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Review article

Technological prospection of microalgae-based biorefinery approach for effluent treatment

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ABSTRACT

Microalgae-based biorefinery coupled with effluent treatment (MBBET) is an approach that makes biomass production economically advantageous. It can produce value-added products from a circular bioeconomy perspective for a variety of industries. Although the global microalgae market is growing at a rapid rate of 7.39% per year and 70–90% of world's technological information is consolidated in patents, this source of technical data is still underexamined in the microalgae field. Technological prospecting is a powerful tool that provides insight into expanding knowledge and innovation in companies, providing inputs for competitive intelligence, improving decision-making, and forecasting technological changes. Thus, this review aims to fill this research gap by presenting a bibliometric analysis of patents conjugated with a natural language processing (NLP) text-mining approach to transforming raw big data into useful information. The state-of-the-art prospective patents on MBBET are disclosed, demonstrating the potential of this integration, its historic-technological trends, the different effluents treated, and bioproducts of market interest. The research was conducted using the Espacenet and Orbit® databases. The data were recovered from patent documents from the last 20 years using a combination of the keyword "Microalgae" and code C02F3 (Biochemical effluent treatment), where 422 documents were recovered from Espacenet and 664 from Orbit®. In this paper, we show that patents are a noteworthy source of information for the advancement of microalgal biorefinery areas, especially when they are compared to scientific articles. Although biotechnological potential of algal biomass is still underexploited, technological gaps and active technologies are highlighted. MBBET is a promising key to mitigate global environmental challenges and is an expression of the new paradigm of water-food-energy. Future challenges are proposed to sketch perspectives on new avenues for technological improvements.

1. Introduction

Environmental pollution, sustainable consumption of natural resources, and mitigation of climate change have become global challenges. It has been estimated that the demand for energy, water, and food will increase by 50, 80, and 60%, respectively, between the years 2000 and 2050 worldwide [1,2]. To meet the proposals stipulated at the 21st Conference of Parties (COP21) held in 2015, the "water-food-energy" nexus has been gaining immense popularity, bringing a new systemic approach to achieve global sustainable development [3,4]. Among the guidelines established by Hoff [5], the principal requisites of this nexus include efficiency in the handling of natural resources; the use of residues as a resource in a multiuse system; partnerships and coherence between academic institutions and the government; and the use of third-generation biofuels. The integration between biorefinery and biochemical effluent treatments using microalgal biomass could help meet the majority of these requirements.

The concept of biorefinery is analogous to the traditional petroleum refinery, which allows the sustainable generation of numerous high-value bioproducts from a single source [6,7]. Microalgae-based biorefinery is an approach that has been gaining attention because microalgal biomass production can be performed in an integrated way by using the circular bioeconomy framework [8]. These microorganisms are promising candidates as feedstock for biorefinery due to their

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versatility. Compared to plants, microalgae have superior photosynthetic efficiency, do not require arable land, excessive amounts of water, and dependence on seasonality. They have a relatively simple cell wall (i.e., without lignin), and notably, can grow in industrial effluents. Microalgae are capable of producing high-value substances such as *exo*-polymers [9], which can be used as potent bio-flocculants [10]. They are also able to produce bioplastics (PHA) with properties similar to polypropylene [11].

Among other products having marketing value, microalgae are used to develop bioactive compounds such as pigments and vitamins, which can be used by humans as an antioxidant, anti-inflammatory, antimicrobial, and antiviral agent [6]; the proteins produced can be utilized for feeding humans and animals [6,12], while the lipid fractions and carbohydrates can be converted into biodiesel and bioethanol, respectively.

Commercially, microalgae are cultivated in synthetic media, which confers a high cost to the final product. However, this value varies according to the scale of the study and the species utilized and can be reduced with the use of effluents. Chisti et al. [13] evaluated the costs of Spirulina biomass production plants and Costa et al. [14] adjusted the values considering inflation in June 2019, conforming to the guidelines of the US Department of Labor, Bureau of Labor Statistics (www.bls.gov). The studies demonstrated that the culture medium represented 13% (USD 677.000) of the total costs. Moreover, Feng et al. [15] showed that the total cost to produce 1 ton of *Chlorella vulgaris* biomass can be up to 3.5 times lower (from US$ 808.79 to US$ 231.59) if it is cultured in 1443 m^3 of an artificial effluent. Thus, the substitution of synthetic media with industrial effluents is an indispensable alternative to reduce the costs of producing microalgae while also treating effluents instead of using conventional physical, chemical, and biological treatments [16]. Wastewaters contain nitrogen and phosphorus in concentrations up to three times higher than in natural water bodies [17,18]. Microalgae require these nutrients for cellular structure development and energy exchange [19]. Therefore, these microorganisms can perform effluent bioremediation by degrading contaminants to obtain nutrients and converting them into less toxic substances [20].

Due to several benefits of microalgae-based biorefinery coupled with biochemical effluent treatment (MBBET) and its growing interest among the scientific community, it is expected that newer technologies will be developed in this area in the coming years. It has been predicted that the global microalgae market will reach US$ 1.143 billion by 2024, expanding at a compound annual growth rate of 7.39% [21]. To evaluate the likelihood of this scenario, technological prospection is a relevant tool that uses bibliographic analysis for mapping developing trends in the field. This process is aimed at identifying strategic areas of research and emerging technologies that are likely to generate greater economic and social benefits [22].

Patents are a source of technological information and measurable output of research and development (R&D) activities worldwide. Besides an increasing number of patent applications as an indication of their growing relevance, it is suggested that depending on the years and the technical domain, 70–80% of the information contained in patents cannot be found elsewhere [23].

Abbas et al. [24] stated that investigating patents within a specific technical domain supports organizations in numerous ways, such as defining novelty in patents; analyzing patent trends; identifying technological vacuums (i.e., fields in which patents have not yet been granted) and hotspots (i.e., fields in which patents have intensively appeared); forecasting technological developments in a particular field; and strategic technology planning. While patents provide insights for innovation in both public and private institutions, there are challenges of working with an extensive volume of structured content (e.g., patent citation, patent classification) and especially unstructured content (e.g., abstracts, claims, and technical description) because of the nature of the data as well as their lack of categorization. To circumvent this problem, different techniques have been proposed within two sets: text mining and visualization approach. Text mining involves extracting information from unstructured data in the text and mining the text to discover rules and patterns by using lists, maps, and trends obtained from publication databases [25]. Visualization approaches, on the other hand, express predominantly structured data as visual information, where patent maps or clustering methods can be employed to extract useful information [24]. Since patents in the area of microalgae as a source of technical information are still underexamined, this hybrid approach that can properly analyze them would benefit technological prospecting as an attempt to advance innovation in this growing field. Garrido-Cardenas et al. [26] developed a bibliometric study on scientific publications on microalgae, and Moreno-Garcia et al. [27] complemented this with a summary of microalgae-based biorefinery using effluents. However, to the best of our knowledge, there is a lack of extensive studies exploring the MBBET in the ambit of patents.

This review presents a bibliometric analysis of patents conjugated with a text-mining approach to transform large amounts of raw data into useful information. The state-of-the-art prospective patents on MBBET are presented in this work, demonstrating the potential of this approach, its historic-technological trends, technological profile of the institutes, geographical representation of the patents, different effluents treated, and the developed bioproducts of great market interest. This review is organized as follows: Section 2 explains the methodology used for the systematic review of patents; Section 3 presents the results and discussion divided into the following aspects: retrieving of worldwide data and technology-time evolution (Section 3.1); major technological domains (Section 3.2); geographical coverage (Section 3.3); profile of the institutions and top applicants (Section 3.4); analysis of effluents (Section 3.5); profile of technology applications (Section 3.6); dominant technology to biofuel purpose (Section 3.7); keyword clustering and microalgae employed (Section 3.8); Section 4 provides the prospects and challenges; and Section 5 outlines the final considerations.

2. Methodology

2.1. Retrieving the data and software used

The technological prospection method was applied based on data recovered from patent documents of the last 20 years, using two search tools: Espacenet and Orbit®. Espacenet is a free access patent information database developed and provided by the European Patent Office (EPO). Orbit® is a private platform of the Questel company dedicated to patent research and analysis. The data obtained from Espacenet were imported using the CSVed software, compiled, and exported to Microsoft Excel.

Orbit® was used in addition to the Espacenet research due to its comprehensive analysis tools, especially to search the predominant technological domains and clusters of keywords. Clusters are a useful visualization approach within patent analysis techniques. Orbit® allows one to analyze the principal depositors in each cluster and the number of times each word is repeated. The search was conducted in November 2019 using international classification codes and keywords relevant to the subject under investigation.

