

Scientometric Assessment and Mini Review of Graphitic Carbon Nitride ($g-C_3N_4$) for Photocatalytic CO_2 Reduction: An Evidence of Progress

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ABSTRACT:

The scientific community globally have sought for alternative energy generation approaches and environmental remediation techniques to address the changes in consumption patterns of fossil fuel. Semiconductor photocatalysis has been reported as a potential technique to address the aforementioned challenge with graphitic carbon nitride ($g-C_3N_4$) identified as one of the novel nanomaterial exploration and applications in diverse fields. Many reviews have been reported on $g-C_3N_4$ focusing on photocatalysis principles, preparation techniques, photocatalytic enhancement strategies and various photocatalytic applications. However, in this study, a scientometric assessment was conducted to detail the research status on $g-C_3N_4$ semiconductor for photocatalytic reduction of CO_2 and visualization of the future research trends. The scientometric review covers 504 published articles retrieved from the SCOPUS database online spanning from 2010 to May 2021. The articles were analysed using the VOSviewer mapping tool software using 10 scientometric indices. The results show 461 authors globally contributed to the topic with 333 articles from China and progressive increase in published articles in diverse journals from 1 in 2010 to 88 in May 2021. Collaborations exist between authors and co-authors from various countries and institutions. The new trending areas for $g-C_3N_4$ photocatalysts include nanomedicine, biomedical systems, toxicological industries, food safety monitoring and mathematics.

Keywords: Scientometric analysis, $g-C_3N_4$, Semiconductor, CO_2 reduction, Photocatalysis.

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INTRODUCTION

The continuous development of human society has resulted in an increase in environmental waste generation and demand for energy globally. The current energy supply globally depends largely on fossil fuel sources such as coal, petroleum reserves and natural gases which have been depleted rapidly.¹⁻² Furthermore, the consumption of energy from these fossil fuels also contributes to generation of harmful gases into the environment. Over three decades, the scientific community are engineering various materials to overcome the aforementioned challenge of environmental

remediation and energy conversion as alternatives to fossil fuel.³⁻⁴ Semiconductor photocatalysis has shown a potential promise as a green technology to generate solar fuels and for environmental remediation applications. Semiconductor photocatalysis has received great interdisciplinary attention for conversion of harmful CO_2 into valuable solar hydrocarbon fuels as well as approach for pollutants removal, water disinfection, and air purification. In addition, semiconductor photocatalysis has shown a great potential in splitting of water into oxygen (O_2) and hydrogen (H_2), where the hydrogen could be used as a source of energy.^{2,5-7} To date, several high quality semiconductor photocatalysts have been developed and utilized under both ultraviolet (UV) and visible light.^{1,8-11}

Among the several semiconductor photocatalysts fabricated is graphitic carbon nitride ($g-C_3N_4$) which is a metal free polymeric material. The research communities have

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conducted in-depth research into g-C₃N₄ because of its high physicochemical properties, easy facile preparation, moderate bandgap (≈ 2.7 eV), suitable electronic band structure, non-toxicity and earth abundant nature.¹²⁻¹⁴ The pristine g-C₃N₄ photocatalyst is easily synthesized through thermal polycondensation of organic precursors such as thiourea, urea, melamine, cyanamide and dicyandiamide. However, the pristine g-C₃N₄ obtained from these synthesis process has limited photocatalytic performance from poor visible light harvesting ability, low surface area and rapid recombination of photogenerated charge carriers.^{1,12-13} To overcome the aforementioned challenges, several modification techniques such as doping, heterostructuring, surface plasmon resonance effect, morphology tuning, metal deposition, semiconductor coupling etc. have been reported to enhance the photocatalytic performance of g-C₃N₄.^{1,12,14-16} There is an increase interest in the study of g-C₃N₄ semiconductor photocatalysis recently with the past few years witnessing a surge in published literature reviews.^{1,13,17-19} Most of the literature reviews regarding g-C₃N₄ photocatalysis focused on photocatalysis principles, physicochemical properties, preparation techniques, photocatalytic enhancement strategies and various photocatalytic applications.^{1,17,20-27} However, to the best of our knowledge, detailed research status on g-C₃N₄ semiconductor photocatalysis and visualization of the future research trends have not been fully explored. Thus, in order to address this existing research gap, a scientometric analysis was conducted. A scientometric analysis involves quantitative study of science, communication in science and science policy.²⁸ It includes assessment and measurement of impact of published articles, authors, journals, institutions, scientific citations and visual mapping in the scientific area.²⁹ In this study, an attempt was made to conduct a scientometric review of published articles in the field of g-C₃N₄ photocatalysis and obtained a snapshot of trends in this field from 2010-2021. In addition, a mini review on the multifunctional applications of g-C₃N₄ as a photocatalyst was also conducted. The results from the study can provide researchers better understanding, development of current trends and hot topics in g-C₃N₄ photocatalysis.

METHODOLOGY

The study employed the bibliometric analysis on g-C₃N₄ and photocatalysis. Scientometric studies involved quantitative aspects of recorded information in a research field. The information for the study was obtained from SCOPUS database in CSV files format. The CSV files were exported into a VOSviewer software which is a mapping tool for Scientometric analysis. A Scientometric analysis is a measure of an evolution of a scientific domain, scholarly publications impact, authorship patterns and scientific knowledge production process. Thus, Scientometric analysis involved

progress of research and researchers in a particular field of study and various relationships such as research institutes, universities, researchers, journal publishers and particulars of journals in a scientific domain.³⁰ In this study, ten (10) scientometric techniques were employed to carry out the analysis. They were analysis of documents, research by subject areas, publication trends, keywords, authors, countries and institutions, citations, co-citations and journals.

