



Review

Global trends and characteristics of nano- and micro-bubbles research in environmental engineering over the past two decades: A scientometric analysis

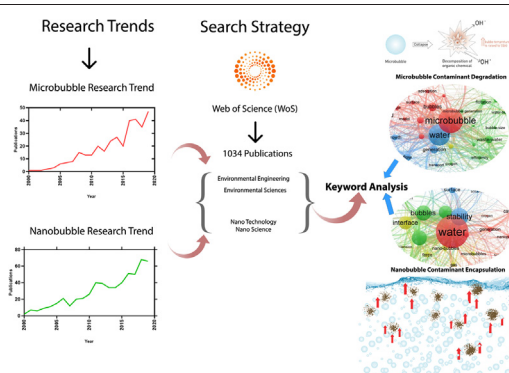
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HIGHLIGHTS

- Environmental sciences and chemistry were the dominant categories for microbubbles.
- Nanotechnology and physical chemistry were dominant for nanobubbles.
- Half of the most productive countries were among the developed and high-income countries.
- Keyword analysis revealed “water treatment” as the focus of microbubbles.
- Nanobubbles are still being investigated for their unique characteristics such as stability.

GRAPHICAL ABSTRACT



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ABSTRACT

The present study has two primary goals, the first goal is to investigate a bibliometric analysis and assess the trends to evaluate the global scientific production of microbubbles and nanobubbles from 2000 to 2020. The aim is to elucidate the cornucopia of benefits the two technologies (micro and nanobubbles) can offer in environmental sciences and environmental amelioration such as wastewater treatment, seed germination, separation processes, etc. The second goal is to explicate the reason behind every chart and trend through environmental engineering perspectives, which can confer value to each analysis. The data was acquired from the Web of Science and was delineated by VOS viewer software and GraphPad Prism. Considering 1034 publications in the area of micro- and nanobubbles, this study was conducted on four major aspects, including publication growth trend, countries contribution assessment, categories, journals and productivity, and keywords co-occurrence network analysis. This article revealed a notable growth in microbubbles and nanobubbles-related publications and a general growth trend in published articles in a 20-year period. China had the most significant collaboration with other countries, followed by the USA and Japan. The most dominant categories for microbubbles were environmental sciences and environmental engineering comprising 22.5% of the total publications, while multidisciplinary subjects such as nanotechnology and nanosciences (8%) were among the dominant categories for nanobubbles. Keyword's analysis results showed that microbubbles had reached the apex since their discovery. Consequently, they are being used mostly in water/wastewater treatment or environmental improvement. On the other hand, nanobubbles are still in their infancy, and their pervasive use is yet to be fully materialized. Most of the publications are still striving to understand the nature of nanobubbles and their stability; however, a critical analysis showed that during the past two years, the trend of using nanobubbles as a cost-effective and environmentally friendly approach has already begun.

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1. Introduction

The availability of water resources in densely populated countries across the globe is a severe issue. In this regard, some of the cities that have made international headlines due to water shortage are Cape Town in South Africa, Bangalore in India, Jakarta in Indonesia, and Sao Paulo in Brazil (Oki and Quiocho, 2020). Limited land area to reserve rainfall exacerbates the problem since a large amount of water is squandered as it cannot be captured or retained (Tortajada, 2006; Wen et al., 2011). Unbridled urbanization, anthropogenic activities, and the perpetual increase in the world's population have strained water resources, thereby necessitating improved discharge quality (Drangert and Sharatchandra, 2017; Fan et al., 2019). If not adequately treated, wastewater can be detrimental to aquatic and terrestrial ecosystems' health (Lee et al., 2019; Li et al., 2019; Wan et al., 2020; Xu et al., 2020). In terms of quantity, municipal secondary effluent is large and has the potential to be used as a sustainable source for reuse (Fan et al., 2019; Larsen et al., 2016; Sun et al., 2016).

According to a recent report, in 2018, the Chemical Abstract Service listed 143 million chemical products (American Chemical Society, 2020; Azuma et al., 2019) which will inevitably find their way into wastewater treatment plants to be treated for reuse (Tiedeken et al., 2017; Verlicchi et al., 2010). Emerging contaminants (ECs), which are also known as emerging organic contaminants or contaminants of emerging concerns (CECs) (Bilal et al., 2019; Castillo-Zacarias et al., 2021), represent a group of natural and synthetic chemicals in water bodies that are able to adversely affect ecosystems and human health (Geissen et al., 2015). The list of compounds and chemicals in this group is significantly large and getting perpetually larger by incorporating new commercial substances and disposal of chemicals currently in widespread use. Moreover, with the state-of-the-art techniques to analyze the water and detect low concentrations of emerging contaminants, new ECs have been recently identified (Gasperi et al., 2014; Rodríguez-Narvaez et al., 2017). Some major categories of ECs include pesticides, pharmaceuticals, and personal care products (PPCPs), disinfection by-products, industrial chemicals, endocrine disrupting compounds (EDCs), artificial sweeteners and food additives, nanomaterials, sunscreen, and UV filters, flame retardants, and Perfluorooctanoic Acid (PFOA) (NORMAN, 2020; Richardson and Ternes, 2014). These chemicals can negatively impact various water resources (Alygizakis et al., 2016; Azuma et al., 2019; López-Serna et al., 2013). Therefore, urgent actions are needed to analyze and remediate such compounds in water bodies as potable water is becoming scarce across various parts of the globe. Even though conventional methods are proven to be

effective, the concept of "biodegradability" constrains the treatment of certain types of wastewater, making it an arduous task to achieve standard discharge quality. Consequently, wastewater treatment plants require sustainable technologies (Jabesa and Ghosh, 2016). In this regard, micro and nano-bubbles possess the capacity to solve this issue and, therefore, are attracting a lot of attention owing to their possible applications in different sectors (Agarwal et al., 2011; Gurung et al., 2016).

Micro and nanobubbles are air bubbles with dimensions of 10–15 μm and less than 200 nm, respectively. These bubbles possess unique characteristics that make them efficient in water treatment. Unlike macro bubbles that reach the surface with a high velocity and then burst, microbubbles rise with much lower velocity, and they burst midway. Upon bursting, ozonated microbubbles release hydroxyl radicals, reacting with the contaminants and degrading them (Yasui et al., 2019b). At the final moment of the violent bubble collapse, the bubble temperature increases significantly to 7200 K, even though the endothermic heat of O_3 considerably cools the bubble. Most oxygen (O) atoms are immediately consumed inside a bubble by the chemical reaction with water vapor $\text{H}_2\text{O} + \text{O} \rightarrow 2\text{OH}$. In addition, more than 10^7 OH (hydroxyl) radicals are produced inside the bubble, as mentioned in the above chemical reaction (Temesgen et al., 2017a), rendering them ideal for disinfection and contaminant degradation. Fig. 1 elucidates the process of bubble burst and radical generation.

Nanobubbles can stay afloat in the water's body for hours to several days if generated correctly. Nanobubbles have three unique properties aside from high durability; first, they have Brownian motion, which means they can cover a larger surface area when rising to the surface or remaining in the water (Sekiguchi et al., 2011); as a result, they are capable of bringing back suspended solids or oil droplets (fat particles) to the surface more efficiently. Second, they possess a very large surface area; if a typical bubble has a 0.1 mm to 0.24 m^2 surface area ratio, nanobubbles have 0.24 nm to 240 m^2 , which is approximately 1000 times more surface per milliliter. This results in higher mass transfer, and third, in the presence of surface-active agents (surfactants), nanobubbles can acquire electrostatic charge (various charges based on the type of surfactant). Consequently, nanobubbles can surround contaminants, similar to forming a micelle (Attard, 2003), providing a unique opportunity to remove contaminants from the body of the water.