The International Patent Classification (IPC) is a classification system of language-independent symbols to organize patents based on their technical fields. Hence, the search began with the collection of words and codes relevant to the topic under investigation. The words “Microalgae” and “Biorefinery” were individually crossed using the operator “and” with distinct IPC codes C02 (water and effluent treatment), C02F3 (biochemical effluent treatment), and C12 (biochemistry and microbiology). On the platforms, these combinations were sought both in the “title” and “abstract” fields to increase the scope of this search.

Once the appropriate combinations were made, the documents were retrieved and processed in CSVed software; all abstracts of the patents were read, and the information was separated and classified in Excel according to their strategic topics. Providing structured data (such as time evolution, geographical coverage, top applicants, and institutions) in patent forms are bibliometric features and obtaining this information requires only compilation and counting without reading whole documents.

In contrast, unstructured data that contain technical information (such as effluent utilized, final technology developed, type of industry, and their specificities) were acquired by reading each document, followed by compiling, categorizing, and counting the files. In this text-mining step, key findings were extracted from the abstracts and transformed into simple language structures using the subject–action–object (SAO) model as a reference throughout the process. To identify the functions of a particular technology, SAO is a frequently used, natural language processing (NLP) method in which subjects and objects can be related to the components of a system and actions can refer to the roles assigned to these components [24,28,29].

In this study, "Subject (S)" portrays the examination of microalgae species that were utilized in patents; "Action (A)" represents the search to determine if the microorganisms were used for biomass valorization, bioremediation, or both; and lastly, "Object (O)" indicates the sources of wastewater and their specificities. Patterns were identified and graphics were generated to show the key factors encountered. Later, comparisons were made between the current patents analyzed in this study and the published scientific articles in this field to identify trends and relationships.

2.2. Schematic representation of methodology

The chronological sequence for the research method was proposed based on the following hybrid approach: part one involved a bibliometric analysis (Fig. 1), while the second part utilized the text-mining method (Fig. 2). These schematics summarize the methodology applied, encompassing qualitative and quantitative analyses. Fig. 1 shows the steps in the development of bibliometric search.

Fig. 2 illustrates the second stage of the process, which consists of reading the documents, extracting and gathering the information according to the NLP-SAO method.

The term "wastewaters" was divided into four distinct blocks according to the origin of the effluent: (i) domestic sewage (residences, institutions, and commerce) and the digestate or biogas slurry; (ii) industrial effluents (refers to those specified according to the type of industry); (iii) specific environmental issues (S.E.I) dealing with eutrophicated water bodies, methods to attenuate algal blooms (using a biological remediation agent), or methods to treat polluted water bodies in the laboratory; (iv) N.T.S. ("not specified") refers to documents that do not specify the origin of the effluent.

3. Results and discussion

3.1. Retrieving of worldwide data and technology-time evolution

As observed in Table 1, the combination of "Microalgae" and IPC code "C02F3" was chosen due to the specificity of the content included and number of patents. Although the crossing of "Microalgae" and the IPC code "C02" retrieved more patents, this combination goes beyond the scope of the work, as physical and chemical treatments were also contemplated.

From the 422 documents recovered from *Espacenet*, only 391 were available for consultation since the others were still in the confidentiality period that lasted for 18 months. On the other hand, using the same word combination in *Orbit*®, 664 documents were found, of which 204 were expired patents.

Fig. 3 illustrates the changes in the number of documents associated with MBBET in the last 20 years indicating the dynamics of the inventiveness in this field.

From 1998 to 2005, the development of this technology was stable, with an average of approximately two documents per year. The data from 2006 revealed an increase with an average of seven documents per year, indicating advances in the development of this technology. This tendency could be associated with the third petroleum crisis (2003–2008) when the price of a barrel of petroleum increased nearly five times [30]. In parallel, during the same period, microalgae were hinted at as a viable source of biofuels, which motivated large energy companies to take an interest and invest in uncovering the biotechnological potential of microalgae. In 2007, Nozaki et al. [31] investigated and expressed the genome sequence of microalgae, arousing interest in the scientific community to perform new research in this area based on the newly acquired information.

Garrido-Cardenas et al. [26] demonstrated that the number of scientific papers published had a vertiginous rise from 2006, analogous to the patent documents presented in Fig. 3. In the ambit of patents, the period from 2010 to 2011 remained constant with an average of 11 documents; however, between 2011 and 2018, the average number of documents was 43. This value was 20 times greater than that at the start of the century and could be linked to the search for alternatives to meet the demands stipulated at global sustainable development events such as COP18–24 (2012–2018), RIO + 20 (2012), and the creation of "water-food-energy" nexus in 2011. A second factor for this trend could be associated with the development of internal policies in each continent. Notably, the European Common Market adopted an ambitious strategy for the development of a bioeconomy based on the use of sustainable

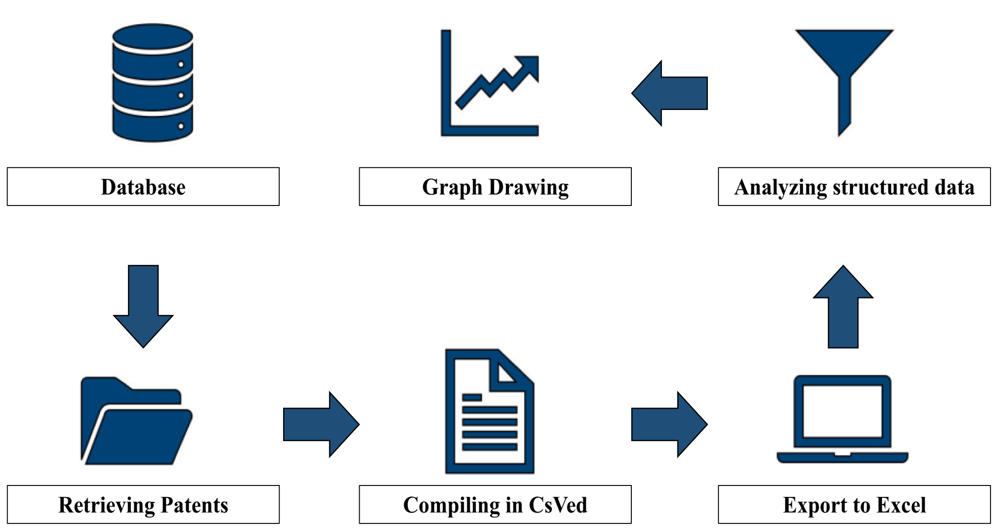

Fig. 1. Schematic representation of the bibliometric analysis (part one).

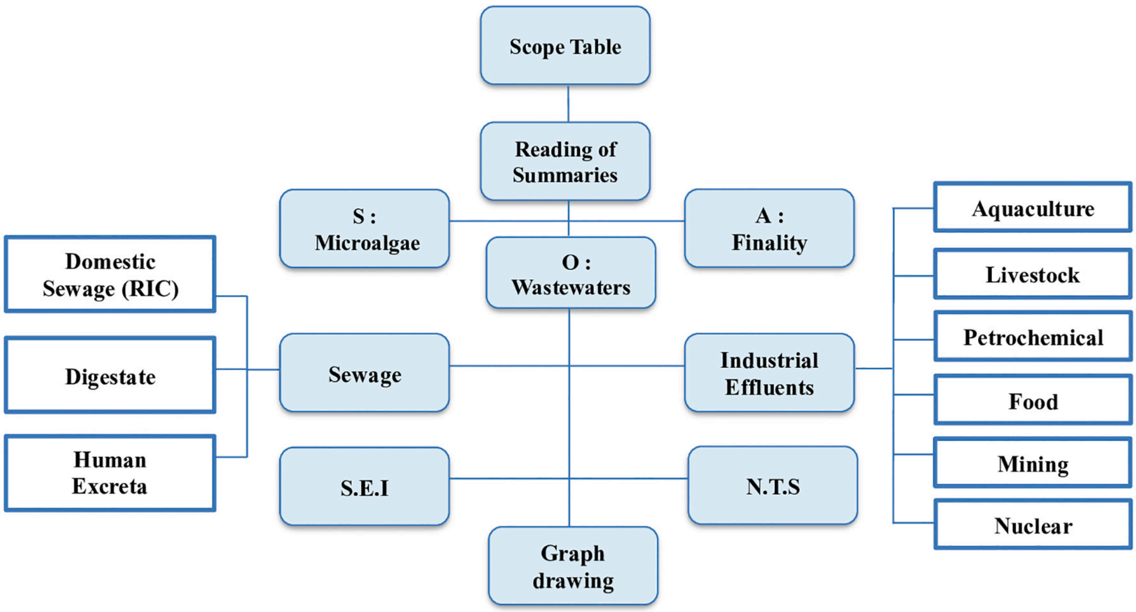

Fig. 2. Schematic representation of the text-mining approach (part two).