Data Collection

The data for the study was obtained from the online SCOPUS database. The SCOPUS database contained about 70 million items and regarded as one of the comprehensive and influential database for academic journal publications related to Elsevier publisher.³¹ The search was conducted in May 2021 with the collection of documents spanning from 2010-2021. Keywords in publication titles, abstracts and keywords were searched in the Scopus database. Specifically, the query searched in Scopus was ((TITLE (“carbon nitride” OR “g-C₃N₄” OR “graphitic carbon nitride” OR “photocatalysis”) AND PUBYEAR > 2009) AND (“CO₂ reduction” OR “carbon dioxide reduction”)). The search returned 504 documents which includes journal articles, reviews, chapter of books and book.

RESULTS AND DISCUSSION

Types of Documents and Numbers

In this analysis, the search conducted returned a total of 504 documents which was exported in a CSV excel statistic data format from Scopus database of Web of Science (WOS). Figure 1 shows the document types recorded with Articles being 390 (77.38%), Reviews were 106 (21.03%), Book Chapters were 7 (1.39%) and Book was 1 (0.20%). The publications were done in two (2) different languages with 99.6 % proportion in English (i.e. 502 documents) and 0.4% in Chinese (i.e. 2 documents). This implies that the almost all the publications relating to g-C₃N₄ for photocatalytic reduction of CO₂ into solar fuels are in English language.

Distribution According to Research Subject Areas

The numbers of subject areas of publication of g-C₃N₄ for photocatalytic reduction of CO₂ were also studied and analysed. Chemistry, Chemical Engineering and Material Science were the top three research subject areas for the field of g-C₃N₄ in photocatalysis (Figure 2). This is because g-C₃N₄ is a novel metal free semiconductor material which have gained enough research attention over the past two decades. The researches focused on application of the g-C₃N₄ materials in diverse fields such as environmental application, green energy system etc. Currently, the research direction of g-C₃N₄ is shifting to medicine and other multidisciplinary areas as depicted in Figure 2. According to Density Functional Theory (DFT)

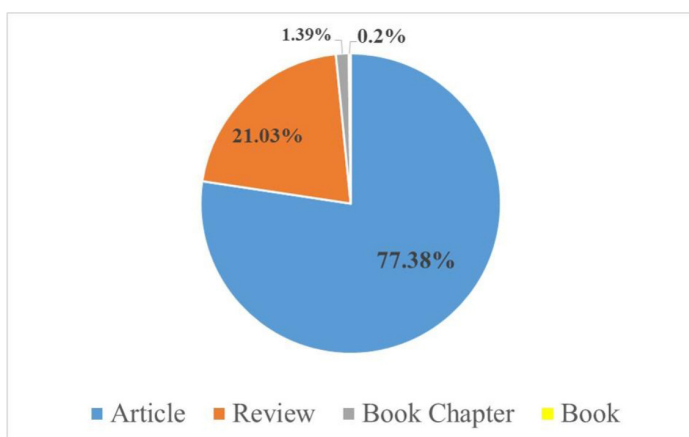


Figure 1: Distribution of proportions of document types in the field of $g-C_3N_4$ for CO_2 photocatalytic reduction.

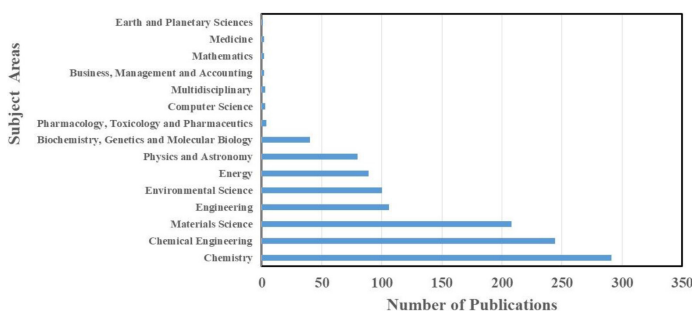


Figure 2: Distribution of publications based on subjects / disciplines.

studies, $g-C_3N_4$ has a chemotherapeutic potential as a drug carrier for carboplatin and cisplatin (anticancer drugs) in cancer treatment and thus applicable in medicine.³²⁻³³

Trend Analysis of Publications from 2010 – 2021

The global publication trend in the area of $g-C_3N_4$ for photocatalytic reduction of CO_2 from 2010–2021 is shown in Figure 3. It can be observed that published documents progressively increased over the years. From 2010 to 2017, the number of documents published was relatively small from 1 to 30 respectively. This could be attributed to the synthesis, characterizations and the application of $g-C_3N_4$ for photocatalytic CO_2 into solar fuels were still not clear which needed further probe. The number of documents published increased from 2018 (i.e. 68 documents) to 88 documents (i.e. May 2021) significantly due to further researches conducted in this field and the application process become very clear and understandable. The issue of climate change and energy shortage globally is attracting a lot of research. Currently, the development of visible light driven photocatalysts for converting CO_2 into a valuable solar fuels is a research hotspot of which $g-C_3N_4$ has more potential compared to other materials.³⁴⁻³⁵ This also accounted for more published documents in this field recently.