Though nano and microbubbles' technology has long been investigated, there is a lacuna of knowledge, especially when it comes to the application in water treatment. Only a few literature reviews have considered nanobubbles and their utilization in water treatment or other

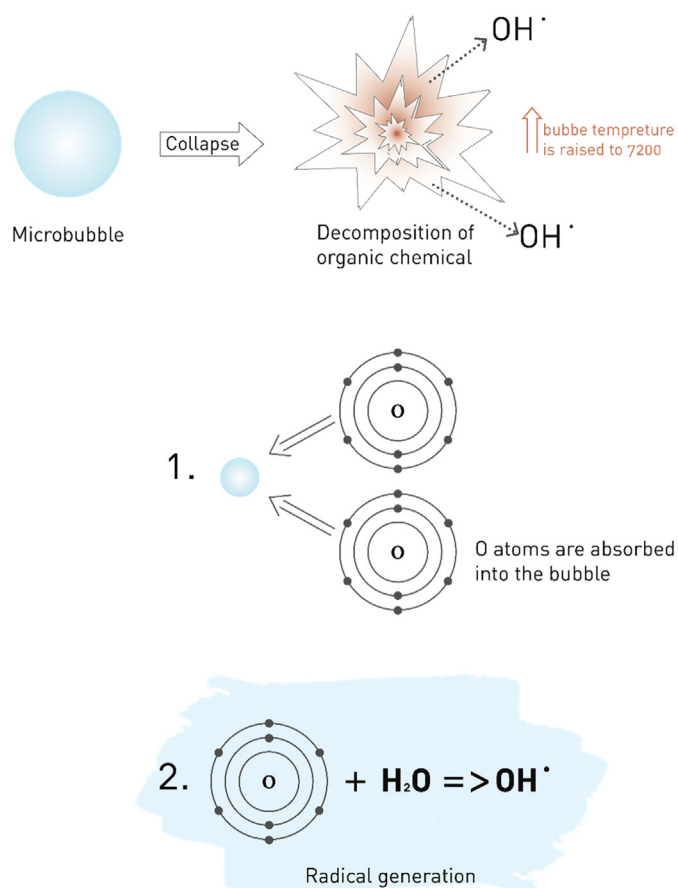


Fig. 1. Microbubble burst and OH radical generation process.

environmental improvements. Based on the Web of Science database (Clarivate, 2020), in the last four decades (from 1978 to 2020), only 17 review papers have been published on nanobubbles, and among those, only two papers assessed nanobubbles role in water treatment (Atkinson et al., 2019; Temesgen et al., 2017b). This, indeed, calls for both review and bibliographic publications that researchers can use to have a better perspective toward nano and microbubbles. Consequently, it is difficult for scholars and researchers to read all the publications in this field. Moreover, to the best of the authors' knowledge, no bibliometric research about nano and microbubbles has been done over the past 42 years. In this study, the authors have strived to provide a general overview of nano and microbubbles research trends to assess their characteristics and their utilization in environmental remediation.

The overarching aim of this study is to conduct a scientometric analysis of nano- and microbubble research from 2000 to 2020, considering the increasing attention given to this topic. Multiple key areas were assessed, including keywords, authors, countries, major journals, number of publications, citation, and keywords co-occurrence network. In order to visualize the results, social network analysis (SNA) was employed (Macías-Quiroga et al., 2020; Zhuang et al., 2013).

2. Methodological approach

A three-tiered approach was adopted in this exercise. The first being the bibliometric analysis involving database and software, keywords associated with the subject matter, year of publication, and data visualization. These approaches are described in detail below.

2.1. The bibliometric method

The bibliometric analysis in this study is the result of the consecutive assessment: (1) opting for the most reliable database and software (2) selecting the keywords, commensurate with the topics (in this case both nano and microbubbles) (3) applying limitations to the search results (year range, document type, etc.) (4) exporting the data via the Web of Science (WoS) exporter to literature tools for further analysis (5) data visualization using social network analysis (SNA).

The web of science core collection was employed to extract the data. Given the number of published studies related to nano and microbubbles was limited (specifically their utilization in environmental improvements and/or water/wastewater treatment), the year range was selected to be 2000 to 2020. The web of science is among the most powerful and reliable scientific publications' repositories (Merigo et al., 2017; Yang et al., 2013). Furthermore, WoS features smart tools to analyze the data in various forms, thus facilitating the process of eliminating the extraneous data and refining the results. It is also compatible with multiple literature analysis tools (Newell and Cousins, 2015).

2.2. Social network analysis

Employing graph theory, social network analysis (SNA) is used to investigate the dataset's social structures (Otte and Rousseau, 2002). To visualize data and bibliometric network, "visualization of similarities" (VOS) was selected. The interconnected nodes in the network could comprise journals, publications, and various studies. Moreover, it can be established upon citations, co-occurrence, co-authorship, countries, to mention but a few. In this study, SNA was employed to establish the connection between countries, authors, and keywords cluster-analysis via VOS viewer (version 1.6.15). It should be noted that there are other methods to analyze the bibliometric data; Canoco is also a software for multivariate data analysis (ter Braak and Šmilauer, 2002).

2.3. Data collection

The literature data was collected and refined on October 12, 2020, from the Web of Science. Two separate searches were conducted to assess two topics simultaneously. First, "nanobubble" was selected to be searched upon in titles, keywords, or abstracts. Then "water" was added with the "AND" function, to be combined with "nanobubbles." The reason behind this decision lies in the fact that a very large portion of nanobubble-related studies -to be more specific, older studies- are concerned with nanobubble structures, characteristics, and their formation. It is of utmost importance to note that nano and microbubbles are relatively new topics. In this regard, utilization of nanobubbles in water or wastewater treatment, floatation, purification, horticulture, and other environmental enhancements appeared mostly alongside the keyword "water." Consequently, the authors decided that it was the most prudent choice to combine these two keywords, even at the expense of decreasing the number of publications.

Given there are a large number of unrelated publications (not related to wastewater treatment), citing those publications would have baffled the readers who are likely to use this article as the benchmark of their studies. It should again be noted that there are no bibliometric reviews available to the best of the authors' knowledge. Thus, we envisage that the current study will be conducive to a concise, right-to-the-point review concerned with modern uses of nano and microbubbles.

The same approach was applied to microbubbles, and even though microbubbles are relatively an older subject than nanobubbles, for both cases, the "search period" was set to 2000 to 2020 (20-year period). A total of 1038 publications (406 for nanobubbles and 687 for microbubbles) whose titles, keywords, or abstracts, simultaneously contained either "nanobubble" and "water" or "microbubble" and "water." To answer the question regarding the difference between inclusion and exclusion of the keyword "water," it is observed that by

excluding this keyword from the search results, the number of publications for nanobubbles increases to 741 at the expense of accuracy since a large portion of these newly included papers is concerned with biology and medicine. Under no circumstances related to “environmental engineering,” therefore, it is ostensibly a wise choice to eliminate these results by including the keyword “water” to ensure that only environmentally related papers are being investigated. Additionally, the same case applies to microbubbles.

Another exquisite point that deserves some words here is that even after excluding the keyword “water,” the total number of publications is only a handful (741), considering the long period of 20 years. This and the fact that many publications have been published in recent years show that these topics -nano and microbubbles and their use in the environment- is exceptionally new and yet to be explored. Consequently, composing a bibliometric review that can incorporate all the necessary data is a prerequisite for future research in this field.

Overall, as shown in Fig. 2, the documents consist of articles 91.1% and 90.3% for nano and microbubbles respectively, review papers (4.2%, 3.5%), proceedings papers (2.5%, 4.5%), early access (1.5%, 1.6%). English was the language used in almost all the papers regarding nanobubbles; Chinese and Japanese were equally used in only four articles (two papers for Chinese and two Japanese papers). The case was somewhat more diverse for microbubbles, as was expected since this is an older topic of interest. Its utilization has been more broadly discussed in many countries, with 637 papers in English, seven papers in Japanese, 2 Chinese, 1 French, 1 German, and 1 Portuguese.

In the current work, Microsoft Excel 2020 was employed to thoroughly analyze the data based on document type, language, title, country, journal, and keywords. Additionally, Mendeley (both Desktop application and Google Chrome add-on) was used to manage references.

3. Results and discussion

3.1. Publication's growth trends analysis

As shown in Fig. 3, the annual number of publications in nanobubble and microbubble cases has increased from 2000 to 2020. The annual number of citations (TC) showed a similar increasing trend with a peak in 2019, indicating a significant amount of attention to the nanobubble discussion; the number of citations decreased in 2020. This decrease in the total number of citations is reasonable because the newly published documents have not been cited pervasively. The same trend can be observed for microbubbles (Fig. 3b); overall, the total number of publications drastically increased, reaching the most significant number of publications in 2020 (77 publications in one year). The TC pattern is almost precisely similar to that of nanobubbles (Fig. 3a), as many 2020 papers have not been cited, resulting in a TC

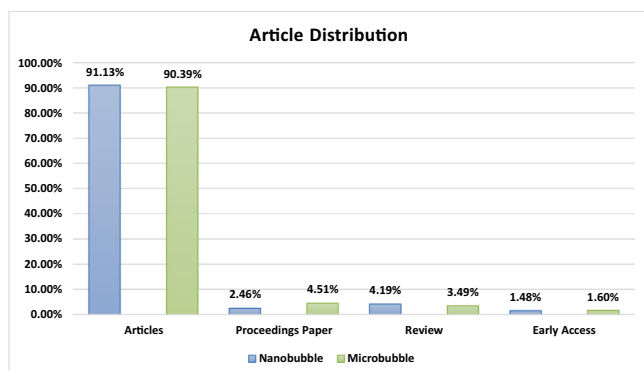


Fig. 2. Global distribution of articles for nano and microbubbles in a 20-year period.

