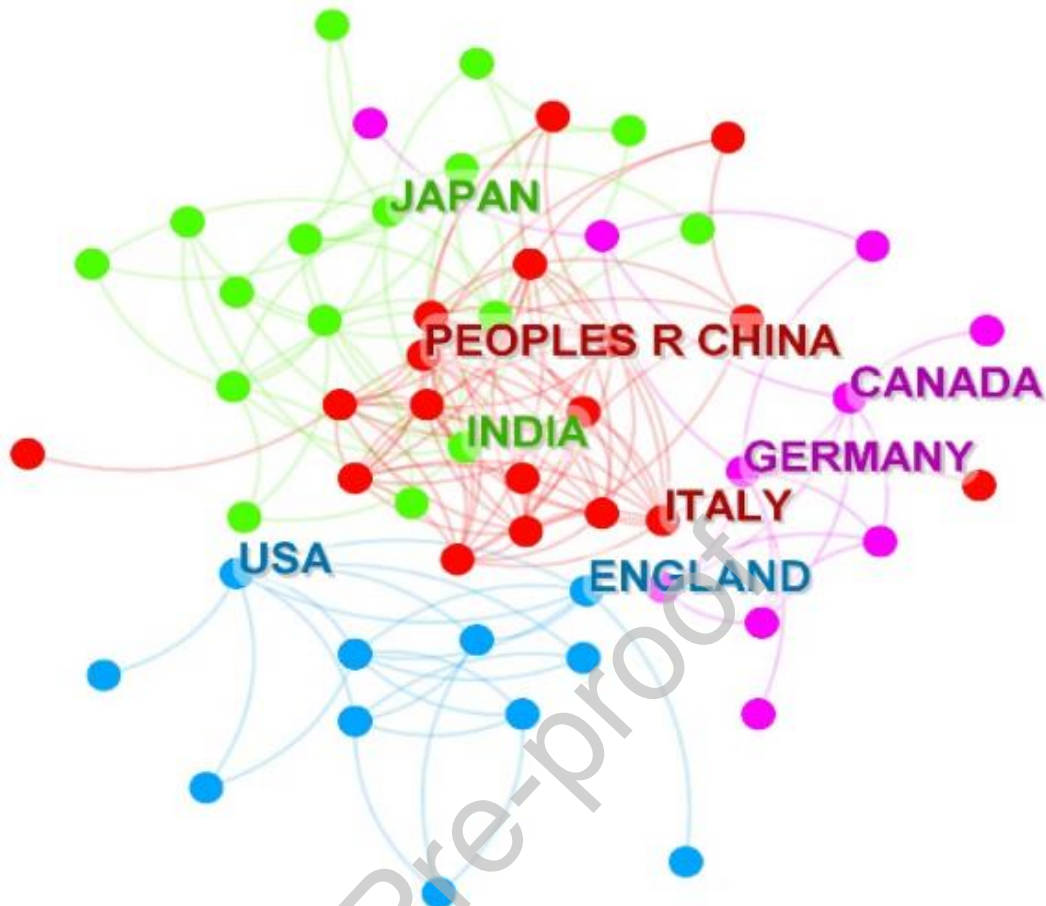


Figure 3 shows that research is highly developed in several countries (e.g., China, the United States, Canada, and India). Figure 4 shows a network map of the collaborative links between the scientific groups analyzed.

Figure 4 – National collaboration network of articles regarding HTG-produced biofuels published between 2006 and 2022 on WoS. The thickness of a line connecting two countries indicates the frequency of co-authorships (thicker lines mean more published articles), and the color clusters indicate groups of countries with a high level of collaboration.