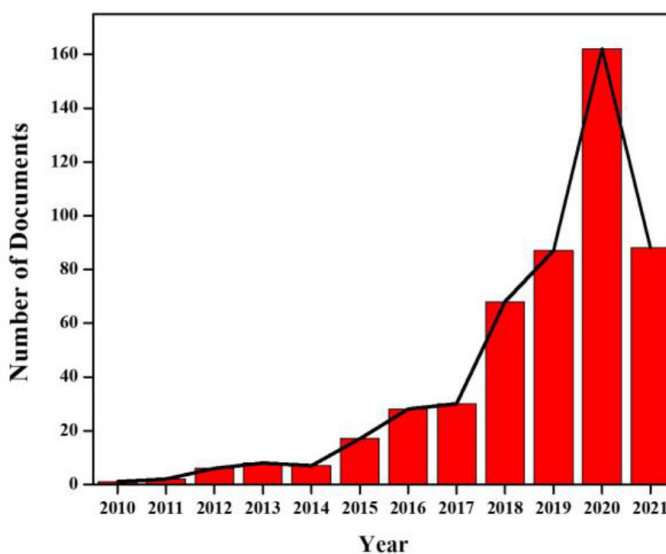


Figure 3: Publication of documents trend over the search period.

Keyword Co-occurrence Analysis and Clustering

Keywords are very important for published documents in a scientific domain for characterization in a specific field. It represents the main content of published articles and display the wide range of the research areas within the limit of any field.^{30,36} In this study, clustering of keywords which is a process of placing analysed items with certain similarities into groups or sub categories was carried out. This implies that each cluster of keywords have items or data with similar characteristics but these characteristics differs among the clusters.³⁰ The cluster of keywords was also used to identify most suitable article or document among a prevailing list of publications in a scientific field and also shows relationship among each word. The Scopus database provides two types of keywords namely the "author keywords" (i.e. keywords that authors provided) and "index keywords" (keywords indexed by journals for article retrieval). Both keywords were employed in this study to ensure that all documents were retrieved.^{29,30} The VOSviewer was used to construct and map the field of knowledge between photocatalysis and carbon nitride ($g-C_3N_4$) using keyword co-occurrence. The result of the scientometric analysis of published documents was shown in the visualization of keywords network of the VOSviewer. The VOSviewer output is a distance-based map where the distance represents the strength of the relation between the two knowledge domains. Generally, the larger or bigger the distance, the weaker the relationship between the two domains. Furthermore, item label size is directly proportional to the number of publications found for the keyword.³⁶

The VOSviewer technique cluster the different knowledge domains using different colours as shown in Figure 4. In this study, the minimum number of occurrences was set to 2 and out of the 3583 keywords, 1085 met the threshold.

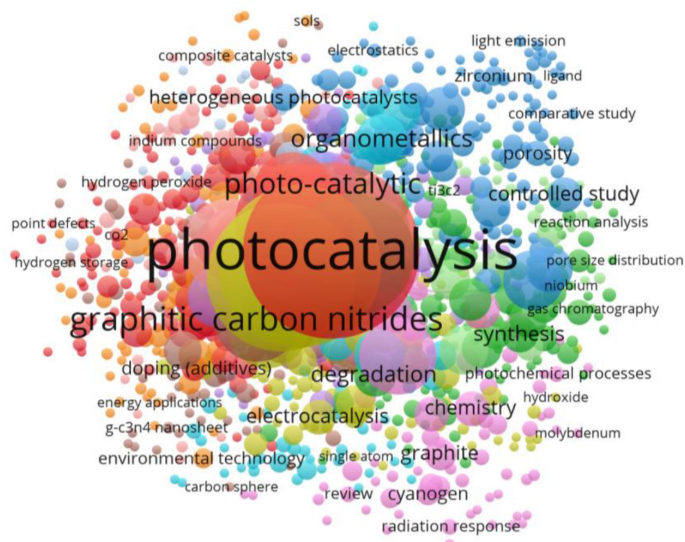


Figure 4: Visual keywords co-occurrence analysis.

To obtain optimal clusters, multiple experiments were used for the threshold selection. The network of co-occurring keywords produced 1000 nodes, 45367 links and 70623 total link strength with 12 clusters. Table 1 shows the first twenty (20) keywords occurrence from the various clusters, their links and total link strength. The top four keywords identified from the VOSviewer output were “Photocatalysis” (Frequency = 240), “Carbon dioxide” (Frequency = 211), “carbon nitride” (Frequency = 182) and “light” (Frequency = 134). The keywords belong to different clusters and were all subject terms in the field. The term “photocatalysis” is generic which is used to describe the process of utilization of light to stimulate a catalytic activity while the term “carbon nitride” represent a specific semiconductor material. Catalytic carbon dioxide conversion can be grouped into different classification namely electrocatalysis, photocatalysis, traditional thermal catalysis, plasma catalysis etc.² Therefore, these keywords evidenced that from 2010–2021, photocatalytic reduction of CO₂ into valuable solar fuels using g-C₃N₄ as a semiconductor has received a lot of attention. This is because it is among the important hottest research areas that has the potential of solving the current energy crisis and environmental problems facing the world. In addition, the g-C₃N₄ semiconductor material can easily be prepared, cheap, possess high chemical and physical stability among other characteristics^{22,37} which has made it an ideal material worth researching.