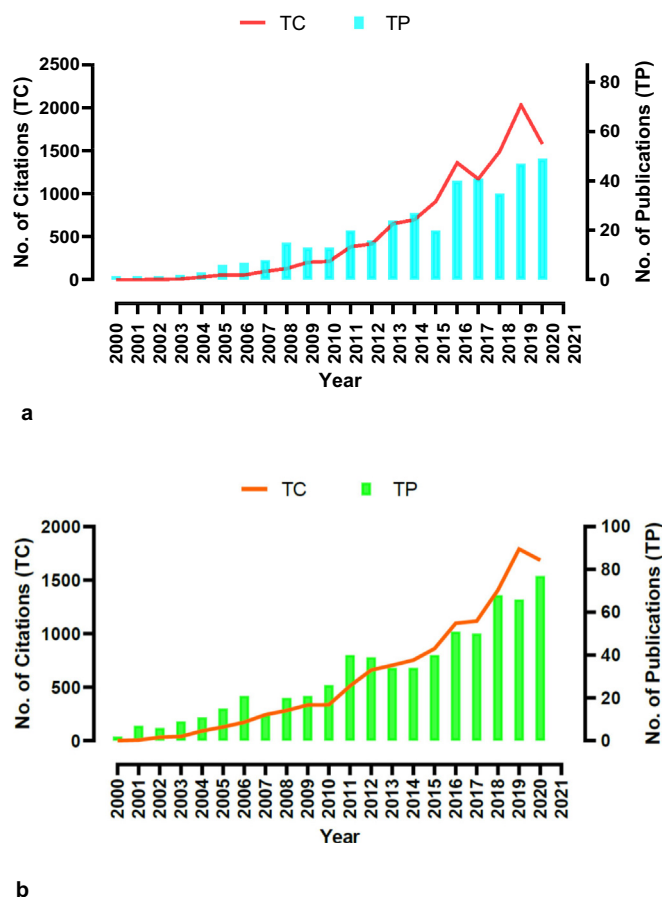


Fig. 3. a Annual number of publications for nanobubbles. b Annual number of publications for microbubbles.

decrease. By comparing these two charts, it is evident that between the years 2000 to 2007, the number of microbubble publications was greater than that of nanobubbles. It is appropriate that the reason for the anomaly observed above need a closer look and the following section discusses this from an environmental point of view.

Microbubbles are a relatively older technology that has been being used primarily in mineral separation and purification technologies in order to float the impurities, removing them, and reaching the refined minerals (Wang et al., 2020b). In the following years, as the other unique aspects of microbubbles were discovered, such as the released shockwaves upon burst, the researchers across the globe began paying attention to their use in environmental remediation, such as removing contaminants (ozonated microbubbles), increasing mass transfer in conveying more oxygen to plants (Zhou et al., 2019) to mention but a few. Therefore, the difference in the number of publications in the early years is commensurate with the inherent nature of these two technologies.

3.2. Subject categories

Nano and microbubbles have various environmental, physical, chemical, and technological aspects. Their pervasive use in different fields turns this area into a multidisciplinary subject. The subject area of papers was determined using the filtering tool provided by Web of Science. This toolbox enables the scholar to precisely indicate the subject area of each paper. That is, by “inclusion” or “exclusion” of certain areas, the showed results alter accordingly. Hence the selected papers have minimum overlap. In this regard, if “environmental science” is selected, the resulted papers are mainly concerned with “environmental

science," which will be the dominant aspect of the papers. To ensure the results' credibility, the authors have rigorously examined the papers in each category. To make sure they do not possess any overlaps. In the final selection, the papers that were present in more than one category were eliminated.

As expected, selected publications -for nanobubbles- incorporate as many as 50 different fields; among them, ten disciplines are suitable for this study as they are related to environmental aspects of bubbles either directly or indirectly. It is evident from Fig. 4 that physical chemistry is dominant and covers topics such as nanobubble stability and generation methods. Moreover, 26.3% of the total publications are related to this category. The next major category is multidisciplinary material science, with a prodigious co-occurrence with the former category (19%). One of the most important categories for this review is, however, nanotechnology and nanoscience. This category comprises various nanobubble usage, ranging from membrane defouling to their positive effect on seed germination, and it takes up to 8% of total publications. It is noteworthy to discuss the environmental sciences publications. This category has a share of 2.6% of total publications, and after in-depth scrutiny, 95% of environmental sciences publications were in 2020. This proves an exhilarating fact that nanobubble use in the environment is still being improved and shows its imminent importance in the following years. To further buttress this notion, older publications are, in fact, in the physical chemistry section as they are mainly concerned with bubbles' nature and generation. The authors predict that shortly this trend will reverse (as it has already begun to do so), and the number of publications in environmental sciences will pace to other fields.

Regarding microbubbles, chemical engineering is the dominant section that mainly includes microbubble characteristics, floatation improvement, microbubble generation methods such as venturi tubes, to name but a few (Fig. 5). In a critical approach, publications in two fields of "environmental sciences" and "environmental engineering" combined will take up to 22.5% (12.8% + 9.7%), which is almost equal to physical chemistry publications. This analysis is of utmost importance because it elucidates the trend in which microbubbles' environmental use is attracting more attention. Moreover, among 123 publications in these two fields, 50 publications were published very recently (2019 and 2020), and the same trend was observed in nanobubbles. The essential idea is that since microbubbles are a relatively older technology, it has matured and enjoys environmental sciences and engineering growth.

3.3. Country analysis

Fig. 6 demonstrates the top 20 productive countries within the dataset for microbubbles; China, accounting for 149 publications (23.1%), and Japan, with 147 (22.8%), are the two most productive countries. The third place is the USA with a total number of 130 publications (20.15%), followed by England, South Korea, France, Canada, Australia, Germany, India, Taiwan, Singapore, Italy, Russia, Netherlands, Brazil,

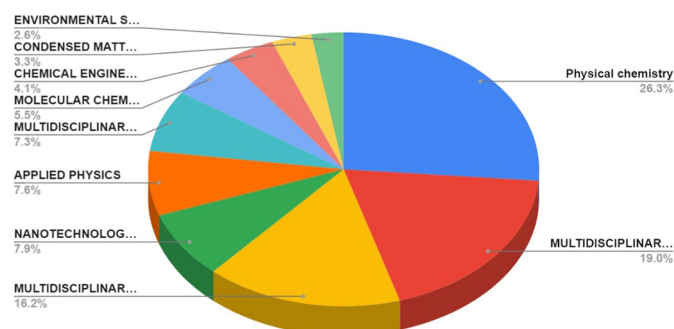


Fig. 4. Top ten of most prominent categories for nanobubbles.

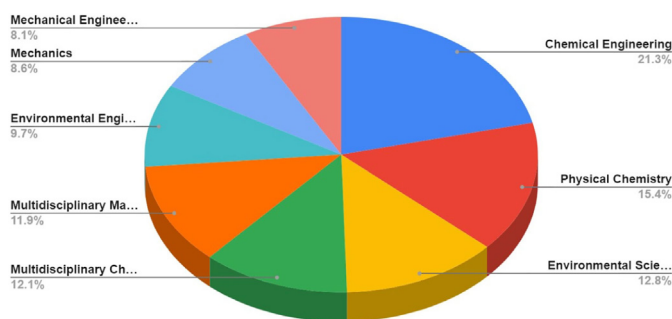


Fig. 5. Top nine of most prominent categories for microbubbles.

Mexico, Iran, Malaysia, and Switzerland. Considering the list, 10 out of 20 countries with the highest contribution are among developed and high-income countries. Employing a critical approach, the authors decided to take a closer look into the nature of publications of the two first countries (China and Japan). It was observed that most of the publications were concerned with utilizing microbubbles in either environmental or specifically water/wastewater remediation.

As most of the countries mentioned above are developed or have a huge population, environmental pollution is considered a major concern for them, and since some of them are technologically advanced, these countries can afford the generation and measurement of micro and nanobubbles (micro and nanobubble generation requires sophisticated apparatuses, especially nanobubbles). Meanwhile, the rest of them (developing countries) such as Iran, Brazil, Mexico cannot properly cope with the wastewater problem. The budget deficit of these countries does not allow them to implement new and novel technologies into their conventional wastewater treatment plants. In Iran, for example, according to the "Financial Tribune" website, Iran's fiscal budget for 2021–2022 has a deficit of 9–10 billion dollars, exacerbated by the COVID-19 pandemic. The perpetual budget deficit over the years has hindered the required investment in exploring new methods such as micro and nanobubbles and their utilization in wastewater treatment.

Consequently, wastewater treatment infrastructures have been the same for decades suffering from a lack of improvement (Behboodian and Shirdelian, 2021; Tabesh et al., 2019). The same case is, to some extent, applicable to other developing countries. According to (Pandey et al., 2021), the detection of SARS-CoV-2 (as fecal contamination) in wastewater during COVID-19 is a severe issue in developing countries, lacking the proper wastewater treatment measures. However, developing a simple ozone-generated microbubble system with highly reactive bubbles can alleviate the problem of COVID-19 transition via aquatic media. Thus, the problem with developing countries, aside from the budget, can be ascribed to lack of management as well.