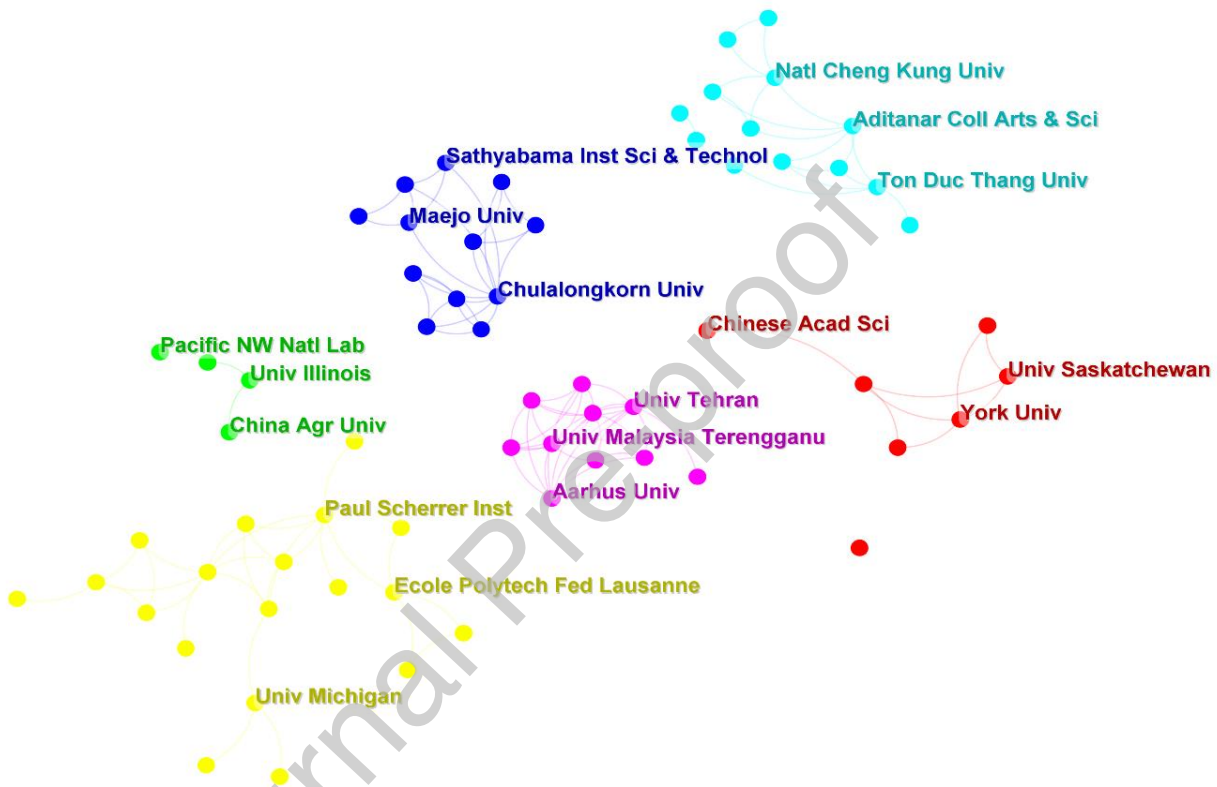


China is the most collaborative country, followed by India (Figure 4). Despite the large amount of research carried out by the United States, there is little interaction between North American researchers and non-English speaking researchers.

The analysis shows that 568 institutions are associated with 1,352 authors from 66 countries who have written 331 published papers. Although many institutions are interested in the topic, 394 institutions (approximately 64.4%) have only one publication in the field, which is characterized by high institutional dispersion; for example, only 45 institutions have more than four published papers. In particular, the University of Saskatchewan in Canada stands out as the institution with the most significant number of published papers on the topic (N = 17), followed by the U.S. Department of Energy (N = 16). To understand how these

institutions interact, a map of the collaborative network between the institutions was created (Figure 5).

Figure 5 – Map of collaboration among institutions. The thickness of a line connecting two organizations indicates the frequency of co-authorships (thicker lines mean more joint published articles), and the color clusters highlight groups of institutions with a high level of collaboration.



We used the citations of the papers to observe how they interacted. A total of 67 institutions (11.82%) were the most cited. It can be observed that even the institutions with the most significant number of published articles have few interactions, probably because their publications are primarily in low-impact journals.

Considering an average density of approximately 4.5 authors per article and 2.4 authors per institution, these data confirm a high dispersion of researchers. If only three authors with at least 10 publications are selected, the result is only three. Among them, Sonil

Nanda stands out with the most significant publications (18 papers and 639 citations in other articles). To better understand how the authors are articulated, Figure 6 summarizes a map of collaboration among the authors.

Figure 6 – Map of collaboration among authors. The thickness of a line connecting two authors indicates the frequency of co-authorships (thicker lines mean more published articles), and the color clusters illustrate groups of authors with a high level of collaboration.