For better clarification and understanding of the research hotspots, the keywords in the field of g-C₃N₄ were clustered into 12 by the VOSviewer software. Cluster 1 (made up of 128 items) focused on the general description and applications of photocatalysts such as artificial photosynthetic, doping, heterojunction, morphology, CO₂ reduction, environmental application, environmental pollution etc. Cluster 2 (126 items)

Table 1: Summary of Top 20 Occurrence Keywords and Each Individual Node Strength.

| Keyword | Cluster | Occurrences | Links | Total link strength |
|---------------------------------|---------|-------------|-------|---------------------|
| Photocatalysis | 1 | 240 | 891 | 3849 |
| Carbon dioxide | 4 | 211 | 844 | 3661 |
| Carbon nitride | 4 | 182 | 792 | 3075 |
| Light | 2 | 134 | 732 | 2548 |
| Photocatalytic activity | 1 | 124 | 674 | 2138 |
| CO ₂ reduction | 1 | 112 | 581 | 1742 |
| Heterojunctions | 10 | 87 | 532 | 1504 |
| Graphitic carbon nitrides | 8 | 74 | 481 | 1368 |
| Nitrides | 8 | 70 | 462 | 1288 |
| Photocatalytic performance | 1 | 70 | 505 | 1230 |
| Graphitic carbon nitride | 4 | 68 | 463 | 1063 |
| Hydrogen production | 4 | 66 | 470 | 1156 |
| Light absorption | 1 | 63 | 492 | 1176 |
| Catalysis | 9 | 62 | 520 | 1290 |
| g-C ₃ N ₄ | 2 | 55 | 373 | 790 |
| Solar energy | 7 | 55 | 448 | 1012 |
| Photo-catalytic | 5 | 49 | 388 | 761 |
| Catalysts | 7 | 46 | 418 | 790 |
| Charge transfer | 10 | 46 | 379 | 760 |
| Catalyst activity | 4 | 45 | 360 | 658 |

generally represent terms which is used to describe the chemical processes and mechanisms of CO₂ reduction such as absorption, adsorption, charge carrier recombination, chemical interaction, chemical structure, light etc. Cluster 3 (120 items) represent in-depth terms used to illustrate chemical and electronic structure or properties such as electron transport, excited state, light harvesting, light scattering, mesoporous etc. The frequency of the highest keywords in cluster 4 is associated with the atomic and electronic structure description. They involved keywords such as binding energy, atoms, DFT calculation, first principle calculation, photocurrent response etc. From Table 2, it can be seen that most of the keywords belongs to cluster 1 which described photocatalysis in general and very broad terms to accommodate other important sub areas.

Co-occurrence Analysis (e.g., countries, institutions, and authors)

Author-Co-authors network

The contribution of authors in the field of photocatalysis and g-C₃N₄ was extracted from the VOSviewer software. This information would help identify and mapped out lead or highly productive researchers in the field as well as collaboration or cooperation between the researchers. A total of 461 authors were retrieved and the top 25 productive authors in the studied field during 2010–2021 was presented in Table 2. Zhang X

Table 2: The Top 20 Productive Authors during 2010-2021 in the Field of g-C₃N₄ and Photocatalysis for CO₂ Reduction.

| Ratings | Authors | Article Frequency | Citations | Country |
|---------|----------|-------------------|-----------|---------|
| 1 | Zhang X. | 27 | 1547 | China |
| 2 | Zhang J. | 25 | 603 | China |
| 3 | Li Y. | 24 | 1300 | China |
| 4 | Li X. | 24 | 786 | China |
| 5 | Wang Y. | 23 | 754 | China |
| 6 | Li Z. | 22 | 1543 | China |
| 7 | Wang H. | 22 | 557 | China |
| 8 | Li H. | 21 | 329 | China |
| 9 | Wang X. | 20 | 1308 | China |
| 10 | Zhang Y. | 18 | 932 | China |
| 11 | Wang L. | 16 | 1006 | China |
| 12 | Wang C. | 13 | 1890 | China |
| 13 | Wang S. | 12 | 819 | China |
| 14 | Liu Y. | 12 | 446 | China |
| 15 | Zhang T. | 11 | 1627 | China |
| 16 | Xu H. | 11 | 575 | China |
| 17 | Wang Z. | 11 | 265 | China |
| 18 | Liu J. | 11 | 251 | China |
| 19 | Zhang H. | 10 | 585 | China |
| 20 | Zhao Y. | 10 | 156 | China |

produced the largest number of articles (27). Zhang J, Li X and Li Y were rated as second, third and fourth productive authors with 25, 24 and 24 articles respectively within 2010-2021 period. Figure 5 shows the author and co-authorship network.

In this network, the nodes denote an author and the relationship of the collaboration between the author and co-authors of a publication are represented by the links. The size of the node represent the number of articles published while the thickness of the link represent the level of collaboration or cooperation between the authors.²⁹ In all, 461 nodes and 3356 links were generated with a total link strength of 4108.

Figure 5 depicts that there were a lot of collaborations between author and co-authors represented by both the closed-loop and small circuits. The researchers in the circuits show establishment of collaboration or cooperation such as circuit of Zhang X and Zhang J. Furthermore, the study also identified other numerous research communities where many authors had a collaborative working relationship with one or two highly productive author(s). Generally, this research field has several international collaboration or cooperation which would be beneficial to the scientific communities globally.

To determine the extent of many connections or links within the nodes (i.e. authors and co-authors), Betweenness Centrality (BC) analysis was conducted. The BC is defined as

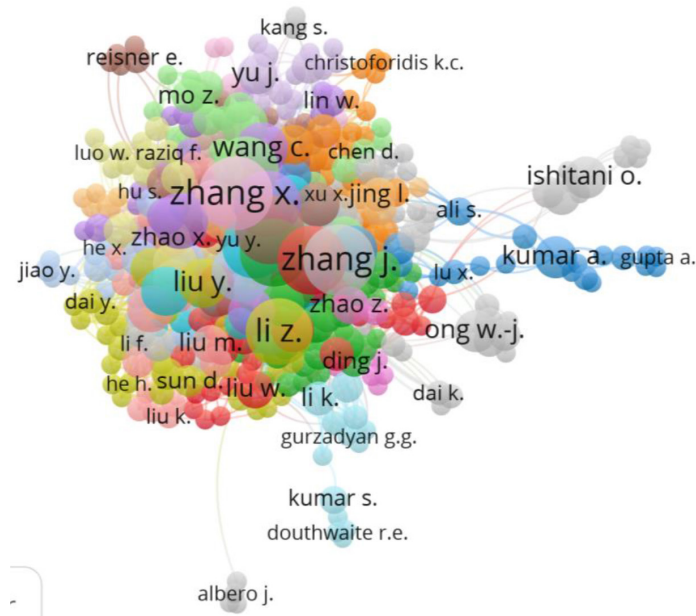


Figure 5: The author and co-authorship map generated from the VOSviewer.