Table 1
Scope of technological prospecting based on patent documents

Microalgae	Biorefinery	C02	C02F3	C12	Total
X					5873
X		X			688
	X				238
X				X	299
X			**X**		422
X			X	X	217

Source: Elaborated by the authors based on Espacenet. C02: Water & effluent treatment (physical, chemical, and biochemical); C02F3: (biochemical effluent treatment); C12: (biochemistry and microbiology).

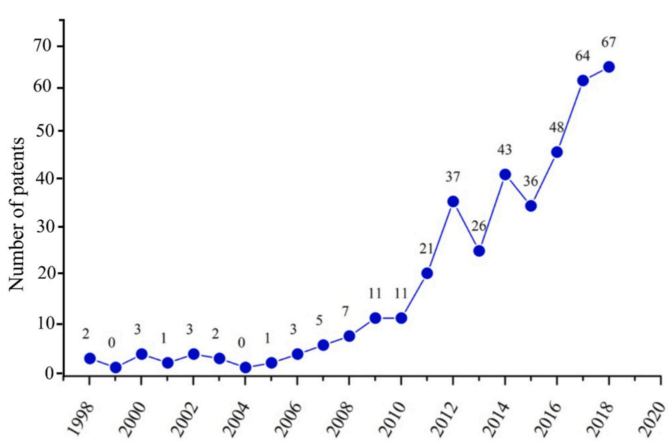

Fig. 3. Frequency of filing for patent documents based on effluent treatment by microalgae in the last 20 years.
Source: Elaborated by the authors based on Espacenet.

resources to meet the increasing demands in the food, energy, and industrial sectors. This "green economy" initiative aims to attenuate emissions, increase the efficiency and use of natural resources, and prevent the loss of biodiversity by 2050 [32,33]. The United States, through the Energy Independence and Security Act (EISA-2007), offered financial support for R&D activities aimed at the production of renewable fuels, including algal biomass as feedstock [34]. They sought to reduce the dependence on oil and move towards higher energy security.

Overall, there is an increasing global R&D interest in the development and application of MBBET. This is evident from the significant increase in the number of patents and scientific papers on this topic, especially after 2011. In addition, energy and environmental policies tend to accelerate this trend, promoting more R&D budgets. While a stable profile is a sign of sector maturity, a sector with intensive growth is indicative of a race for patents to overcome technological obstacles.

3.2. Major technological domains

Patent portfolios can be divided into different domains. Fig. 4 shows the predominant technological domains of the prospective patents, where a single patent may be linked to more than one segment. Environmental (659 documents) and biotechnological (340 documents) sectors have the highest contributions, which is evidence for the recognition of the biotechnological potential of microalgae by many countries. All the patents prospected in the current analysis are thus related to the technological domain that focuses on the environment.

The heterogeneity between the technological domains can be understood by two biases: (i) the technical interdisciplinarity involved in the bioremediation process of effluents; and (ii) the high-value bioproducts that can be obtained and applied in industrial sectors, such as food (exopolysaccharides and proteins), pharmaceutical and cosmetic (phycocyanin and lipids), biofuels (lipids and carbohydrates), and materials (polyhydroxyalkanoates). Since small firms based on microalgae will grow in the next few years, understanding the predominant technological domains by learning from outside sources can guide them in their initial process. In this context, less eminent sectors are gaps for R&D as they are opportunities to explore and initiate new technologies, whereas the most notable fields tend to indicate potential products and processes for commercialization [35]. Even though this is highly acknowledged by the scientific community, it portrays a lack of development on its market potential.

3.3. Geographical coverage

An analysis of Fig. 5 reveals that of the total documents of patents

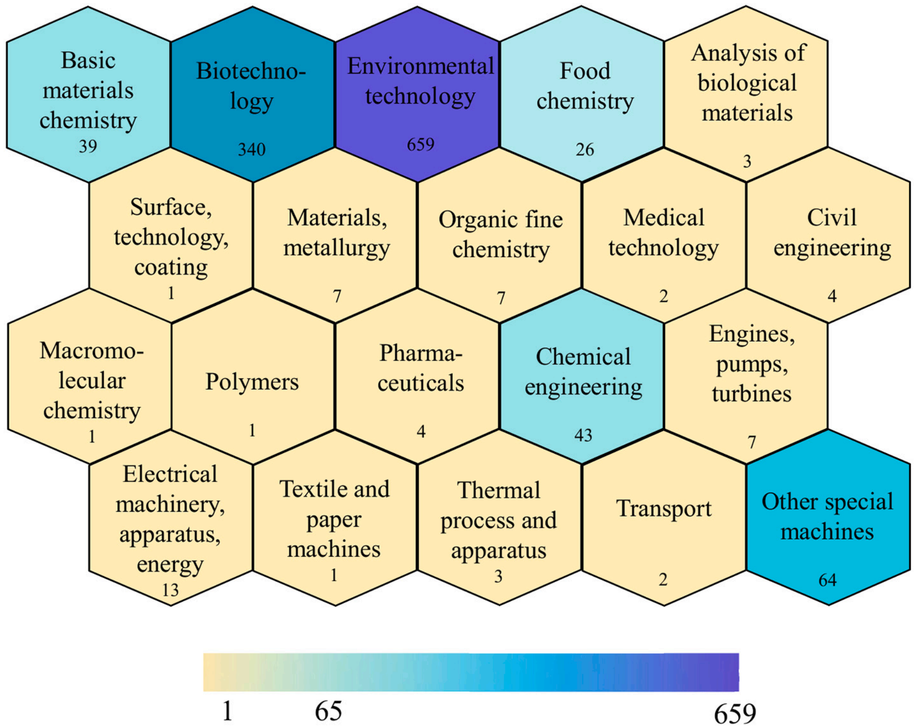

Fig. 4. Patent documents based on effluent treatment by microalgae in various technological domains. Source: Q. Orbit, ®.

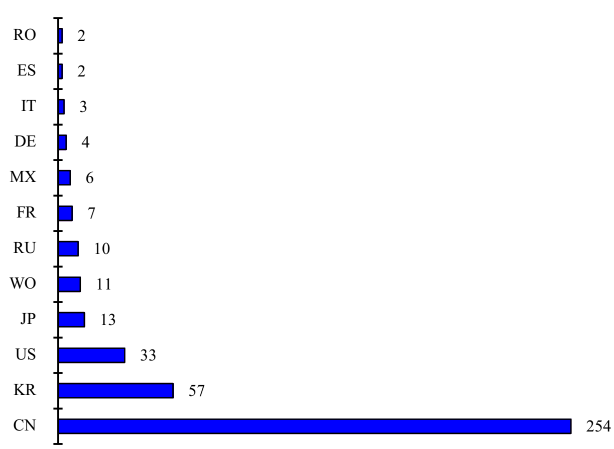

Fig. 5. Number of patent documents based on effluents treatment by microalgae versus their origin.
Source: Elaborated by the authors based on Espacenet.

recovered, 65% are from China, 15% from South Korea, 8% from the USA, and the rest are distributed among the remaining countries. This trend is not the same for the number of research articles related to technologies involving microalgae. As emphasized by Garrido-Cardenas et al. [26], although the development of technology is mainly in China, the USA is in first place in terms of scientific publications, demonstrating that China is more focused on developing technologies with market applications.

Besides being the most populous country in the world, China has several challenges that could justify the massive investment in MBBET technologies. It has limited arable land for food production since desertification is widespread in many areas [36,37]. Although cattle breeding has increased, the land available for pasture has diminished [38]. Additionally, the import of European cattle breeds such as Friesian and Simmental, which demand greater food requirements, has further aggravated the scenario [39].

Despite its high energy consumption, China also suffers from limited petroleum reserves, lack of freshwater [40] and environmental pollution [41]. As an alternative to petroleum reserves, Zhao and Yan [41] stated that the Chinese Ministry of Science and Technology is heading a biomass energy development project as a governmental priority, making significant funding available in this area. In this emerging green economy, public finance plays an important role in dealing with the high-risk and uncertain nature of the innovation process, which takes time to develop new technologies. Mazzucato [42] argues that a “green revolution” will depend on proactive governments that have an entrepreneurial mindset. This refers to the state's willingness to invest in uncertain areas envisioning the changes across public agencies and government departments and the public-private coalition.