According to the map, Nanda Sonil has the largest network of connections and the highest number of articles. Observing the trend of other research in the area, one would expect that small groups in this type of map would be obtained from small groups of researchers from the same country; however, the trend was different. The prominent authors interact with each other and create a system that includes all the other authors.

3.3 Quantitative analysis of cited references

Among the factors that most contribute to disseminating a scientific article, we can highlight the selected articles, their references, authors, number of citations, and the most relevant journals [352]. Table 3 shows the 12 most cited publications in the analyzed period.

Table 3 – Main publications on biofuel production by HTG between 2006 and 2022.

NO. OF CITATIONS	YEAR	1st AUTHOR	TÍTULO	JOURNAL	COUNTRY
1,408	2008	Petterson	Thermochemical biofuel production in hydrothermal media: A review of sub- and supercritical water technologies	Energy & Environmental Science	USA
540	2015	Elliott	Hydrothermal liquefaction of biomass: Developments from batch to continuous process	Bioresource Technology	USA
527	2010	Brown	Hydrothermal Liquefaction and Gasification of <i>Nannochloropsis</i> sp.	Energy & Fuels	USA
444	2013	Barreiro	Hydrothermal liquefaction (HTL) of microalgae for biofuel production: State of the art review and future prospects	Biomass & Bioenergy	USA
370	2014	Tekin	A review of hydrothermal biomass processing	Renewable & Sustainable Energy Reviews	Turkey

365	2008	Kruse	Supercritical water gasification	Biofuels Bioproducts & Biorefining	Germany
351	2011	Meyer	Technical, Economical, and Climate-Related Aspects of Biochar Production Technologies: A Literature Review	Environmental Science & Technology	Switzerland
336	2012	Biller	Nutrient recycling of aqueous phase for microalgae cultivation from the hydrothermal liquefaction process	Algal Research-Biomass Biofuels and Bioproducts	Denmark
331	2015	Chen	Thermochemical conversion of microalgal biomass into biofuels: A review	Bioresource Technology	China
321	2013	Elliott	Process development for hydrothermal liquefaction of algae feedstocks in a continuous-flow reactor	Algal Research-Biomass Biofuels and Bioproducts	USA
292	2008	Elliott	Catalytic hydrothermal gasification of biomass	Biofuels Bioproducts & Biorefining	USA
190	2015	Kruse	Water - A magic solvent for biomass conversion	Journal of Supercritical Fluids	Germany

Table 3 shows many highly cited publications by North American authors and

highlights Elliott as the first author in three of them and coauthor in one, although he is not among the authors with the most publications. North American authors maintain relationships to publish more. Compared with Figure 6, the authors with the highest number of publications in the field, except Kruse and Barreiro, are not among the authors with the highest number of citations. Energy & Environmental Science's impact factor influenced the dissemination of the research (39,714 points), becoming the scientific journal with the highest score among those recorded in the present bibliometric analysis.

4 HOT RESEARCH TOPICS

In order to obtain a mapping of trends and future perspectives, an analysis of keywords and research areas is adopted, which allows the evaluation of the formed clusters.

4.1 Quantitative analysis of frequent Keywords

In addition to the points already mentioned (i.e., authors, affiliations, and impact factors), keywords are also important because, in addition to being a search tool in bibliographic queries, they present the main concepts that are related to the topic [353]. Table 4 shows the ranking of the top 12 keywords used in this study.

Table 4 – Frequency analysis of the 12 most used keywords in research on biofuel production by HTG between 2006 and 2022.

N	KEYWORDS	FREQUENCY	%
1	GASIFICATION	115	3.960
2	BIOMASS	90	3.099
3	HYDROTHERMAL LIQUEFACTION	73	2.514
4	BIO-OIL	69	2.376
5	HYDROGEN-PRODUCTION	62	2.135
6	SUPERCRITICAL WATER GASIFICATION	52	1.791
7	BIOFUEL PRODUCTION	52	1.791

8	CONVERSION	46	1.584
9	SUPERCRITICAL WATER	43	1.481
10	LIGNOCELLULOSIC BIOMASS	43	1.481
11	MICROALGAE	39	1.343
12	SEWAGE-SLUDGE	37	1.274

A total of 2,904 keywords were obtained from 881 terms. Although different, many keywords had the same meaning, such as “supercritical water gasification” (6th keyword and 52 hits) and “supercritical water” (9th keyword and 43 hits), and “pyrolysis fast” (13th keyword and 35 hits) and “pyrolysis” (14th keyword and 31 hits). Although they have the same meaning, they were treated as independent terms.