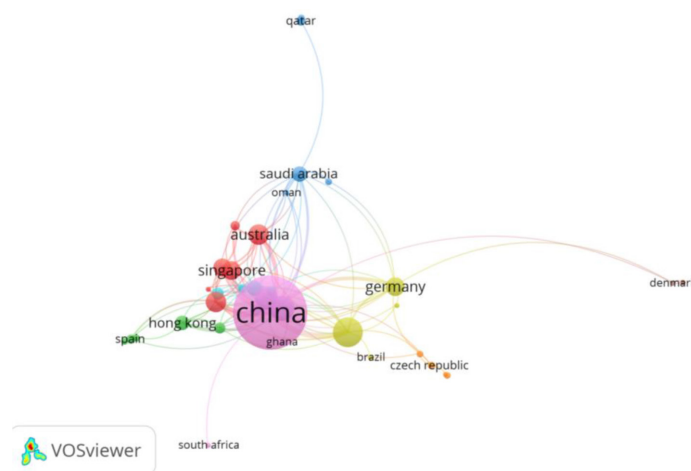
the ratio of the shortest path between two nodes to the sum of all such shortest paths.³⁸⁻³⁹ In other words, BC determines the centrality of nodes and nodes value lying in a range of 0 to 1. A node with BC one (1) means it has maximum intermediates (i.e. there is usually connection of two or more large groups of nodes with the node itself in-between) while BC of zero (0) implies that only one line connects two ending nodes.³⁰ In this study, Zhang J have the highest BC of 0.03, which is a quite low, an indication of a weak collaboration between researchers which may need further improvement.

Network of countries and institutions

To further explore the distribution of research publication outputs in field of g-C₃N₄ and photocatalysis for CO₂ reduction, a network map based on countries and institutions were generated. The VOSviewer produced 37 nodes, 118 links and a total link strength of 265. The size of the node represents the total number of published articles within the study period of 2010-2021. China (333 articles), United States (54), India (38) and Japan (30) were the top four main published article contributing countries on the field of g-C₃N₄ and photocatalysis (Table 3). The large number of published articles in these countries suggested that advance research has been conducted in the area of g-C₃N₄ and photocatalysis. In this search, China had the highest number of articles of 333 and could be attributed to the fact that almost all the Universities are having Material Science, Engineering and Processing Departments where published articles were produced. Furthermore, researchers from China have a wide range of international collaboration with other researchers as shown in the author and co-authorship map (Figure 6)

Table 3: Contribution of Published Articles from Different Countries on g-C₃N₄ and Photocatalysis for CO₂ Reduction.

| Rank | Country | Articles | Citations |
|------|---------------|----------|-----------|
| 1 | China | 333 | 14467 |
| 2 | United States | 54 | 6260 |
| 3 | India | 38 | 1384 |
| 4 | Japan | 30 | 1866 |
| 5 | Australia | 27 | 4894 |
| 6 | Unite Kingdom | 26 | 1318 |
| 7 | Germany | 23 | 864 |
| 8 | Singapore | 22 | 1041 |
| 9 | Malaysia | 20 | 4227 |
| 10 | Saudi Arabia | 15 | 1366 |
| 11 | South Korea | 15 | 824 |
| 12 | Hong Kong | 13 | 1195 |
| 13 | Canada | 9 | 61 |
| 14 | France | 8 | 364 |
| 15 | Taiwan | 8 | 1213 |
| 16 | Iran | 6 | 585 |
| 17 | Pakistan | 6 | 261 |
| 18 | Qatar | 6 | 28 |
| 19 | Spain | 6 | 650 |
| 20 | Vietnam | 5 | 44 |

**Figure 6:** A visual map generated from the VOSviewer based on contribution of publications by countries.

which could be a contributing factor to the high number of published articles been churned out.

The BC for the top five countries were India (0.51), Australia (0.40), Japan (0.38), United States (0.30) and China (0.17). This also shows there is research activities connection and network between different countries globally. The contributions of organizations or institutions in the field of g-C₃N₄ and photocatalysis were also produced.

A total of 97 different organizations or institutions were found. Among the top twenty (20) organizations or institutions, seventeen (17) were from China, two (2) from Japan and one (1) from Australia. The Chinese Academy of Sciences was rated as the first organization or institution contributing seven (7) articles, followed by Jiangsu University, (5) articles and Tianjin University (4) articles. This implies that Chinese organizations or institutions are leading in the research field of g-C₃N₄ and photocatalysis.

Authors and citation

Authors and citation relationship established the frequency of which published articles of author(s) are mentioned or cited in documents. It also shows the collaboration among the authors and the impact of cited articles in the field of study. The academic impact of an article is measured based on the number of citations the article received. In all, 472 authors were generated into sixteen (16) clusters with 11702 links and a total link strength of 17012. The top twenty (20) most cited author(s) are presented in Table 4 with Ong W. J. (Citation frequency = 2986) being the top-ranked highly cited author. Zhang T. (Citation frequency = 1351), Zhao Z. (Citation frequency = 1037) and Wang C. (Citation frequency = 1029) were ranked second, third and fourth respectively. The highly cited article was published in Chemical Reviews in the year 2016. This was a review article which have received many readings and hence citations from the scientific community. Other journals such as Chemical Society Reviews, Nanoscale and Journal of the American Chemical Society were the publishers of the second, third and fourth highly cited articles. The first four highly cited articles were published by Chinese researchers. This clearly shows that the highly cited author(s) published articles in diverse journals and in different years. This is an indication that research on g-C₃N₄ and photocatalysis is on a wide increase and receiving global attention.