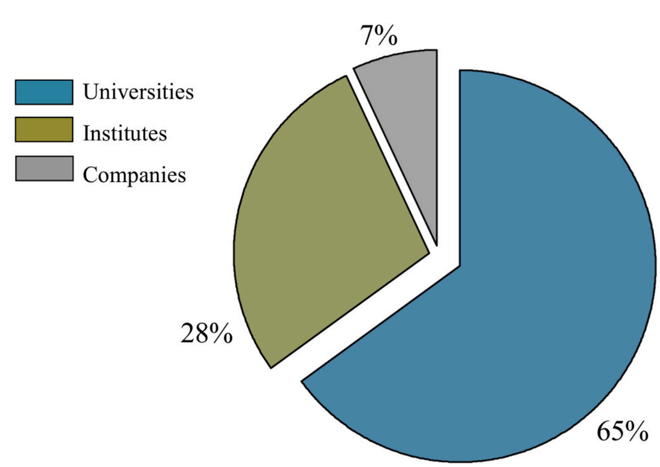

Fig. 6. Percentage of institutions that develop technologies based on effluents treatment by microalgae.

3.4. Profile of the institutions and top applicants

According to Fig. 6, there are no independent inventors for the development of prospected technology, and universities contribute to a major part, represented by 65% of the documents, as opposed to 28% for research institutes and 7% for companies. One should consider that partnerships between companies and universities or research institutes could be relevant for the development of patents since companies will be able to provide funding and different effluents necessary for the analysis and this synergy will, in turn, create a circular bioeconomy.

Another noteworthy factor for the 65% output is due to the changing role of universities in the last few decades. Within the knowledge-based economy paradigm, universities have been shifting from the former mission of creating and providing scientific information to more entrepreneurial institutions [43]. This model plays an enhanced role in technological innovation by incorporating two new tasks: knowledge marketing and active contribution to the development of local and regional companies. Notably, technology transfer offices (TTOs) established by universities can provide business models and explore local partnerships (e.g., venture capital, entrepreneurs, firms) to commercialize innovations and generate revenue.

It is worthwhile to understand the innovation chain (from basic research to commercialization) existing in the current market dynamics. Phaal et al. [44] provide a framework to understand the phases of emerging technologies by dividing them into five stages that are preceded by technological maturity. These phases are (i) science-dominated (precursor), (ii) technology-dominated (embryonic), (iii) application-dominated (nurture), and (iv) market-dominated (growth). According to the authors, as this chain progresses, the risk associated with commercial uncertainties decreases, technological development occurs, and there is a tendency for a shift in funds and controls, usually from universities to industrial level research and development. Nevertheless, as demonstrated in Fig. 6, the patents from research institutes are still less than half the percentage obtained from universities (28%), and companies only represent a small part (7%). This could indicate that the technologies encompassing MBBET are still far from reaching the maturity phase and ultimately the final stage of market dominance.

As it is a multidisciplinary sector that encompasses different types of technologies at different levels, it is challenging to precisely characterize the stage in which the MBBET group currently belongs. It could be placed in the transition towards the nurture phase since there are improvements in performance and price that need to be made to attain its full potential and develop into a sustainable business.

With the new paradigms for achieving technological sophistication in the current century, universities have been playing an important role in furthering microalgal research by providing solutions for companies, especially in China and South Korea. It is hoped that this framework can be scaled up globally, especially in developing countries.

To further examine the various contributing institutions, the top 12 major developers in the MBBET group are presented in Fig. 7. This figure enlists the principal inventors within universities, research institutes, and companies, providing a degree of internationalization in the patent portfolio.

The Chinese state university Zhejiang Ocean University holds 16% of the world's patents in this area and is 2.3 times more than the second-place holder, the Korea Institute of Science & Technology. It is worth emphasizing that there are no American, French or Spanish institutions in the list, although these countries publish many scientific articles in this field [26]. South Korea is listed as the tenth country in the number of scientific publications. However, Figs. 5 and 7 demonstrate that they are the second-highest in technological development. Two aspects should be highlighted when it comes to South Korea. First, they have a well-established culture of innovation through a collaborative relationship between the university, industry, and government (UIG), known as the triple helix framework [45]. In the rush for industrialization, the creation of the Korean Advanced Institute of Science and Technology (KAIST) has substantially contributed to technological development and economic growth in Korea [46].

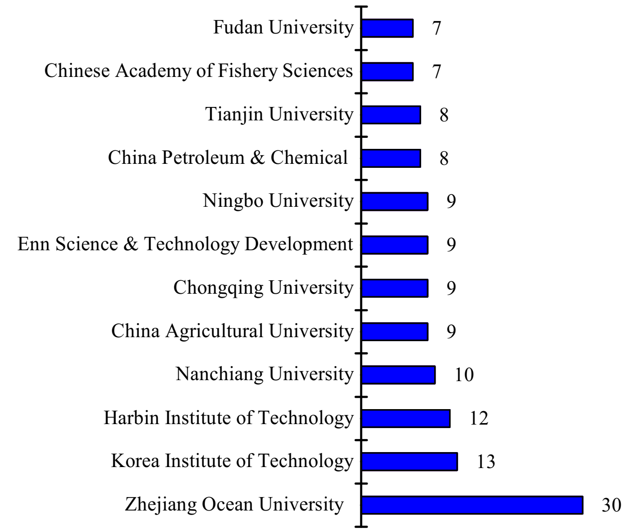

Fig. 7. The major depositors of patent documents based on effluents treatment by microalgae.
Source: Q. Orbit®.

The second point concerns the "Biotech 2000" and "Bio-Vision 2016," national programs headed, respectively, in 1994 and 2006 by South Korea, making it one of the largest biotechnology producers in the world [47,48]. Korea and China are eminent countries in developing technologies in the MBBET sector and have two aspects in common: an innovation ecosystem based on collaboration between public and private initiatives as well as a robust and national technological development project.

In general, Table 2 summarizes the differences in the profile of technologies among Zhejiang University, KAIST, and China & Petroleum, since they are the principal institutions of each category. Zhejiang University, which stands out among the institutions, is primarily focused on developing microalgae-based adsorbents to treat distinct contaminants. KAIST is centered upon treatment devices and methods to take advantage of microalgal biomass, mainly through anaerobic digestion technology.

Another feature presented in this institute is the development of new microalgae strains, including Micractinium sp., (**KR101624176**) [49] and Nephroselmis sp. KGE 1 (**KR101799943**) [50]. The second is a method to simultaneously treat industrial effluent and cultivate microalgae, aiming to accumulate carbohydrates, that is, the MBBET perspective. The China Petroleum & Chemical company still has little participation in the patent portfolio, but it also has devices and methods for the continuous cultivation of microalgae while treating its effluents from industrial processes, such as low reuse effluents rich in nitric acid (**CN109337820**) [51], wastewater rich in nitrate (**CN109467191**) [52], and the denitrification of flue gases without a catalyst (**CN109939548**) [53]. Although the profile of these institutions might differ, the three technological biases (e.g., ecological adsorbents, novel strains, and devices and methods in alignment with MBBET) are relevant to different market needs.

Table 2
Main characteristics of technologies developed by the institutions.

Institution	Type of patent	Features
Zhejiang Ocean University	Products and methods	Bioadsorbents compounds
Korean Institute of technology	Devices and methods	Novel strains and anaerobic digestion technology
China Petroleum & Chemical	Devices and methods	Continuous microalga cultivating industrial process

3.5. Analysis of effluents

To determine the main effluents used by the technologies, the different categories were quantified, as shown in Table 3.

Table 3 illustrates that the category N.T.S predominates over the others, but that the categories industrial and sewage are close, such that one cannot clearly affirm which effluent predominates in the field of these technologies. However, Fig. 8 highlights their variety, where sewage stands out. It must be mentioned that the columns "Industrial N.T.S" and "Sewage N.T.S" refer to the patents where the text described the effluent as coming from a specific industry, or it was just sewage, respectively, without specifying which segment. Moreno-Garcia et al. [27] reported the use of a wide spectrum of effluents as cultivation media for microalgae, which converged with the prospection of technologies carried out in the present study, principally for effluent digestates, agriculture, and oil. However, these authors did not include effluents from aquaculture or radioactive reactors, in contrast to those found in the present study in terms of technologies.

Concerning livestock, the majority of effluents are coming from swine breeding in China (**KR20180092545**, **CN105985917**, **CN108991281**, **CN108892243**, **CN108249574**, **and CN107916226**) [54–59] and also a mixture of effluents coming from cattle and poultry raising (**CN203474790**, **CN103451091**, **CN103086520**, **CN102161550A**) [60–63]. For aquaculture, the effluent is from shrimp preparation (**CN109601446**) [64]. With respect to the food and beverage industries, all the registered patents were also from production in China for soy sauce (**CN109626584**) [65], tofu (**CN109621699**, **CN107760605**) [66,67], starch (**CN10729962**, **CN104789603**) [68,69], and effluents from the alcohol or beverage industries (**CN109337816**, **CN106867953**, **CN104556566**) [70–72]. These effluents are sources of the principal nutrients, such as ammonia (NH_4 – source of Nitrogen) and phosphate (PO_4 – source of Phosphorus). Studies have reported that ammonia is the nitrogen source preferred by algae, even though high concentrations are toxic to them [73,74]. The effluents containing starch are also advantageous since they are potential carbon sources for microalgae. This diversity in the use indicates how China is engaged in adhering to the "water-food-energy" nexus and how the Occident could learn to add further value to their effluents.