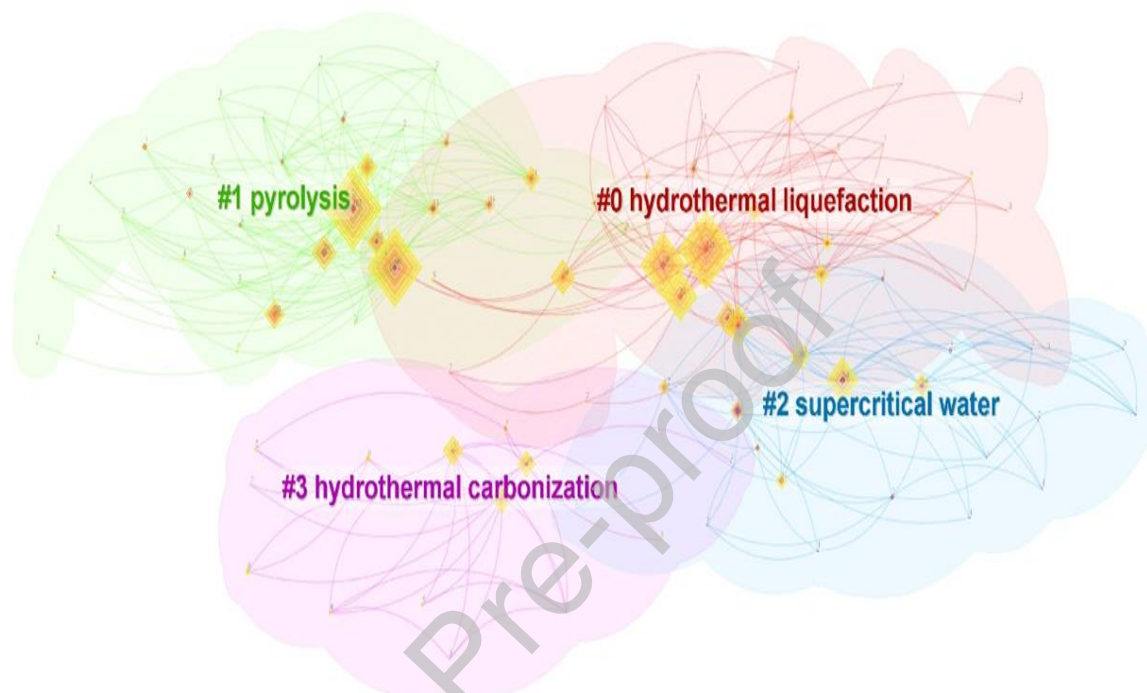
Two of the three keywords selected in the search tool were ranked in the first three positions, especially in the 3rd position, which considers fuel liquefaction. The term “biofuel” appears only in the 7th position with 52 hits. The 34th position is occupied by the term “biofuel” (N = 15), the 40th by the keyword “biofuels” (N = 14), the 114th by the term “solid biofuel production” (N = 4), the 194th by the term “biofuel production” (N = 2), and four other matches with only one citation.

4.2. Research areas

In addition to plotting graphs and creating maps, CiteSpace can be used to analyze and organize research data and understand possible trends in the field, as the software allows visualization of what is being developed [354]. Through CiteSpace, we can identify more projectable data based on various factors (e.g., keywords). Thus, CiteSpace is one of the most essential tools for developing viable methodologies for biofuel production using HTG

processes [354, 355]. Figure 7 shows the top five cocitation sets among the articles related to the topic of the study.

Figure 7 – Map of the relationship between citations on biofuel production through HTG between 2006 and 2022.



In Figure 7, four major groups can be observed—“hydrothermal liquefaction”, “pyrolysis”, “supercritical water”, and “hydrothermal carbonization”. We have the main keywords within these groups and how they are interconnected and related. Although the “pyrolysis” group has a cluster with a higher number of hits (e.g., “gasification” and “biomass”), the techniques are less numerous than those addressed within “hydrothermal liquefaction”.