Documents and co-citations of Journals

The sources of g-C₃N₄ and photocatalytic articles from 2010 to 2021 were retrieved from 142 journals with 734 links. Table 5 shows the 20 top-ranked journals and their citations. The top ranked journal “Applied Catalysis B: Environmental” published 36 documents that have received 2327 citations. The “Chemical Engineering Journal” have 19 documents and received 437 citations. The “Journal of Materials Chemistry A” (citations = 400) and “Applied Surface Science” (citations = 233) were ranked third and fourth with published documents of 16 and 15 respectively. This clearly shows that articles relating to g-C₃N₄ and photocatalysis published in these top four journals have received high attention from researchers in the field.

Table 4: The Top Twenty (20) Highly Cited Authors and Journals in the Field of g-C₃N₄ and Photocatalysis for CO₂ Reduction.

| Rank | Author(s) | Title of article | Year | Name of Journal | Citation |
|------|---------------------|---|-------|---|----------|
| 1 | Ong W. J. | Graphitic Carbon Nitride (g-C ₃ N ₄)-Based Photocatalysts for Artificial Photosynthesis and Environmental Remediation: Are We a Step Closer to Achieving Sustainability? | 2016a | Chemical reviews | 2986 |
| 2 | Zhang T. | Metal-organic frameworks for artificial photosynthesis and photocatalysis | 2014a | Chemical Society Reviews | 1351 |
| 3 | Zhao Z. | Graphitic carbon nitride- based nanocomposites: A review. | 2015 | Nanoscale | 1037 |
| 4 | Wang C. | Doping metal-organic frameworks for water oxidation, carbon dioxide reduction, and organic photocatalysis | 2011 | Journal of the American Chemical Society | 1029 |
| 5 | Hou W. | A review of surface plasmon resonance-enhanced photocatalysis | 2013 | Advanced Functional Materials | 926 |
| 6 | Fu J. | g-C ₃ N ₄ -Based Heterostructured Photocatalysts | 2018 | Advanced Energy Materials | 903 |
| 7 | Zhang X. | Plasmonic photocatalysis | 2013 | Reports on Progress in Physics | 820 |
| 8 | Wang J. L. | Metal-organic frameworks for light harvesting and photocatalysis | 2012 | ACS Catalysis | 566 |
| 9 | Gao G. | Single atom (Pd/Pt) supported on graphitic carbon nitride as an efficient photocatalyst for visible-light reduction of carbon dioxide | 2016 | Journal of the American Chemical Society, | 555 |
| 10 | Mamba G. | Graphitic carbon nitride (g-C ₃ N ₄) nanocomposites: a new and exciting generation of visible light driven photocatalysts for environmental pollution remediation | 2016 | Applied Catalysis B: Environmental | 550 |
| 11 | Dhakshinamoorthy A. | Catalysis and photocatalysis by metal organic frameworks | 2018 | Chemical Society Reviews | 522 |
| 12 | Fateeva A. | A water-stable porphyrin-based metal-organic framework active for visible-light photocatalysis | 2012 | Angewandte Chemie International Edition | 475 |
| 13 | Wang S. | Multifunctional metal-organic frameworks for photocatalysis | 2015 | small | 366 |
| 14 | Sun D. | Studies on Photocatalytic CO ₂ Reduction over NH ₂ -Uio-66 (Zr) and Its Derivatives: Towards a Better Understanding of Photocatalysis on Metal-Organic Frameworks | 2013 | Chemistry-A European Journal | 366 |
| 15 | Zhou Y. | Monolayered Bi ₂ WO ₆ nanosheets mimicking heterojunction interface with open surfaces for photocatalysis. | 2015 | Nature communications | 362 |
| 16 | Park H. | Photoinduced charge transfer processes in solar photocatalysis based on modified TiO ₂ . | 2016 | Energy and Environmental Science | 360 |
| 17 | Luo B. | Recent advances in 2D materials for photocatalysis. | 2016 | Nanoscale | 357 |
| 18 | Wen J. | Photocatalysis fundamentals and surface modification of TiO ₂ nanomaterials | 2015 | Chinese Journal of Catalysis | 347 |
| 19 | Samadi M. | Recent progress on doped ZnO nanostructures for visible-light photocatalysis | 2016 | Thin Solid Films | 314 |
| 20 | Zheng J | Zn phthalocyanine/carbon nitride heterojunction for visible light photoelectrocatalytic conversion of CO ₂ to methanol | 2019 | Journal of Catalysis | 305 |

Multifunctional Applications of g-C₃N₄ Based Photocatalyst

Graphitic carbon nitride (g-C₃N₄) has emerged as a metal free semiconductor photocatalyst over the last three decades. It possesses exceptional properties which are favourable for multifunctional applications such as environmental remediation, renewable solar fuel generation, water splitting, optoelectronic uses among others.^{19,25,40-41}

Environmental Degradation of Pollutants

Population growth and industrialization of economies has resulted in the emission of high toxic and hazardous substances into the environment. These issues have a potential of posing health risks to human health. Photocatalysis promised to be a green technology the scientific community has explored and considered to be a potential remedy to preserve the environment.¹ A lot of studies have been conducted extensively on g-C₃N₄ for environmental applications such as water purification, air purification, and hazardous

Table 5: Top Twenty (20) Productive Journals from 2010-2021 in the field of g-C₃N₄ and Photocatalysis.