Micro and nanobubbles are very efficient and convenient methods for various aspects of the environment, and therefore, are being used pervasively. For example, China is a vanguard using bubbles (micro and nano) in various fields ranging from environmental sciences to material science; in 2020, China was one of the first countries to have used micro-nanobubble aeration with regards to eutrophication of urban landscape water (Wang et al., 2020a). China is the first country with 113 publications (29.05%) (Fig. 7). Japan, however, has been replaced by the USA, with 80 publications (20.56%). The third country with the most contribution is Japan, with 62 publications or ~16%, followed by Netherlands, Australia, Germany, Russia, England, South Korea, France, Taiwan, Singapore, Belgium, Czech Republic, Iran, Sweden, Canada, India, Denmark, and Israel. It is noteworthy to understand that the same developed and high-income countries that have been active in using microbubbles and publishing about them are the same pioneers in the nanobubble field.

In Figs. 6 and 7, co-authorship network analysis among the top 20 productive countries is illustrated to better understand the global

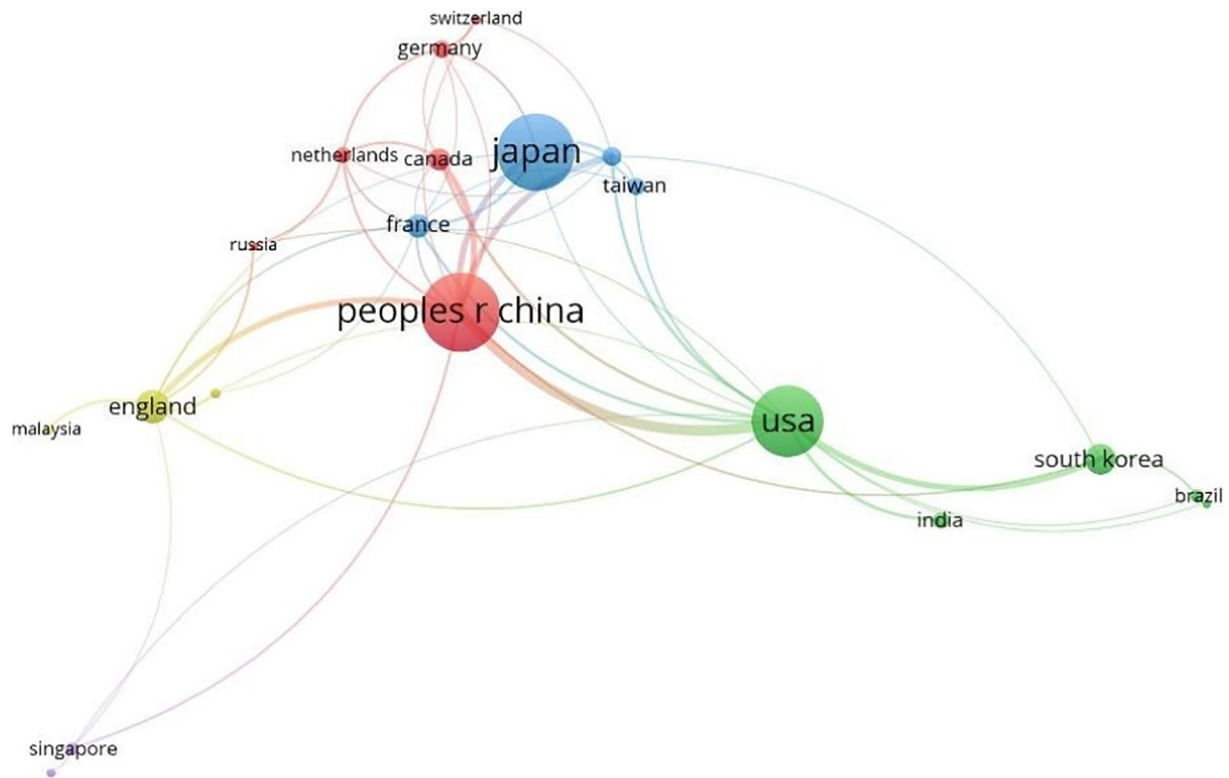


Fig. 6. Collaboration of countries on microbubble based on the co-authorship relations.

research status and trends. Considering the clusters of Fig. 6, it is evident that China and Japan's total publications are greater than other countries, and the link strength of them is 46 and 18, respectively. While the USA has relatively fewer publications than Japan, its total link strength is 40. In both cases (nano and microbubble publications), China has the greatest link strength, and a plausible reason could be attributable to China's student export to various overseas countries. Considering the clusters in Fig. 7, China and the USA have both the highest number of publications and total link strength of 66 and 46, respectively. In both cases, the USA has the second link strength. Therefore,

we speculate that the USA is in the perpetual search of new and advanced technologies, which could be attributed to the total economic growth that paves the way for more advanced and simple-to-use technologies such as nano and microbubbles. Therefore, it is conceivable why the USA is heavily investing in this area of research. According to the Federal Research and Development (R&D) Funding in 2021 with the amount of 142.2 billion dollars ([Congressional Research Service, 2020](#)), it is expected for high-income countries such as the USA to pursue state-of-the-art technologies such as bubbles and thus publish more papers in this area.

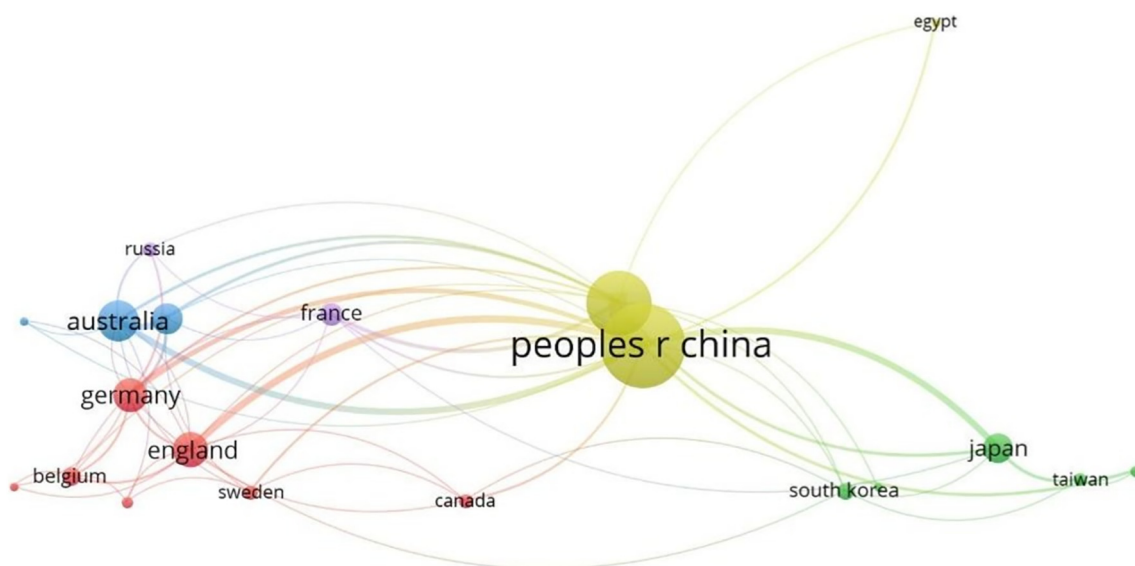


Fig. 7. Collaboration of countries on nanobubble based on the co-authorship relations.

In order to critically look into the nature of publications, the case for the USA was investigated, approximately 80% of USA publications regarding nanobubbles are either about understanding their formation and stability (as their stability has become a controversial issue, due to the fact that the Laplace pressure inside the bubbles is so high that it should collapse in a matter of seconds) (Boshenyatov et al., 2019; Kim et al., 2019) or are concerned with their use in water improvement and treatment. It was interesting to observe the same trend in China's publications as they were mostly about nanobubbles formation and stability and their use in environmental engineering, such as membrane defouling or microbial activity (Chen et al., 2020; Xiao and Xu, 2020; Xiao et al., 2020). However, in developing countries, the nanobubble is still in its infancy, and most publications are focused on floatation techniques and nanobubble use in mineralogy (Nazari et al., 2019; Pourkarimi et al., 2018, 2017). This implies that micro and nanobubble utilization in developing countries with low-income is mainly due to lack of technology and devices and a dearth of monetary support and therefore not yet fully developed.