4.2.1 Research fields

Cluster #0 has “hydrothermal liquefaction” as its main keyword. Hydrothermal liquefaction involves the conversion of biomass at high pressure and high temperature using

water as a solvent and other cosolvents as necessary [147, 210, 220, 235, 356-357]. In hydrothermal liquefaction, there is a catalyst that, together with water and other solvents, allows secondary processes such as hydrolysis, fragmentation, and repolymerization of the biomass to take place, prioritizing the formation of liquefied products from products derived from different feedstocks [82, 292, 335, 358].

A predominance of lipidic feedstocks, refined or not, of animal and plant origin, derived from discards or residues from other processes [124, 187, 241, 291, 335]. According to the analysis of the articles published on this topic, algae are used to produce oils that are converted into biofuels with low cost and high yield. In the second group, algae from sewage treatment or other liquid waste are used to reduce pollutants and produce biofuels [130], [250-252, 289]. Most studies present the hydrothermal liquefaction process in a temperature range of 300°C to 450°C and pressure between 10 and 20 MPa.

Cluster #1 has the keyword “pyrolysis”. Pyrolysis is a biomass decomposition process at high temperatures in the presence of oxygen and other compounds in the gaseous, liquid, and solid phases [133, 163, 359-360]. The gas comprises mainly CO, CO₂, hydrogen, and hydrocarbons with low molar mass. The liquid phase of the product comes from gas condensation, known as pyrolytic liquid, while the solid phase is called biochar [113, 211, 308, 361-363]. Thus, Cluster #1 aims to discuss the decomposition process to obtain a new product and the analytical process to identify the composition of the phases, aided by other techniques to study the components [364]. Another completely different technique is microwave-induced pyrolysis to obtain a purer bio-oil. In the vast majority of works in this study, pyrolysis is just another process among the extraction, purification, and conversion of oil into biofuel [55, 316, 365-367].

Considering this type of occurrence in several keywords, Figure 7 illustrates how these keywords reflect and relate to each other.

4.2.2 Emerging trends

Looking at the emerging proposals, we have two clusters that show evident differentiation characteristics (number 2). Supercritical fluid screening techniques have proven to be increasingly applicable to current technologies. However, many authors note that while it has shown value in bench tests and for high value-added products, its use exponentially increases the price of the final product, making it unattractive for application in biofuel production in the current scenario [52, 74, 105, 368-370].

A supercritical fluid is an extractive process consisting of a system of liquids and gases confined in a given space and remaining in equilibrium [88, 248, 371]. By heating the system, the physical and chemical properties (density, rise, refractive index, thermal conductivity, etc.) of the liquid/gas interface begin to interact and converge toward a common point and become equally as well as characterize this situation called the critical point and create a single supercritical phase [46, 127, 131, 370, 372-373]. Therefore, a supercritical fluid is any substance that, under conditions of pressure and temperature above its critical criteria, remains stable [370-371, 374].

Cluster #3 has the keyword “hydrothermal carbonization”, a thermochemical conversion process at moderate temperatures. Hydrothermal carbonization uses lipid- and carbohydrate-rich materials or lignocellulosic feedstocks as biomass to produce a product known as solid hydrothermal carbon [240, 248, 324].

Many works use hydrothermal carbonization, especially microwave-assisted hydrothermal carbonization, as a possibility to replace pyrolysis techniques. It is considered

more promising because it can be applied to organic materials with high carbon content and feedstocks that do not necessarily undergo a previous drying process; in addition, the average pressures and temperatures are lower, and the products obtained, depending on the purpose, can be more promising than those of pyrolysis [58, 153, 189, 215, 310].

As described in Cluster #0, the plants analyzed use hydrothermal carbonization, primarily as a method of treating and reusing wastewater components, industrial wastes (such as sawdust and ash), and byproducts of other extractive processes, as well as feedstocks such as grains, vegetables, and animal remains [56, 89, 189, 227, 321]. Hydrothermal carbonization performs well with wet and residual biomass feedstocks as with supercritical fluids, but the process becomes too costly for power generation [118, 139].

4.3 Quantitative analysis of categories and areas of knowledge

The articles analyzed the production of biofuels using HTG are grouped into a total of 38 knowledge areas, according to the Web of Science database (Table 5).