On the other hand, effluents from the nuclear industry require attention because the contamination of water with radioactive compounds represents a serious problem. The Republic of Korea has the largest number of patent documents in this area (3). Patents (**KR20190017447A-KR101556655B1-KR20170123044**) [75–77] deal with the development of a system to decontaminate water polluted with radioactive compounds by way of microalgae. In addition to Korea, a patent developed in Spain and protected by the World Organization of Intellectual Property (**WO2014087030**) [78], consists of a method for recovering and enriching uranium by bioaccumulation in genetically modified microalgae. Because the cell walls of microalgae contain negatively charged groups, the adsorption of positively charged compounds, the heavy metals, becomes an energetically favorable process [79].

For the problem of eutrophication, 14 patent documents (8 from China, 3 from Korea, and 3 from Russia) deal with methods to treat "algal bloom" or "flowering", mainly between the red tide, brown tide, and green tide. The patents aimed to develop an ecological remedial

Table 3
Main effluents used in the cultivation of microalgae.

Wastewater	Number of documents
N.T.S	116
Industrial	105
Sewage	106
S.E.I	64

N.T.S: "not specified", S.E.I: "specific environmental issues".

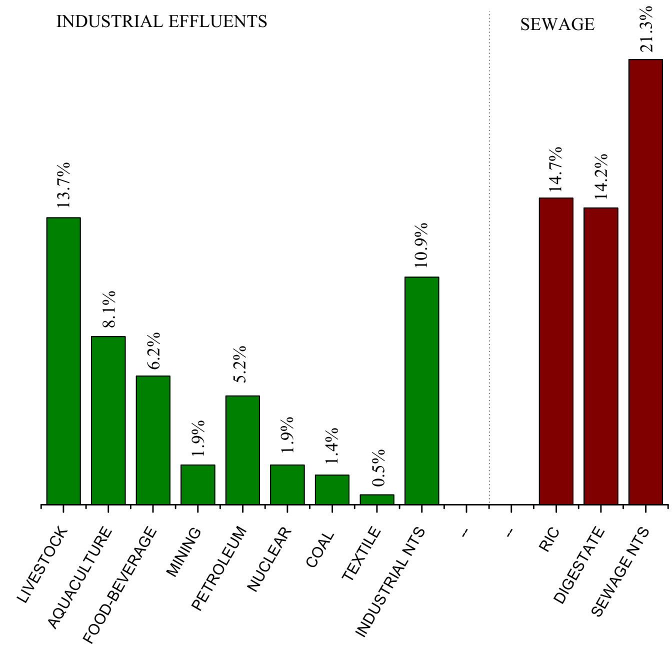

Fig. 8. Percentage distribution of patent documents per segment of effluents used in the technologies involving the cultivation of microalgae.

agent that, in most cases, used microalgae as the main compound. The document (**CN108949618A**) [80] deals with a method using the algaecide bacterium *Lysinibacillus* sp., which has an excellent effect on the death of the majority of cyanobacteria prone to flowering, including *anabaena*, *cylindrospermopsis raciborskii* and microcystis. The document (**CN107096565**) [81] deals with the development of a microcystin-degrading catalyzer prepared by the uniform mixing of various substances (including powdered microalgae). In addition, the document (**CN109618997**) [82] deals with an ecological purification method for oyster cultivation, removing the heavy metals using microalgae and aiming to increase the quality of their consumption.

Table 4 summarizes the different relevant contaminants. A wide range of heavy metals, radioactive complexes, and compounds can be seen, proving the bioremediation potential of microalgae on a technological scale.

At the end of Table 4, note the presence of specific adsorbents and microalgae-based bio-compounds used to remove diverse contaminants. As can be seen, the possibility of developing microalgal biomass is independent of the nature of the contaminant. The document (**CN105754860A**) [91] mentions the synergy between two distinct microalgae, promoting a greater assimilation of nitrogen and phosphorus at 8% and 15%, respectively. In addition to the better bioremediation capacity, another factor that the microalgae compound technologies highlight is the ease in recovering the material. The patent presented in (**CN109395700**) [93] states that the microalga used was *Chlorella*, and also points out that one of the problems in the treatment of effluents with microalgae is the difficulty in separating the bio-compounds, as well as the high cost associated with this practice. To resolve this inconvenience, the technologies have been using magnetic nanoparticles such as magnetite (Fe_3O_4) to aid in the separation of the bio adsorbent from the medium, allied to sodium alginate to improve the adsorption capacity. This technology is an example of a synergic approach between biotechnology and nanotechnology cited by Koyande et al. [97], and thus eliminating the costly processes involved in harvesting algae.

Considering that this biocomposite technology is a simple, low-cost solution encompassing a broad spectrum of recalcitrant contaminants, as shown in Table 4, they have high market potential. Companies

Table 4
Principal organic, inorganic, and radioactive contaminants used as cultivation media for the microalgae and the bio-compounds based on microalgae applied to these contaminants.

Document	Depositor	Wastewater	Contaminants	Compound
KR20170123044 [77]	UNIV SOGANG RES FOUNDATION	Industrial effluents - nuclear	Radionuclide	–
KR20190017447 [75]	ORION ENC CO LTD	Industrial effluents - nuclear	Radioactive pollutants	–
CN102586117 [83]	GUANGZHOU EP ENVIRONMENTAL ENGINEERING LTD	S.E.I	Lead	–
CN107381821 [84]	MARINE FISHERIES RES INSTITUTE OF ZHEJIANG	S.E.I – contaminated seawater	Mercury	–
CN109574235 [85]	UNIV WUHAN TECH	S.E.I	Mercury	–
JP2007209365 [86]	NIPPON SHEET GLASS CO LTD	S.E.I	Arsenic	–
WO2018158752 [87]	SYMBIOSIS INTERNATIONAL UNIV	S.E.I	Copper- cyanide complex and a zinc-cyanide complex	–
CN102745818 [88]	UNIV CHONGQING TECH & BUSINESS	S.E.I	Cadmium	–
CN107364971 [89]	MARINE FISHERIES RES INSTITUTE OF ZHEJIANG	S.E.I	Copper	–
CN108970588A [90]	UNIV ZHEJIANG OCEAN	NTS	Lead	(Fe_3O_4) nanoparticles + microalgae
CN105754860A [91]	UNIV HUNAN	Sewage (biogas slurry)	The removal efficiency of the composite microalgae on nitrogen and phosphorus is improved by 8%, and 15%, respectively	The synergistic effect of two kinds of microalgae is used
CN105198095A [92]	UNIV ZHEJIANG OCEAN	Industrial effluents	Xylene	Microalgae + fungi to form symbiotic spheres
CN109395700 [93]	UNIV ZHEJIANG OCEAN	Sewage	Cadmium (Cd) – Good removal effect was obtained	*Chlorella* + nanoparticles + (Fe_3O_4) + sodium alginate
CN109355278 [94]	UNIV ZHEJIANG OCEAN	S.E.I - seawater	(PAHs) the rate of degrading PAHs can reach 82–93%	Microalgae + bacteria immobilized in pellets
CN109225164 [95]	UNIV ZHEJIANG OCEAN	Industrial effluents – aquaculture (marine culture wastewater)	Antibiotic pollutants can be effectively removed, and the composite is easily recovered	(Fe_3O_4) nanoparticles + microalgae + sodium alginate + inorganic salt
CN109174025A [96]	UNIV ZHEJIANG OCEAN	NTS	Good removal effect of chromium (Cr) can be obtained	Microalgae + magnetic nanoparticles + konjac glucomannan + modified carbon fiber

employing this technology are already moving from the linear paradigm of "extract, produce and dispose of" to the circular paradigm because the treated effluents will have the potential to be used as new inputs for their respective processes. Moreover, these technologies are generally developed by the institution that most filed patents in this area, according to Table 3, and belong to the two most explored technological domains shown in Fig. 4. Because of these factors, this is a very active technology in the prospected portfolio, that is, it is a hotspot technology.

To see other hotspot technologies as well as technological vacuums, Fig. 9 shows the profile of the application of the technologies associated with the patents recovered.