Figs. 8 and 9 delineate the total number of publications from 2000 to 2020 for each country, explained in the last paragraphs. In Fig. 11, the six most productive countries' annual publications in microbubble are depicted. The pioneer in article publication, overall, was China. However, prior to 2016, Japan surpassed China by far. It is interesting to notice the same trend for the USA. It is evident that Japan, after 2007, went ahead of the USA as well, even though the trend was in reverse before 2007. The reason behind the proliferation of China was explained earlier. This caused the authors to look for the stimuli behind Japan's proliferation in the years 2010 to 2013. It was observed that a total number of 14 papers were on the topics of environmental sciences, environmental engineering, and water resources; incorporating issues such as removing pesticides from water, using microbubbles for separation of oil in water-oil systems, or the removal of other contaminants in water (Ikeura et al., 2011; Sekiguchi et al., 2010; Van Le et al., 2012). Since employing microbubbles in environmental engineering and water resources was a perspective that other countries opened their eyes to later (China, USA, etc.), this data analysis yields a fundamental understanding that Japan reached maturity in microbubble use earlier than other countries, and it was only later that China started to pace Japan (2017 to 2020) in this regard. For the USA, aside from the fluctuations, a general increasing trend may continue to grow, but it is improbable to surpass China. It is also important to note that almost all other countries have an increasing trend in microbubble publications, thus suggesting the field of microbubble is yet to be fully explored. Among the six most prolific countries, no developing countries are observed (India, Iran, etc.). This indicates that micro and nanobubbles (illustrated in Fig. 11) are technologies that cannot be utilized without sophisticated generation and measurement methods.

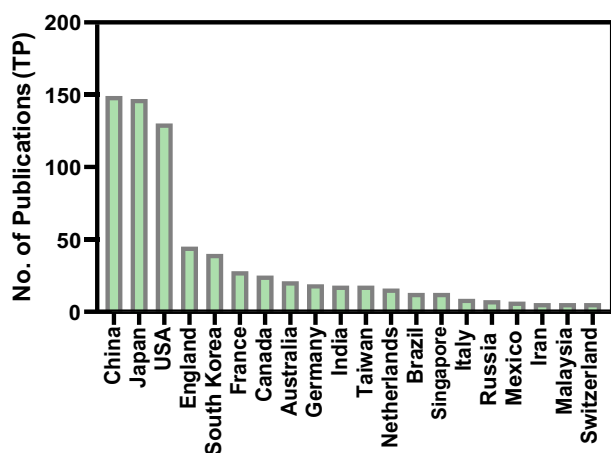


Fig. 8. Top 20 productive countries from 2000 to 2020 (Microbubble).

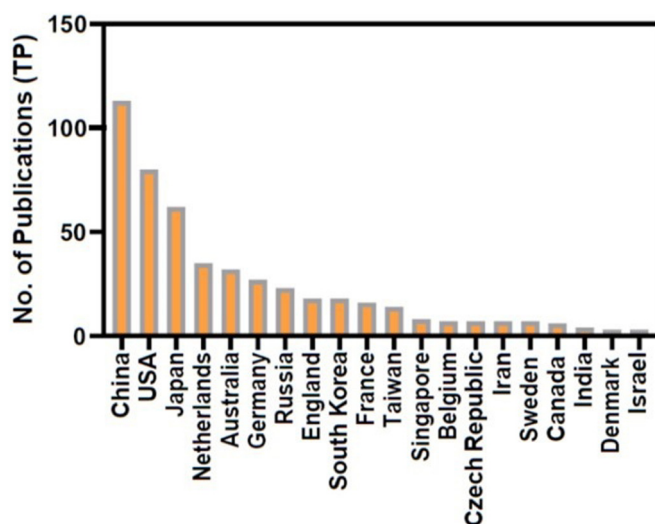


Fig. 9. Top 20 productive countries from 2000 to 2020 (Nanobubble).

In Fig. 12, the total annual publications of nanobubbles are delineated. As was expected, due to the relative novelty of this concept, before 2006, almost all the major countries have a negligible number of publications. In 2006, China became the most productive country; however, most of the publications were on the subject of "surface nanobubbles" formed on a hydrophobic solid-water interface (Wu et al., 2006; Zhang et al., 2006). Since many research papers in the nanobubble area are concerned with nanobubbles on the surface, and as they are inherently different from "bulk nanobubbles," it is important to understand the spontaneous nucleation of surface nanobubbles. To be more specific, the nucleation process is explained using the Gibbs energy. The relationship between the Gibbs energy graph and surface nanobubbles radius shows the possibility of spontaneous generation of these nanostructures (Koshoridze and Levin, 2020). The process of nucleation is depicted in Fig. 10. The authors have redrawn the graph to present a simpler view for the readers and maintain this work's originality. G_{Hom} showed with the blue line is the homogeneous Gibbs energy that reaches its maximum when the nucleation process is completed. Further in the process, as both the homogeneous and heterogeneous Gibbs energy (showed by the green line (G_{Het})) decrease the radius of the nuclei increases, thus producing the bubble.

It was only during the past few years that the significance of "bulk nanobubbles" has been found out. Consequently, many recent publications or on the topic of bulk nanobubbles and their use in environmental engineering and their stability started to be investigated, the same trend that was observed at the beginning of "surface nanobubbles" discovery (Koshoridze, 2020). Between 2006 and 2011, Netherlands enjoyed a steady growth in the number of publications; a more in-depth look into the nature of these publications showed that even in 2015, most of the publications were again concerned with "surface nanobubbles" (Lohse and Zhang, 2015a, 2015b). From 2015 to 2020, Japan and the USA had a close race until, in 2020, Japan published more articles compared to the USA.

However, after 2015, the growth in the number of publications on the nanobubbles topic was considerably drastic and sharp. This is because only recently are scientists acquiring the necessary knowledge to understand nanobubbles' various environmental applications. For example, their use in horticulture or even for soil remediation purposes has recently been documented. Moreover, it is essential to note that recent research combining nano and microbubble systems (MNBs) is worth investigating as their simultaneous use can be very productive. To be more specific, (Sung et al., 2017) used micro-nano-bubbles to remove trichloroethene from water. Elsewhere, (Aluthgun Hewage et al., 2020) recently employed ozonated nanobubbles to remediate

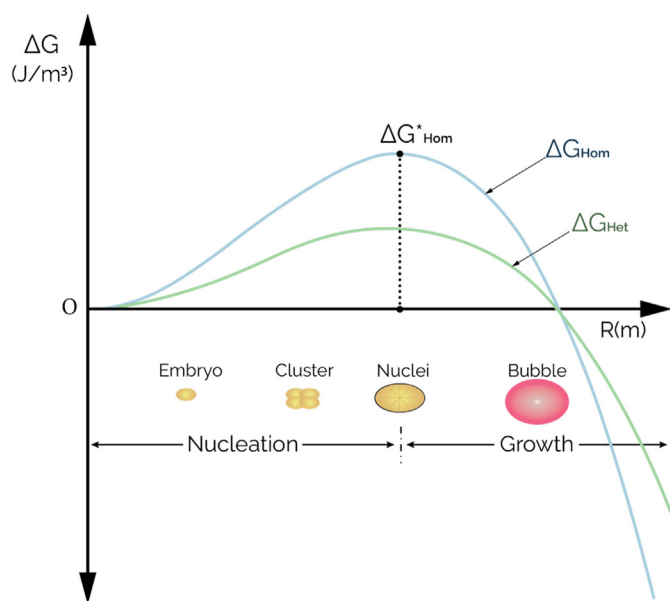


Fig. 10. Bubble's radius and its relationship with Gibbs energy during the nucleation process.

contaminated sediments. The authors used an in-situ remediation method with a combination of low-frequency ultrasound and ozone nanobubbles. The employment of ultrasound enables the soil to go into a suspension state, resulting in the desorption of contaminants. The desorbed contaminants are then oxidized via the introduction of ozone nanobubbles that possess a higher mass transfer efficiency and retention time (compared with normal bubbles).

Therefore, it is envisaged that following the current trend and pervasive use of nanobubbles in a plethora of environmental and non-environmental fields, the number of scholarly attempts will increase significantly and that various combined systems such as MNBs, oxygen nanobubbles, ozonated nanobubbles, etc., will become dominant topics in this regard.

3.4. Journal analysis

The total of 645 publications of microbubbles resulted from approximately 100 journals, among which only 27 journals had more than five publications during the last 20 years, accounting for nearly 30% of the total publications. The impact factor (IF) is one of the most reliable indices to evaluate research quality. Researchers and the authors used it to opt for a specific journal (Suiter and Sarli, 2019). On the other hand, quartile rankings (QRs), in this article provided by WoS citation report,

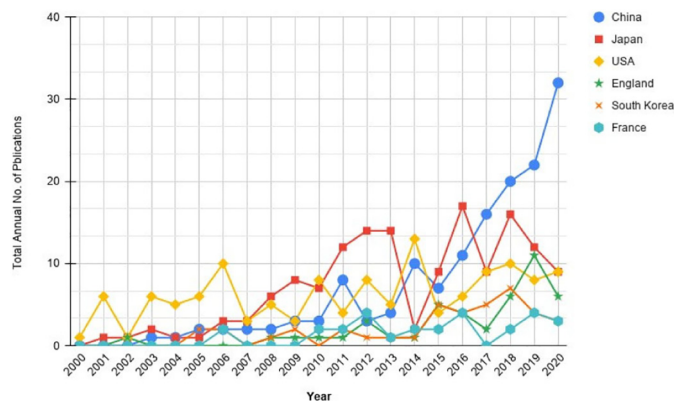


Fig. 11. The annual trends of total publications of the top 6 most productive countries on microbubbles.