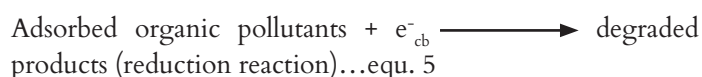
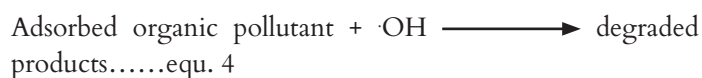
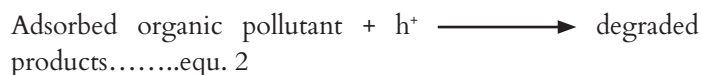
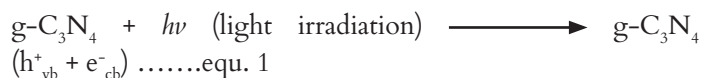
| Rank | Journal Sources | Documents | Citations |
|------|---|-----------|-----------|
| 1 | Applied Catalysis B: Environmental | 36 | 2327 |
| 2 | Chemical Engineering Journal | 19 | 437 |
| 3 | Journal Of Materials Chemistry A | 16 | 400 |
| 4 | Applied Surface Science | 15 | 233 |
| 5 | Angewandte Chemie - International Edition | 13 | 1384 |
| 6 | Journal of The American Chemical Society | 12 | 2131 |
| 7 | Chemcatchem | 11 | 203 |
| 8 | Journal of Colloid and Interface Science | 10 | 210 |
| 9 | ACS Applied Materials and Interfaces | 9 | 621 |
| 10 | ACS Catalysis | 8 | 1003 |
| 11 | Catalysis Science and Technology | 8 | 226 |
| 12 | Chemical Communications | 8 | 377 |
| 13 | Solar RRL | 8 | 46 |
| 14 | Chemistry - A European Journal | 7 | 695 |
| 15 | Chemsuschem | 7 | 107 |
| 16 | International Journal of Hydrogen Energy | 7 | 86 |
| 17 | Journal Of Catalysis | 7 | 315 |
| 18 | RSC Advances | 7 | 80 |
| 19 | Small | 7 | 592 |
| 20 | Advanced Materials | 6 | 579 |

waste treatment.^{19,42-43} This is because the modified g-C₃N₄ photocatalysts possess high visible light harvesting ability, efficient charge separation and transportation abilities, and high capacity for adsorption of pollutants for enhanced photocatalytic environmental performance.^{1,20}

The industrial wastes such as textiles dyes, effluents, etc. must be treated to meet the acceptable standard before being released into the environment. Thus, their released into the environment even in very low concentration without treatment possess health risk to both human and animals. But treatment of these industrial wastes as well as purification of surface and ground water for drinking either through a biological or chemical process is challenging and requires alternative approach (es). A lot of researches have shown that semiconductor photocatalysts is an excellent alternative method for treatment of low concentrated organic compound waste waters.⁴⁴⁻⁴⁶

The mechanism of photocatalytic environmental remediation involves important active species of photoexcited holes. These photoexcited holes are produced under light irradiation of the semiconductor photocatalyst in the valence band ($h\nu$) while the electrons are in the conduction band (e^-_{cb}) as shown in equation 1. The photoexcited holes are the most important oxidants reacting directly with the adsorbed organic pollutant molecules (equation 2) or water to generate hydroxyl radicals ($\cdot\text{OH}$) which has a strong oxidation potential of 2.8 eV (NHE)

(equation 3) which consequently oxidize the adsorbed organic pollutant molecules (equation 4). A reaction can also occur between the electrons and the organic pollutants to produce reduction products (equation 5). This mechanism has been discussed extensively in literature^{44-45,47-48} and a summary is shown below using g-C₃N₄ as an ideal photocatalyst. Furthermore, Table 6 shows the summary of some g-C₃N₄ based photocatalysts and their organic pollutant removal rates.

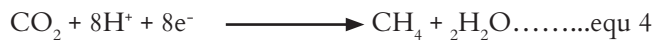
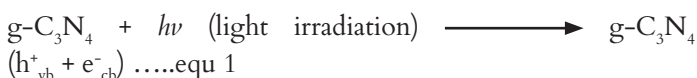


Photocatalytic reduction of CO₂ into Renewable Solar Fuels

One of the important sources of green renewable energy is solar fuels which can be obtained from photocatalytic conversion of CO₂ and water. This process is mimicking of the natural photosynthetic steps involved in the conversion of solar energy into chemical energy.¹³ The valuable hydrocarbon fuels that can be obtained from this process include methane (CH₄), methanol (CH₃OH), formaldehyde (HCHO), carbon monoxide (CO) among other hydrocarbon fuels as a sustainable energy source and production of chemicals. The mechanism involved in the CO₂ photocatalytic generation of solar fuels over a photocatalyst is a multi-electron transfer process which comprises generation of photoexcited electrons and holes. It is also involved in the breaking of C–O bond and formation of C–H and O–H bonds.⁵⁶ The photocatalytic process begins with the generation of photoexcited holes and electrons in the valence and conduction bands respectively by light irradiation on the photocatalyst as shown in equation 1. The molecular CO₂ is adsorbed on the surface of the photocatalyst and reacts with the photoexcited electrons. Based on the reaction kinetics and its thermodynamics, one CO₂ molecule would require 12, 10, 8, 6 and 4 photoelectrons for conversion to ethanol (C₂H₅OH), ethane (C₂H₆), methane (CH₄), methanol (CH₃OH) and formaldehyde (HCHO), respectively (equations 2-6).^{44,56} Furthermore, the photoexcited holes will react with adsorbed water on the surface of the photocatalyst to produce oxygen (equation 7). The summary of the CO₂ photocatalytic conversion into renewable solar fuels and hydrocarbon chemicals is shown using g-C₃N₄ as an ideal photocatalyst.