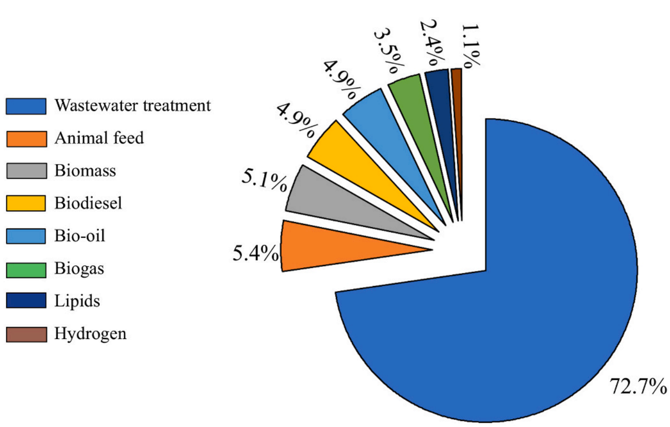

Fig. 9. Percentage distribution of patent documents based on effluents treatment by microalgae per purpose.

3.6. Profile of technology applications

It appears from Fig. 9 that most of the documents contained methods focused only on the treatment of effluents with microalgae. It should be noted that biorefinery can only be achieved if one takes advantage of the biotechnological potential of the microalgal biomass. The International Energy Agency (IEA) reports that biorefinery is still a concept under construction to be fully realized [98]. Thus, the absence of greater use of microalgal biomass could be evidence of the incipience of biorefineries in the ambit of developing technologies. However, three possibilities were identified regarding the purpose of patents: (i) the methods are focused on treating the water to recycle it in the process, reducing the costs; (ii) the technologies were developed to solve environmental challenges (algae bloom) or remove heavy metals from polluted water bodies; or (iii) in the concept of biorefinery, the technologies aim to simultaneously treat the effluents and offer a finality for the microalgal biomass.

As Koyande et al. [97] analyze some trends in the microalgae market, they point out that although the focus of recent years has been on biofuels, these would be losing first place to another market that is also rapidly expanding, namely proteins. As shown in Fig. 9, this trend appears to be a reality from the standpoint of filing patents.

Animal supplementation was the second principle of finality, mainly concerning the areas of livestock and aquaculture. The document (**CN107487859**) [99] highlights the use of microalgae in the treatment of water and its subsequent use as a poultry additive (protein source), whereas the document (**PH12017500826**) [100] highlights the treatment of the waters of an aquarium conjugated with its use as a food source for the fish. The inventors in (**CN108991281**) [56] disclosed a method to use the microalgae in piggery wastewater, which grows rich in carotenoids, as golden carp feed, resulting in a lower mortality rate and a faster growth rate. The method is divided into three steps, one of which is designed to pre-treat the effluent in order to dilute, disinfect, and sterilize it. Notwithstanding the relevant concerns about using microalgae grown in effluents for food production, these technologies may begin to demonstrate some viability. There are still no technologies for direct human consumption, but there is a commencing presence of technologies that may have the goal of indirect feeding, that is, through animal feed. Hence, the type of effluents utilized and the pre-treatment steps before the inoculation of microalgae in the wastewaters are pivotal factors that need to be taken into consideration for future methods aimed at this endeavor.

According to Hemaiswarya et al. [101], microalgae have distinct applications in aquaculture, and their use is mainly to provide nutrition and to enhance the color of the flesh of salmonids. The scientists also reported that several larvae of mollusks, echinoderms, crustaceans, and fish feed on microalgae, and the most frequently used species are the following: *Chlorella*, *Tetraselmis*, *Isochrysis*, *Pavlova*, *Phaeodactylum*, *Chaetoceros*, *Nannochloropsis*, *Skeletonema*, *and Thalassiosira*. These patents converge with what was reported in the literature, as according to the review proposed by Koyande et al. [102], microalgae are already a source of food for ruminants, pigs, birds, and aquatic species.

On the other hand, metallic nanoparticles (NPs) are a promising technology gap that has not yet appeared. Even though it was mentioned that these particles are used as a constituent part of bioadsorbents, they are likely not to come from the biosynthesis performed by microalgae. In contrast to the conventional chemical and physical routes, biological synthesis of NPs has numerous benefits as it is an economical path that does not require the use of high temperatures, high pressures, or harmful chemical solvents [103]. The advantage of this ecological route is also the alignment with MBBET because the effluents contain metal ions that are substrates for producing commercial nanoparticles by the bioremediation mechanism. Recently, it has been reported in the literature that the biosynthesis of NPs using microalgae is a new field that has potential for large-scale technological applications in various sectors, whereby some nanoparticles, such as copper (CuNPs), silver (AgNPs), zinc (ZnNPs), gold (AuNPs), and iron (II) and (III) [97,104,105]. Although microalgae have been considered as "nano-factories," metallic NPs are still a technological vacuum in the scope of patent research.

The term "biomass" in Fig. 9, refers to the documents containing technologies developed to produce biomass or those that mention that it was recycled without naming the ringed bioproduct. Although biofuels (biogas, biodiesel, bio-oil, and hydrogen) are the key topics of more recent publications, in terms of the development of technology, they are still relatively incipient, contributing to approximately 15% of the total number of documents. Since it is one of the pillars of the nexus "water-food-energy", the use of third-generation biofuels was divided into a separate graph, as shown in Fig. 10.

Of the biofuels produced from microalgae, the majority consists of bio-oil and biodiesel, each with 31.6%, the smaller part being hydrogen with 7%. Biodiesel is produced via the extraction and transesterification of algal oil, bio-oil is obtained via a thermochemical process, the commonly used process of pyrolysis, which is an economical method compared to other methods because it shows high efficiency and low energy consumption [106,107].

Most technologies aimed at bio-oil are probably related to its potential to replace petroleum, since bio-oil has an average of 80% of the energy content of petroleum [108,109] and is, therefore, a promising source of transportation fuels for the future [110,111]. Recently, the following four microalgae strains have been indicated as the most promising for biofuel production (*Tetraselmis suecica*, *Spirulina*, *Nannochloropsis* sp., *and Chlorella vulgaris*), as reported in [112]. In this technological prospecting only *Chlorella* species have been highlighted.

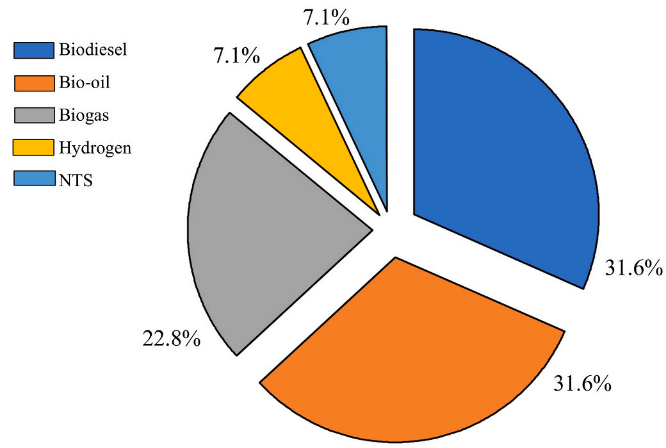

Fig. 10. Percent distribution of the types of biofuel produced by technologies involving the cultivation of microalgae in effluents.

Hydrogen is another fuel that can be derived from microalgae and has been extensively researched in recent years for future transport applications. The production of hydrogen by microalgae is still in its early development phase and can be carried out via three mechanisms: anaerobic fermentation, photo-fermentation, and electrolysis. By utilizing genetically modified strains, bio-hydrogen production can be improved in the coming years [113]. However, the patents encountered around hydrogen production have not dealt with the use of genetic engineering as yet. Three patents stand out, two being held by China and one by Japan, as from electrolysis processes (**CN105084674**) [114] and anaerobic fermentation of the microalgae biomass (**JP2003250519A**, **CN103981220A**) [115,116]. The part designated as "N.T.S" refers to patents involving biofuels, without specifying the segment.

It should be highlighted that although the production of third-generation ethanol has been widely discussed in the literature [117,118], the production of bioplastics as from microalgae biomass [119,120], no technologies are directed at producing bioethanol as from carbohydrates accumulated in the biomass or intracellular bioplastics have been reported in the literature. Since these technologies do not appear in Figs. 9 and 10, they can potentially be defined as technological vacuums.

3.7. Dominant technology to biofuel purpose

Concerning the hotspot technology to produce biomass for subsequent conversion into biofuels, different methods, including anaerobic bio-digestion, stand out. Fig. 11 shows a synthesis of the usual working mechanism of this technology, hereafter known as 5- steps technology. As its name suggests, this is a compilation of the methods used in a succession of five steps (i.e., pretreatment of residual waters, anaerobic bio-digestion, pretreatment of the digestate, microalgal biochemical treatment, and separation of the resulting biomass), which constitute the backbone of the technology represented by the rectangles, whereas the points represent the specificities found in each method.