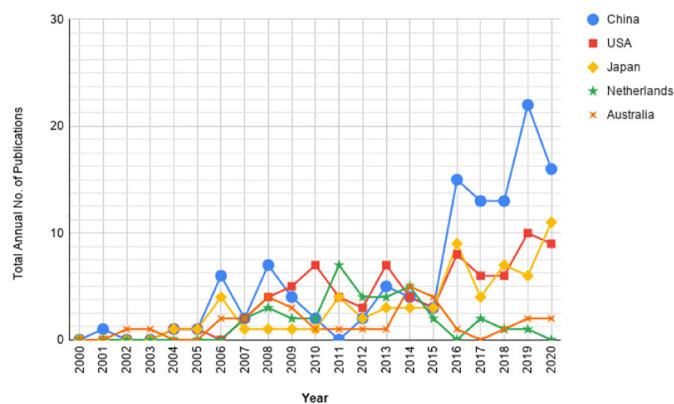


Fig. 12. The annual trends of total publications of the top 5 most productive countries on nanobubbles.

is another critical indicator worldwide, and different databases such as Google Scholar, Scopus, and WoS, use slightly different methods to calculate the "influence score" (Jangid et al., 2014). In the present bibliometric and mini-critical review paper, the top 10 most productive journals for microbubbles and nanobubbles in accordance with IF and QR rankings were presented as supplementary material in Tables S1 and S2.

It is evident that the journal 'Langmuir' was the most productive with 27 publications, which is about two times the number of the second journal in the ranking (colloids and surfaces a physicochemical and engineering aspects). It is to be noted that except for one journal (Journal of chemical engineering in Japan), all other journals were in the first and second quartile rankings (Q1 and Q2), with most of them possessing an IF above 2. This is important because it shows that various authors considered many high-ranking and eminent journals to publish microbubble articles. A more in-depth assessment revealed that with regards to nanobubbles, 'Langmuir' was also the most productive journal and had published even more papers: (considering the recent developments of nanobubbles) 54 articles, accounting for 13.88% alone. In the second rank was the journal of chemical physics with only 13 papers. This shows that Langmuir can be considered at the forefront in micro and nanobubbles-related research. All the top 10 most fecund journals had Q1 rankings, with IF above 2.6, and two journals: physical review letters and journal of colloid and interfaces sciences- had 8.38 and 7.49 impact factors, respectively. Aside from Langmuir, the journal of Applied physics letters in the rank 5 of microbubble studies, had rank 10 in nanobubbles studies. The fact that the overall quality (based on provided criteria) of journals publishing nanobubble research was higher than microbubble-involved journals confirm the authors' previous conjecture that the trend of nanobubbles studies will probably enjoy a drastic and notable increase as it has already started to attract the attention of many strong journals and authors.

3.5. Keywords co-occurrence network analysis

For popular topics in various research fields to be discovered, "keyword co-occurrence analysis" is employed which is composed of current science development and can be implemented even for very complex datasets (Gao et al., 2017; Lozano et al., 2019). Of the 3252 keywords, 207 keywords met the threshold of 5 times as the minimum number of co-occurrence. Among these 207 keywords, 100 keywords were opted to simplify the complex nature of the network. Additionally, some modifications were made to the final keywords, for instance, "microbubble" and "microbubbles" are almost identical. Therefore, choosing both at the same time can have a detrimental effect on the cluster. Moreover, some of the outlier keywords that were unrelated to environmental engineering were excluded, such as "contrast agents," "drug-delivery," "kinetics," "ultrasound contrast agent," "therapy," "in-

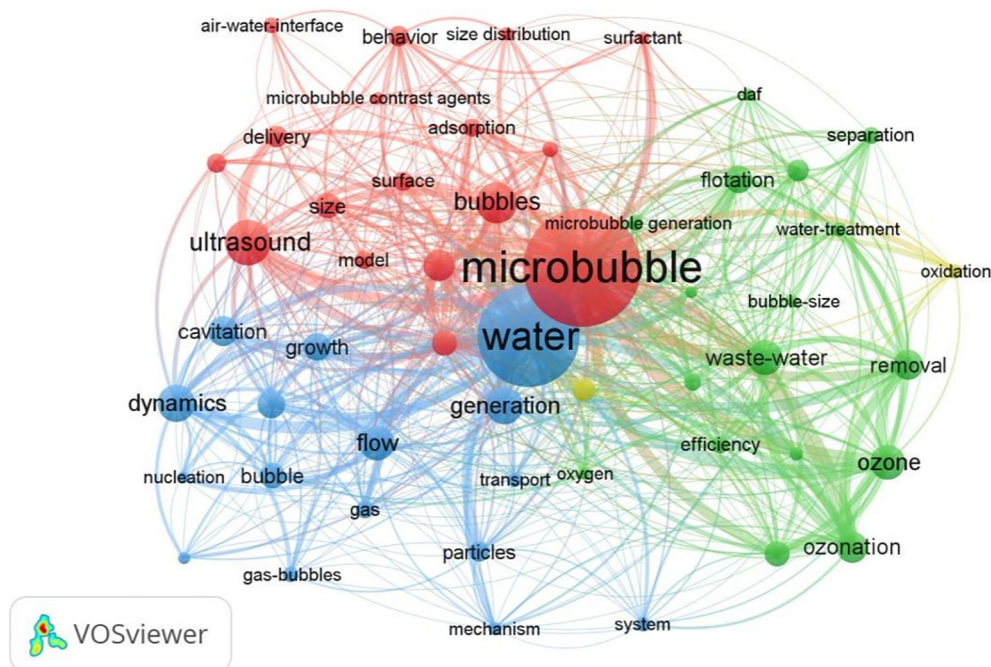


Fig. 13. Co-occurrence network of top 52 keywords delineating the frequency of co-occurrence (microbubbles).

vivo”, “gene delivery”, “drug”, “focused ultrasound”, “sonoporation”, “bloodstream”, “diagnostic ultrasound”, “tissue”, “blood-brain-barrier”, “cells”, “manipulation”, “phospholipid monolayer”, “phospholipids”, “lipid monolayers”, to mention but a few and the final result was comprised of 52 keywords. Many researchers often have the wrong assumption that including as many keywords as possible is beneficial. However, it has two deleterious effects; first, the final network will be so complex that it would become incomprehensible. Secondly, many

identical keywords (with slight differences) are redundant, creating extra clusters that are not related to the main topic's utilization or concept.

In VOS viewer software, using “size variation” and “scale” the graph for microbubbles was created. Both the density visualization and network map illustrated in [Figs. 13 and 14](#) ([de Toledo et al., 2019](#)) suggest the importance of both; The “curves,” are the representatives of co-occurrence between the connected “nodes.” Consequently, each node

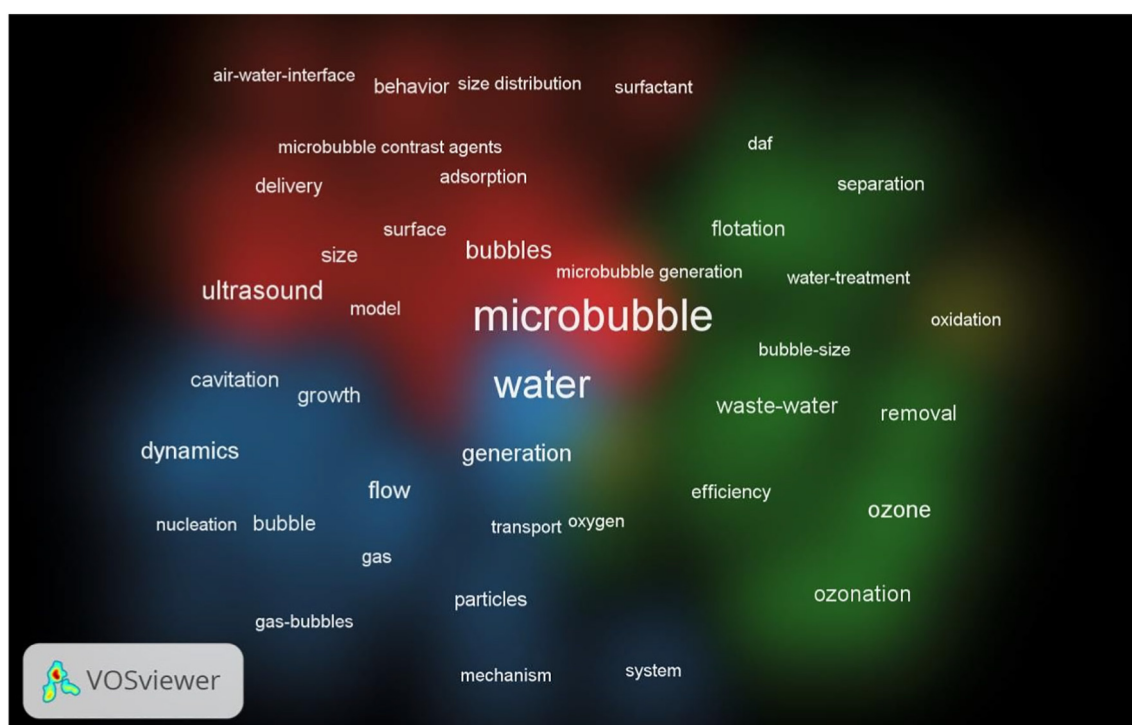


Fig. 14. Co-occurrence density visualization for 52 top keywords (microbubbles).