Table 5 – Frequency analysis of the 12 categories with the most articles in research on biofuel production by HGT between 2006 and 2022.

CATEGORIES OF WEB OF SCIENCE	FREQUENCY
Energy Fuels	198
Engineering Chemical	97
Biotechnology Applied Microbiology	84
Environmental Sciences	53
Agricultural Engineering	45
Green Sustainable Science Technology	44
Engineering Environmental	35
Chemistry Multidisciplinary	26
Chemistry Physical	21
Thermodynamics	21

Mechanics	12
Chemistry Applied	8

The main category of articles is “energy fuels”, which is quite acceptable since one of the search terms was “biofuel” (Table 5). In addition, “chemical engineering” is observed in second place, followed by “biotechnology applied to microbiology”, which stands out for many researchers that used feedstocks for fuel production, organic waste, and water for reuse and wastewater.

5 OPPORTUNITIES AND PROSPECTS

The production cost of biofuels, and the use of first-generation biomass products, is still an associated economic challenge [375-377]. Using biomass as a generator of biofuels can lead to loss of the food supply chain, mainly when used in emerging countries [378, 379].

The transition from the production of first-generation biofuels to second-generation biofuels is necessary. However, the industry is a plural segment with its associated challenges. Thus, as this transition gains strength, other measures to reduce associated costs are employees [380–382].

The circular bioeconomy and the principles of biorefinery show that one of the transition opportunities is the use of biomass from macroalgae [383, 384]. There is a great advantage in the use of macroalgae over the production of biomass, in addition to being a component that does not interfere in the food chain, which is widely accessed in the production of species [385, 386]. Thus, the literature shows a growing perspective of implementing third-generation biorefineries capable of processing aquatic biomass through applications of hydrothermal system engineering platforms [387–389].

The use of first-generation biomass for the production of biofuels can also generate a logistical problem, such as, for example, excessive production of biomass that has not been transformed into biofuel [390, 391]. In addition to bioeconomic waste, this problem brings unnecessary food waste. Another opportunity that the clusters of this research showed as a perspective is the use of anaerobic digestion systems integrated with pyrolysis to treat food waste [392, 393]. The use of only anaerobic digestion of waste produces biogas with the presence of CO₂, hydrogen sulfide (H₂S), ammonia (NH₃), and volatile organic content (VOCs) that reduce the quality and calorific value [394–396]. Pyrolysis is one of the options that can be integrated into the digestion system that effectively improves the biogas product [397, 398].

The opportunities and prospects for the growth of biorefineries, with platforms for hydrothermal systems and biogas products from pyrolysis, raise studies on the subject of gasification, generating better results in energy use in the natural gas network, diesel engines, cogeneration (CHP) for the production of electric and thermal energy [399, 400].

6 RESEARCH LIMITATIONS

Systematic reviews present data and conclusions within a limited research universe, the amount of data found, formed clusters, and the research period. Although limited, the analysis was conducted to provide future directions for the area's development, showing more cited articles, authors, and more productive countries. For more consolidated conclusions, it is advisable to use tools such as text mining use of other databases, not limited to WoS, in addition to a broader period, identifying the initial research in the area.

The use of patent data and gray literature review articles are excellent means of providing more data for the work, since in this systematic bibliometric review, with the

keywords used (“hydrothermal”, “gasification”, and “biofuel”), the amount of data is still in its initial phase.

7 FINAL CONSIDERATIONS

A comprehensive overview of a systematic bibliometric review was provided with the keywords “hydrothermal”, “gasification,” and “biofuel”. Three hundred thirty-one articles were presented, which were refined to 311 articles. The RQs questions were analyzed using WOSviewer, CiteSpace, and Microsoft Excel (Microsoft Office 365[®])

The three journals with the most publications on the topic were: Bioresource Technology; Algal Research: Biomass Biofuels And Bioproducts; Biomass & Bioenergy; they are from England, Netherlands, and England, respectively. However, the most productive countries were China, USA, and Canada, not being the country of any of the first three journals. The most productive authors also cooperated well with peers, showing that this theme has been produced globally.

The analysis of the 12 most cited keywords identified the formation of 4 large clusters: hydrothermal liquefaction, pyrolysis, supercritical water, and hydrothermal carbonization. These showed a possible direction in which the research has been heading. From future perspectives, it is possible to identify that the studies are focused on bioeconomics with subtopics related to biorefinery processes, hydrothermal processes, gasification, and pyrolysis.