The predominance of anaerobic digestion is in line with that demonstrated in Fig. 8, where the digestate contributed to 14.2% of the patents in the sewage category, as well as that which has been published in the literature concerning the use of the digestate as a culture medium for microalgae [121,122]. The residual water, most cited as coming from pig and cattle breeding, is pretreated in the first step so as to separate suspended solids. Subsequently, the digestate is designated for another pretreatment, where the probable focus would be to reduce any eventual turbidity and chromaticity, since its dark brown color can hinder light penetration to the cells, reducing the efficiency of microalgal photosynthesis [27]. These treatments can be carried out by natural solid-liquid separation or using flocculants. The digestate is then mixed

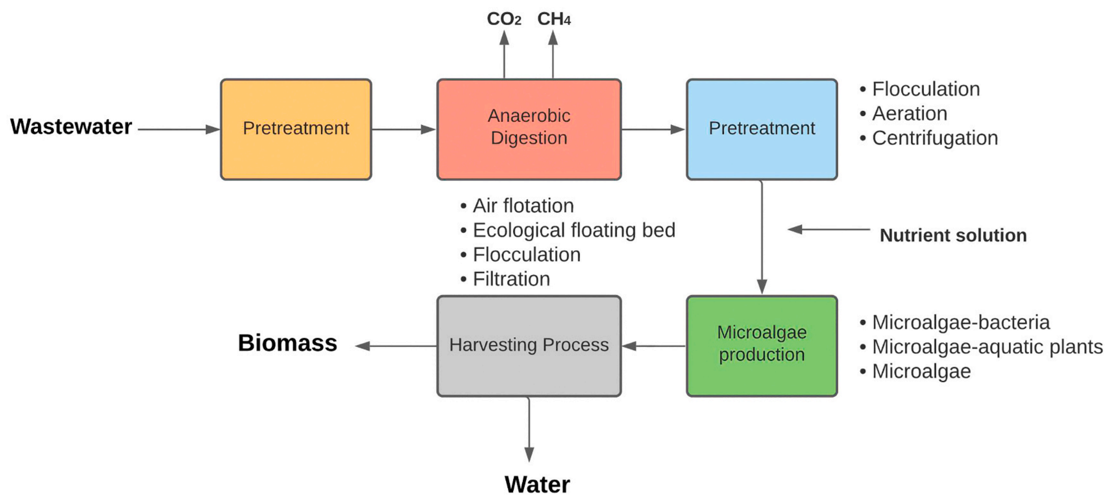

Fig. 11. 5-Steps technology flow diagram.

with a saline solution with a view to correct the pH of the medium and propitiate eventual adjustments in the available nutrients, since, although an ammonium-rich medium, the digestate generally presents a low phosphate concentration [27]. In the fourth step, the biochemical treatment is carried out by way of the growth of microalgae in photobioreactors, for which technology presents three alternative routes: (i) symbiosis of microalgae with bacteria (**CN106396112**) [123]; (ii) microalgae combined with aquatic plants (**CN106348451**) [124], or (iii) microalgae alone (**CN102433362**), (**CN108865892**), (**CN103663715**), (**KR20170039017**), (**CN101549932**) [125–129]. The bacteria-microalgae synergy exposed in some articles also shows a consistent and enhanced path in the scope of patents, since, in addition to increasing the efficiency of the biochemical treatment, it increases the growth rate of both microorganisms [122,130]. The bacteria degrade the organic matter producing CO_2 at the expense of the O_2 produced by the microalgae, while the latter harvest the CO_2 emitted to carry out photosynthesis. Moreover, the growth of many microalgae requires vitamins such as B12, and only prokaryotes can synthesize this vitamin [122,131]. Subsequently, in the last biomass separation step, the patents present varied and relatively simple methods (i.e., air flotation, ecological floating bed, filtration, flocculation). Because the separation process is responsible for approximately 30% of the total production costs [132], it can be seen that the cheapest and simplest methods were employed in the patents prospected.

The 5-steps technology also presents five main benefits, which were described in a pulverized way by inventors and are shown in Fig. 12: (i) low operational costs, (ii) simple operation, (iii) low ecological footprint, (iv) reduced biodiesel production costs, and (v) no secondary pollution.

Note that the role of this technology can be acquired in developing countries, since in these countries, about 80% of the sewage is disposed of into water bodies without prior treatment, culminating in environmental impacts, such as eutrophication [133]. Likewise, as mentioned previously, the majority part of these patents was developed in universities, showing the benefit of forming symbiotic innovated ecosystems in these countries, that is, strengthening public-private initiative ties, which is an essential condition to foment economic and sustainable growth. Such practices conform with the new paradigms of the 21st century, such as the water-food-energy nexus, as well as with the new knowledge economy and circular economy. The benefit of reducing the ecological footprint is welcome in any process, especially when conjugated with the economic factor and inserted in the current context of alarming environmental degradation indexes, where the current ecological footprint of the planet is actually higher than nature's "biocapacity" [98]. Some singular measures were noted for the 5-steps technology, where these patents are, except for those that used centrifugation, opting for increasingly simpler, cheaper biomass separation methods, and reporting strategies to increase energy efficiency. The inventors in (**CN102618446**) [134] cited the introduction of a purification step for the biogas CO_2 and its subsequent use as an additional

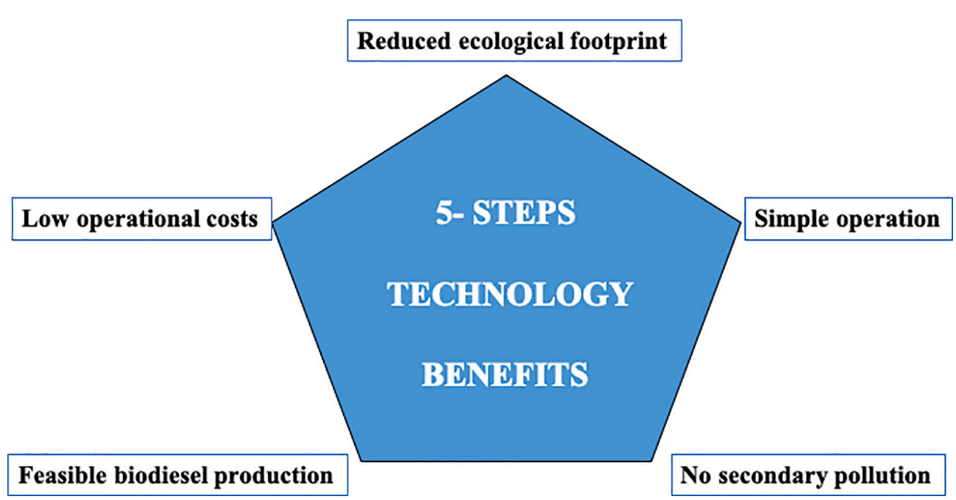

Fig. 12. Five benefits of the 5-steps technology.

carbon source for the cultivation of microalgae, reducing the total process costs. An analogous procedure described in the literature by Nguyen et al. [121] showed that Scenedesmus sp. was capable of concomitantly assimilating biogas CO2 and digestate as carbon sources. Although this strategy is interesting, it still has very small participation in the scope of the technologies. Another interesting aspect is that even with the rapid development and consolidation of the anaerobic digestion process of cattle residues in Europe, especially in Germany, as mentioned by Marcilhac et al. [135], the linking of this process with microalgae was not observed in the patents for these countries. Finally, one must emphasize that four products are obtained with this technology: biomass, methane, CO_2, water, and the latter can be destined for irrigation and virescence, thus contributing to the three nexus blocks, considering that the two effluents can be reused (i.e., digestate and wastewater), water consumption is reduced by reusing it, energy is produced as from the biogas, and it can be imagined that the biomass would not only be used in the scope of third-generation fuels, but also for other biotechnological ends, manufacturing products of greater value.

3.8. Keyword clustering and microalgae employed

Fig. 13 shows how the clusters related to the topic under discussion were organized.

As can be seen, some of the concepts are repeated in more than one agglomerate, which can corroborate the observation that patent documents are more focused on treating the effluents than on aggregating value to the resulting biomass. For example, the words regarding the treatment of effluents (e.g., wastewater treatment, 137; wastewater purification, 54; sewage treatment, 96; water treatment, 49) are predominantly about the production of bioproducts (biodiesel production, 39; biomass production, 39). Biodiesel is alone in the green agglomerate entitled centrifugation, where Shandong University is the greatest depositor with four patent families beside Enn Science & Technology with three. The word "bioreactor" has 44 patent families, of which Guangxi University appears as the main depositor. The word "microalgae" appears in most documents without specifying the genus, but in those that described it, *Chlorella* appears in 16% of them, followed by *Scenedesmus* (6.8%), *Microcystis* (1.4%), *Desmodesmus* (1.2%), *Botryococcus braunii* (0.9%), *Cladophora* (0.5%).