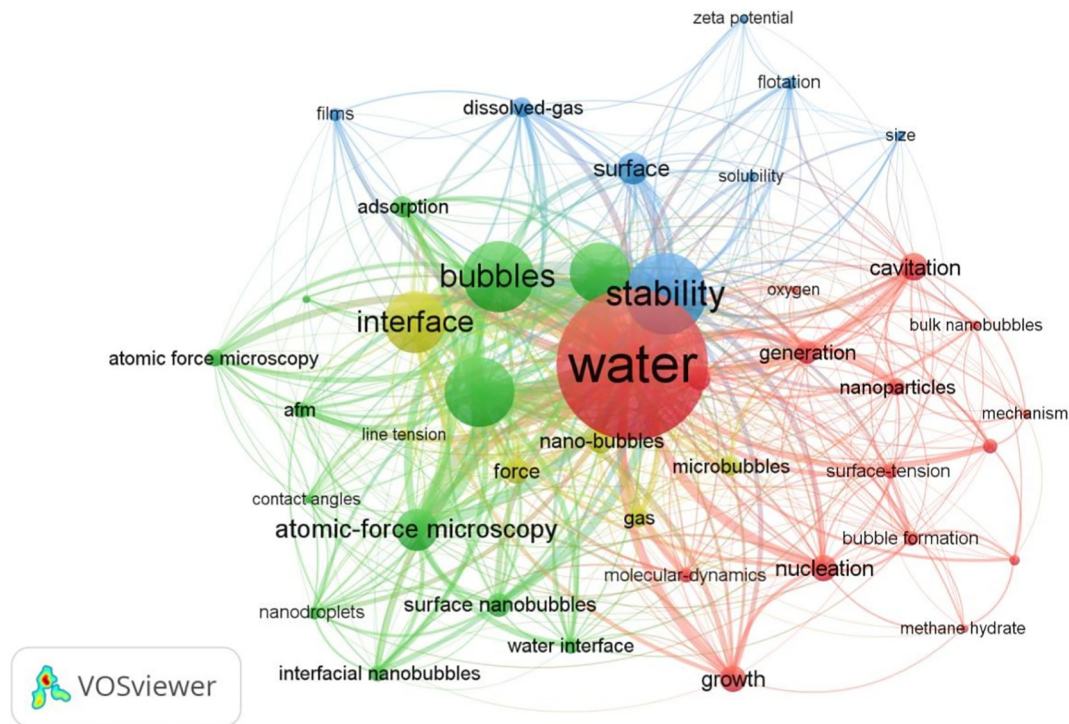


Fig. 15. Co-occurrence network of top 52 keywords delineating the frequency of co-occurrence (nanobubbles).

denotes a “keyword.” The keywords and their connection are depicted in Fig. 13, and it can be observed that they have been segregated into three distinct clusters, shown in three different colors. To explore further details of each cluster, Table 1 delineates the specifics of each cluster. To look at the clusters from an environmental perspective, a brief discussion is provided below.

As it was sporadically discussed throughout this article, many publications regarding micro and nanobubbles have endeavored to explore the nature of bubbles and the concomitant subjects. These subjects include “stability,” “size,” “the effect of ultrasound in producing microbubbles,” and the effect of microbubble “collapse,” which is a vital topic in pollutant degradation. As mentioned before, microbubbles,

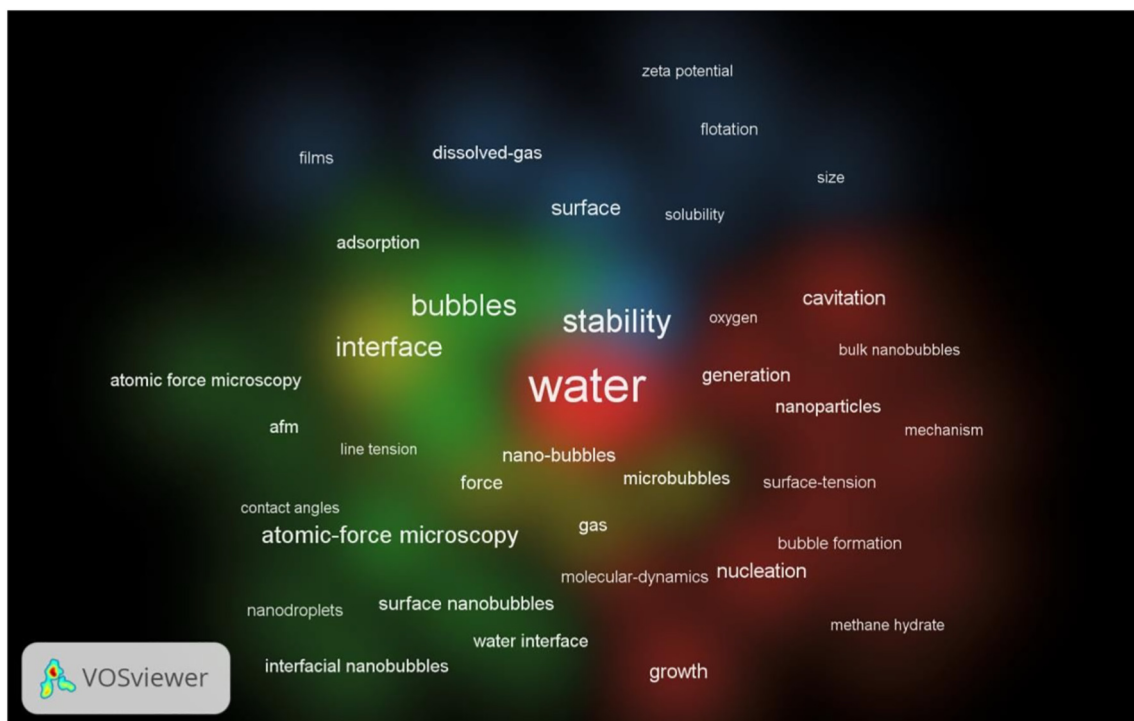


Fig. 16. Co-occurrence density visualization for 52 top keywords (nanobubbles).

Table 1
Clusters, color division, and most-occurred keywords (microbubbles).

#Cluster	Color	6 top keywords within the cluster (occurrences)
1	Red	Microbubble (176), Ultrasound (53), Bubbles (46), Stability (34), Size (26), Collapse (18)
2	Blue	Water (147), Generation (42), Dynamics (41), Flow (39), Cavitation (32), Growth (28)
3	Green	Ozone (39), Waste-water (38), Ozonation (33), Removal (32), Flotation (28), Degradation (26)

upon collapse, release a shockwave capable of degrading hazardous substances (Collince et al., 2020).

The mentioned topics can be seen as the major keywords with the highest link strength in Fig. 13 and with the highest density in “density visualization” in Fig. 14. Chronologically, the nature of these topics (except for stability, which is still a controversial matter) is relatively old, and therefore there are many publications in the last 20 years on these topics. To address the subject of stability, it should be noted that nanobubbles are nanoscopic gaseous domains in aquatic media. The controversy behind their elongated stability is the assumption that spherical bubbles are unable to reach a stable equilibrium. Considering the Young-Laplace pressure, the nanobubbles undergo a high internal pressure due to their smallness. In the equation $\Delta P = 2\gamma/R$, ΔP is the pressure difference of inside and outside the bubble, γ is the coefficient of surface, and R is the bubble radius (Li et al., 2021). So, in theory, the nanobubbles should burst within milliseconds because of the small radius and the extremely high Laplace pressure. This theory is in stark contrast with the empirical evidence of nanobubbles' long-lasting stability.

The Second cluster that is represented by blue color discusses the role of microbubbles in water and processes involving water such as “generation of microbubbles,” “cavitation processes,” “dynamics and kinetics of microbubble formation,” to mention a few. For microbubbles, specifically, “cavitation” that can damage the media in which the microbubble is produced (such as pumps) is an important topic; however, most recent studies on topics such as “cavitation” are in the field of biology and medicine, which is out of the scope of the present work (Wrede et al., 2018). As mentioned previously by the authors, the most recent use and technological advances in both micro and nanobubbles fields is employing bubbles for “treatment.”