Thus, applying bibliometric techniques proved to be an instigating tool to identify the main characteristics of articles on biofuel production using HTG.

Acknowledgments

The current study was partially supported by the following Brazilian Agencies for Scientific and Technological Development: Fundação Cearense de Apoio ao Desenvolvimento Científico e Tecnológico (FUNCAP) (PS1-0186-00216.01.00/21), Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq) (311062/2019-9), and Coordenação de Aperfeiçoamento de Ensino Superior (CAPES) (finance code 001).

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Journal Pre-proof

FIGURE LEGENDS

Figure 1 – Distribution of publications on biofuel production from hydrothermal gasification between 2006 and 2022.

Figure 2 – Structure representing the search and analysis criteria.

Figure 3 – Geocoding of the organizations responsible for the publication of the 311 articles analyzed.

Figure 4 – National collaboration network of articles regarding HTG-produced biofuels published between 2006 and 2022 on WoS. The thickness of a line connecting two countries indicates the frequency of co-authorships (thicker lines mean more published articles), and the color clusters indicate groups of countries with a high level of collaboration.

Figure 5 – Map of collaboration among institutions. The thickness of a line connecting two organizations indicates the frequency of co-authorships (thicker lines mean more joint published articles), and the color clusters highlight groups of institutions with a high level of collaboration.

Figure 6 – Map of collaboration among authors. The thickness of a line connecting two authors indicates the frequency of co-authorships (thicker lines mean more published articles), and the color clusters illustrate groups of authors with a high level of collaboration.

Figure 7 – Map of the relationship between citations on biofuel production through HTG between 2006 and 2022.

TABLE LEGENDS

Table 1 – Ranking of the 12 journals that have published the most on biofuel production by HTG between 2006 and 2022.

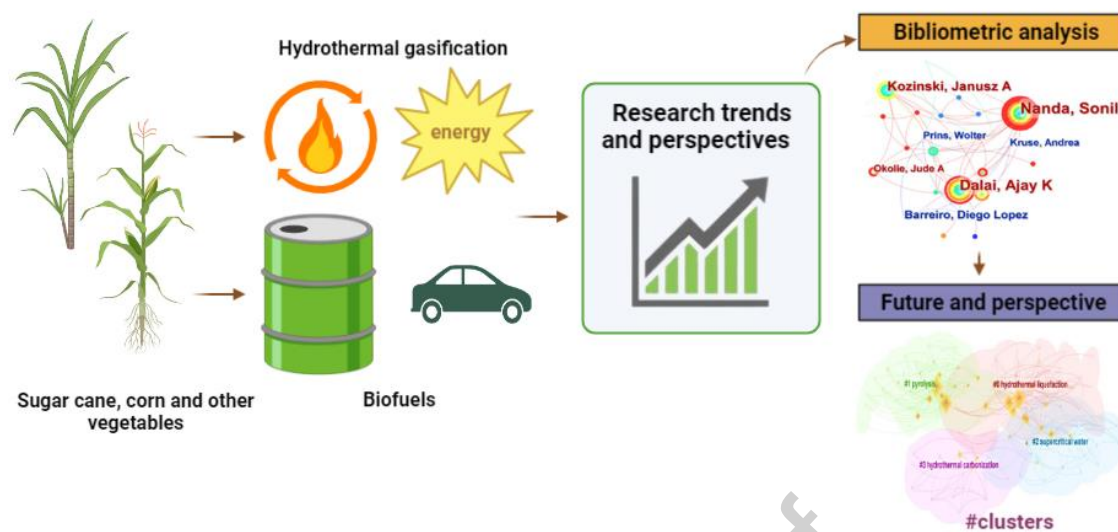
Table 2 – The 12 most productive countries in terms of HTG-produced biodiesel between 2006 and 2022.

Table 3 – Main publications on biofuel production by HTG between 2006 and 2022.

Table 4 – Frequency analysis of the 12 most used keywords in research on biofuel production by HTG between 2006 and 2022.

Table 5 – Frequency analysis of the 12 categories with the most articles in research on biofuel production by HGT between 2006 and 2022.

Graphical abstract



Conflict of Interest

The authors declare no competing financial interests.