On a different line, one should highlight a point of convergence between that observed in Fig. 13 and the literature. As also reported by Garrido-Cardenas et al. (2018) [26] in the ambit of scientific publications, the microalga most used throughout the world for the MBBET, as demonstrated in the present study, was *Chlorella*. Five particularities can be listed for this microalga, as follows: (i) the *Chlorella* species adapt well to heterotrophic cultivation [136,137]; (ii) they have an appreciable capacity to accumulate lipids and produce lutein (carotenoids) as compared to the others [137–139], reaching values of 55% of accumulation [32,140]; (iii) they grow quickly [137,139]; (iv) their small cell diameter is beneficial for reducing the levels of xenobiotic compounds in wastewaters due to their high specific surface area [141]; and (v) these microalgae have been studied for a long time [137] and are already consolidated in the market, since the large-scale cultivation of *Chlorella* species started in 1961 in Japan by the company Nihon *Chlorella* Inc. [142,143]. Thus, in comparison to other microalgae, *Chlorella* species have been well studied, published, and patented for the development of technologies involving the integration expressed in the MBBET.

4. Future prospects and challenges

This technological prospecting was dedicated to mapping global technological trends in the targeted field; however, some gaps can be explored and complimented for future work considering two perspectives: tools for prospecting and technologies for MBBET. First, other techniques of text mining and visualizing approaches can be explored. For example, patent networks can be used to scrutinize the indicators of collaboration between institutions and applicants, as this will allow a deeper insight into the degree of interaction between the public and private sectors, as well as global partnerships for different types of technology. Moreover, tracking, and quantifying patents that are protected beyond their borders can be advantageous to obtain a sense of their economic relevance since the more countries a patent is extended, the greater the costs the applicant is willing to assume for the extension.

Concerning the second perspective, four points are relevant for the

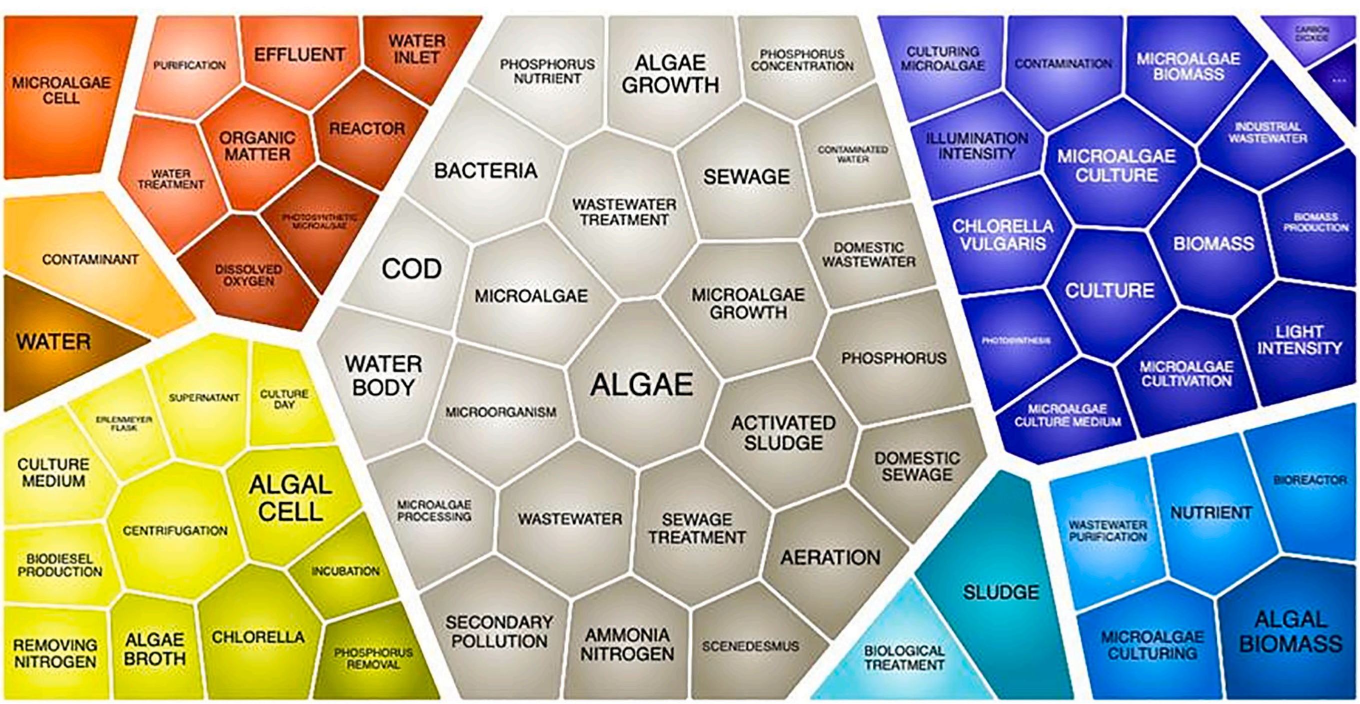

Fig. 13. Clusters of keywords related to patent documents involving biorefining and the biochemical treatment of effluents.
Source: Q. Orbit®.

future of microalgae production: dewatering, harvesting, medium cultivation, and the size of the plant, where the first and second points are the most capital-intensive steps [144,145]. This prospecting focused more on harvesting technologies and culture media due to the breadth of codes used; nevertheless, a discussion of bioreactor patents and their nuances is also a future topic on which Guaxi University appears to excel. Another point worth mentioning is that similar to what was perceived by Yu et al. [146] in the context of scientific research, relatively few technologies have been developed to date with strains other than *Chlorella* and *Scenedesmus*.

Three challenges related to microalgae bioproducts were demonstrated as future notes to be evaluated: bioethanol, bioplastics, and metallic nanoparticles. Despite its important role in energy transition and for the insertion of the paradigm of the water-food-energy nexus, bioethanol from microalgae is still a challenge that is not supported by technologies. In the same line, algae bioplastics are promising for having similar mechanical properties to conventional petroleum plastics, but they are also a challenge that has not yet been found in technologies. Finally, metallic nanoparticles from microalgal biosynthesis are also a potential challenge for future nanobiotechnology and, consequently, for MBBET.

5. Conclusions

This study evaluated patents spanning over the last 20 years through a hybrid method, showing that patent databases are useful for acquiring strategic information. To the best of our knowledge, this is the first attempt to propose a state-of-the-art analysis encompassing microalgae-based biorefinery coupled with biochemical effluent treatments (MBBET). The compilation of technological trends is relevant for decision-makers in research and development (R&D) management because an understanding of the current scenario is useful for providing a better forecast of technological changes.

Since 2011, there has been a growing interest and global race to foster R&D in MBBET. Therefore, this sector presents many opportunities for companies in the coming years, and the technological prospects compiled in the current analysis can assist these firms. It is expected that companies and research institutes will assume a greater role in the development of technologies, despite the relevant part that universities have played in the innovation ecosystem. From the technological profile of these three key players, different market purposes can be observed since there is a trend of institutes and companies turning to patents for developing devices and methods for algal biomass valorization.

This review also illustrated that MBBET is a promising method for mitigating global environmental challenges. It stems from the new paradigm of the water-food-energy nexus since it consists of a synergistic system these three elements as evidenced by the applications in bioadsorbent technologies, animal supplementation, and bioenergy. This systemic approach is the essence of an emerging circular bioeconomy and should be considered by policymakers as well as R&D managers.

The evidence for the biotechnological potential of microalgal biomass is promising for conducting future studies in this area, despite being in its infancy. China was found to be the pioneer, issuing 65% of the world's patents and demonstrating the potential of generating high-value products from bioremediation of sewage and industrial effluents. The technological contributions from China, as well as South Korea's practice of strengthening the ties between private and public initiatives by creating visionary projects in the biotechnology field at a national level, can be beneficially replicated in the West. Notwithstanding the predominant and emerging technologies enclosed in this review, many important technology gaps still exist which are challenges for the advancement of microalgal research and applications.

CRediT authorship contribution statement

Luiggi Cavalcanti Pessôa: Conceptualization, Investigation, Software, Writing – Original Draft, Data Curation, Visualization, Editing final manuscript. **Kricelle Mosquera Deamici**: Writing, Review, Editing final manuscript. **Luiz Antônio Magalhães Pontes**: Supervision. **Janice Izabel Druzian**: Project Administration, Funding acquisition. **Denilson de Jesus Assis**: Methodology, Resources, Writing – Review & Editing, Supervision.

Declaration of competing interest

None.

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