As shown in the third cluster-green- “wastewater,” “water treatment,” “removal,” “flotation,” “ozone” and “ozonation” were the most important keywords and based on Fig. 14 (density visualization), “wastewater,” “removal” and “ozonation”, ostensibly, were the focus of most of the studies. Authors are of the opinion that “cluster 3” is the most important cluster. In clusters 1 and 2 publications, the authors were still trying to understand the nature of the technology (microbubbles' characteristics), then after this phase, they have tried to implement that knowledge into real-life environmental problems. Therefore, it is sensible for the number of studies in this regard to be fewer since it is still an inchoate area of research. Many recent publications are concerned with microbubbles and how they can be used to treat water and wastewater. Microbubbles (and nanobubbles) are technologies that are cost-efficient and environmentally benign (Fan et al., 2019). In a recent work (Li et al., 2020), the flotation performance of coal was improved using nanobubbles as an economical and cost-effective tool as a substitute for polyacrylic acid flocculant. These flocculants can harm the environment and are usually expensive (since they need to be used in excessive doses to be effective). As for microbubbles Lee et al. (2019) reported using ozone-generated microbubbles for total petroleum hydrocarbon stripping and decomposition of oil refinery sludge; at the same time, the ozonation of biologically treated municipal solid waste was investigated by Cheng et al. (2019).

The same procedure was followed for nanobubbles by excluding identical and irrelevant keywords, and a total of 43 keywords were

selected by dividing them into 4 clusters with four different colors (Table 2). A deeper look into the network of Fig. 15 and the density visualization of Fig. 16 provides the following insights.

The first cluster that is distinguished by red color, the most-occurred was “water.” Moreover, other connected keywords are mainly concerned with how nanobubbles are created rather than their water treatment use. As was the case with microbubbles, “cavitation” is also an important keyword for nanobubbles; however, “cavitation” here is also usually used in a different context. To be more specific, using venturi tubes and cavitation can result in nanobubbles generation, either hydrodynamic or acoustic cavitation, and they work as cavitation nuclei (Michailidi et al., 2020; Yasui et al., 2019a). Another noteworthy keyword in this cluster is “generation” that is due to the fact that producing nanobubbles is not as easy as microbubbles; therefore, many publications have strived to find optimum –for example, venturi geometry– for nanobubbles generation (Pourkarimi et al., 2017; Rak et al., 2019; Xiong and Peng, 2015).

In the second cluster (blue cluster), “stability,” “zeta-potential,” and “size” are especially important as they all contribute to nanobubbles formation and stability. “Stability” of nanobubbles is one of the major aspects, distinguishing them from microbubbles and macrobubbles (normal bubbles). If generated correctly, nanobubbles can remain in the water body for days and even weeks due to complex molecular dynamics and thermodynamics behavior. The concept of nanobubbles stability has caused heated arguments between scholars for years, and this has yet to reach a consensus. Nevertheless, nanobubbles possess very high stability and are not easily destroyed, as documented in the literature (Boshenyatov et al., 2019; Bui et al., 2019; Yasui et al., 2018); this is also true for surface nanobubbles (Wang et al., 2019).

The third cluster described by green color is less popular in nanobubble publications. It employs “atomic force microscopy” or “AFM” –another keyword in this cluster– to investigate nanobubbles nucleation's structures and the formation and destruction processes. However, these methods are costly, making the researchers reluctant to employ them, especially in institutions in countries with financial constraints resulting in fewer publications (shown by lower total link strength in Fig. 15) and lower densities, as shown in Fig. 16 (An et al., 2016).

In the fourth cluster, the most interesting matter is the presence of “microbubbles” as a keyword in nanobubbles studies. It was mentioned in the microbubble cluster analysis that the combination of micro and nanobubbles is a novel approach that utilizes unique characteristics of both nano and microbubbles to reach the most effective environmental remediation in various topics.

It is evident from the foregoing discussion that nanobubbles are still in the first phase that microbubbles were at the beginning of their pervasive use, that is, the study of “stability,” “formation,” and “understanding the intricate nature of nanobubbles.” The authors do envisage the second phase of nanobubbles publications concerned with water/wastewater treatment is yet to emerge, even though from 2017 to 2020, this phase has already started at a slow pace (Fan et al., 2020; Kim et al., 2020; Tekile et al., 2017).

Table 2
Clusters, color division, and most-occurred keywords (nanobubbles).

#Cluster	Color	5 top keywords within the cluster (occurrences)
1	Red	Water (78), cavitation (28), Nucleation (26), growth (26), Generation (22)
2	Blue	Stability (93), surface (32), Dissolved-gas (19), Flotation (12), Zeta potential (7)
3	Green	Bubbles (79), Hydrophobic surfaces (78), Nanobubbles (66), Atomic-force microscopy (44)
4	Yellow	Interface (68), Nanobubbles (28), Microbubbles (19), Gas(19), Line tension (9)

4. Concluding remark and future directions

Based on the 1034 number of nanobubbles and microbubbles publications, this bibliometric and critical analysis includes an up-to-date overview of research trends on micro and nanobubbles. Moreover, since the aim of the paper was not to be a mere statistical analysis, and considering the authors' involvement in "environmental engineering," each section is explained and fully elaborated from an environmental point-of-view. Therefore, the scientific reasoning behind every chart, graph, network, etc., is clearly explained. The growing number of publications in a 20-year period from 2000 to 2020 in both topics is an indication of the perpetual increase in this topic's significance. This increasing trend is evident in nanobubbles and microbubbles, with nanobubbles experiencing sharper growth in recent years. The authors anticipate this growth to be even more drastic in the near future based on the various reasons as explained in this article.

The most popular topics among researchers for "nanobubbles" were physical chemistry, multidisciplinary material science, nanotechnology and nanoscience, and environmental sciences. The recent papers being published in the "environmental science" category and older publications in physical chemistry. This indicates a paradigm shift in the nature of the topics, from studying the nature of bubbles to their utilization in the environment.

As for the microbubble, "environmental sciences" and "environmental engineering" combined took a large share among other categories, showing a promising trend toward this topic.

The journal analysis indicated that the Langmuir journal was the most prolific journal on microbubbles and nanobubbles and had the highest quality of research papers (based on IF and quartile rankings). Alongside journal analysis, the country analysis revealed that developed and high-income countries such as the USA and Japan that can afford the generation and measurement techniques of bubbles (especially nanobubbles) were the most productive in recent years in the 20-year period. Additionally, due to its population and the number of students it exports to other countries, China has surpassed all the other countries.

In the near future, new nanobubble generation methods and devices will probably emerge capable of improving the stability and increasing the population of the produced bubbles. A shift in conventional methods of nanobubble generation is evident, from hydro cavitation to using temperature variations to induce nanobubble nucleation or employing hollow honeycomb structures.

Another possible future application is the combination of nano and microbubbles and surfactants (surface-active agents). Using different surfactants can generate "charged bulk MNBs," a promising perspective for removing charged particles in water.

One of the most important aspects of bubbles is the fact that they are environmentally benign. Therefore, their application should enjoy a greater possibility and not be constrained to treatment subjects. Very recently, microbubbles have been employed in the food industry to facilitate the ripening process of some crops.

The researchers can also focus on developing industrial scale nanobubble generator devices that employ methods other than hydro cavitation. At present, there are only a few companies that can manufacture such devices, and they use heavy water pumps. Since this technology has not matured yet, it is a great opportunity for researchers to pursue more subtle and efficient methods to produce bulk nano and microbubbles at an industrial scale.

In the end, the keyword analysis showed that most of the microbubble publications were, in recent years, concerned with "water/wastewater" treatment, which the authors believe is the second phase after a technology is introduced. The same phase is beginning to initiate nanobubbles. In the future, the nanobubbles used in water and wastewater treatment will receive immense attention, and their use in environmental amelioration such as separation methods, horticulture, agriculture, membrane fouling prevention, and regeneration, etc., will be far greater than microbubbles. Moreover, nanobubbles and

microbubbles combined use is yet to be explored and has great potential. It is recommended that other databases such as Scopus, Google scholar, etc., can validate the present study's findings.

It is worth mentioning that both these topics (micro and nanobubbles) are considered very young, yet to reach maturity. These topics are unlike other topics that possess more than 4000 articles in a 20-year period. We believe it is, in fact, this aspect that makes this publication worthy. Because it can be used as the basis for prospective research that has the incentive to explore either one of these topics, understand them better, and even compare them. There have been no bibliometric studies in this area to the best of the authors' knowledge, let alone a critical review to elucidate the trends and confer environmental insight. The authors believe this tabula rasa is not conducive to this field, proving the value of this publication.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2021.147362>.

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