Table 6: Summary of some g-C₃N₄ based photocatalysts and their pollutant removal rate.

| Photocatalyst | Photocatalytic Experiment | Percentage Degradation or Removal | Ref. |
|--|---|---|------|
| Graphitic carbon nitride (g-C ₃ N ₄) hollow sphere | Photocatalytic degradation of Rhodamine B (RhB) in an aqueous solution under visible light irradiation (λ=4420 nm). | About 100 % concentration of RhB was decomposed after 180 min illumination | 49 |
| Activated carbon (g-C ₃ N ₄ /AC) | Photocatalytic AO7 dye degradation in aqueous solution activated by PMS degrade AO7 | 96.4 % of AO7 was removed within 10 min | 50 |
| Silver-decorated ultrathin g-C ₃ N ₄ nanosheets (Ag@U-g-C ₃ N ₄ -NS). | Photocatalytic degradation of MB and phenol under visible-light irradiation. | 88.5 % of MB degraded in 40 min under visible-light irradiation 89 % of phenol degraded in 80 min | 51 |
| 2D graphitic carbon nitride | A photocatalytic degradation of bisphenol A (BPA) was carried out. | 99 % of bisphenol A removal was achieved by exfoliated graphitic carbon nitride after 90 min | 52 |
| Oxygen-doped (O-doped) graphitic carbon nitride (g-C ₃ N ₄) nanosheets | A photodegradation of aqueous RhB solution under visible light (λ=400) nm irradiation was carried out. | 99 % degradation of RhB was achieved after 3 min | 53 |
| Sulfur doped g-C ₃ N ₄ microrods with silver chromate (g-C ₃ N ₄ /Ag ₂ CrO ₄) | A photocatalytic degradation of Methyl orange (MO), organic dye, was carried out under visible light (λ=420 nm) | 90.27 % degradation of MO was achieved with within 20 min | 54 |
| Pipe-like g-C ₃ N ₄ | Photocatalytic activity of was evaluated by the photodegradation of methylene blue (MB) under visible-light irradiation | 96 % degradation of MB within 30 min | 55 |



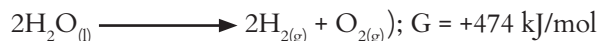
In addition, a summary of some of the various g-C₃N₄ based photocatalysts and solar fuels generated is shown in Table 7.

Photocatalytic Hydrogen Generation

One of the green energies been explored by the scientific community as alternative source of energy to fossil fuels and addressing the global energy crisis is hydrogen. Hydrogen can be generated photocatalytically by splitting water over a semiconductor photocatalyst. But the challenge facing this process for practical application is the large energy barrier required.^{42,44}

Generally, the photocatalytic hydrogen generation process is made up of three stages: (i) the first stage involved irradiation of the photocatalyst to generate electrons and holes (i.e. charge carriers) by the photons. (ii) in stage two, there is separation of the charge carriers and their migration to the surface of the photocatalyst. (iii) the stage three involves a reduction process of H⁺ to H₂ by the electrons and oxidation of H₂O to O₂ by holes.⁶²

For effective overall H₂ generation by water splitting, the photocatalyst should satisfy basic requirements such as the valence band potential must be more negative than H⁺/H₂ and more positive than OH⁻/O₂ potential. This standard is achieved by photocatalysts that have a minimum bandgap energy of 1.23 eV. However, practically water splitting process needed higher energy of 1.8 eV because of overpotential associated with the evolution reactions for hydrogen and oxygen. This makes the process very challenging under visible solar light for efficient water splitting with a single photocatalyst.⁶³⁻⁶⁴ However, composite photocatalysts with and without co-catalysts and hole scavenger have high hydrogen generation capacity⁴⁴ Generally, the overall reaction is:



The mechanistic stepwise reaction for the hydrogen generation process is summarize from equ. 1 to 4. The overall reaction redox potential at a pH = 7 is E_H = -1.23 eV (NHE).⁴⁴

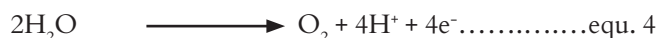
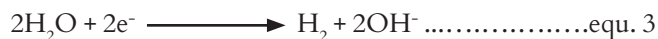
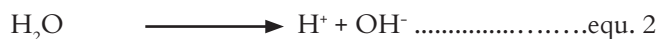


Table 8 shows a summary of g-C₃N₄ based photocatalysts with enhanced hydrogen generation rate.

Emerging Trends in g-C₃N₄ Photocatalysis

This scientometric evaluation indicated that they are new emerging areas where g-C₃N₄ based photocatalyst are being applied. In medicine, it is applied in drug delivery because it's

Table 7: Summary of some g-C₃N₄ based photocatalysts and solar fuel generation.

| g-C ₃ N ₄ based Photocatalyst | Photocatalytic Experiment | Enhancement factor over pure g-C ₃ N ₄ | Ref. |
|---|---|--|------|
| Noble-metal Pt nanoparticles and graphitic carbon nitride (Pt/CN) | Photocatalytic reduction of CO ₂ using water as a sacrificial reagent under visible light irradiation | g-C ₃ N ₄ (2Pt/CN) nanocomposites produced the highest CH ₄ yield of 13.02 μmol g catalyst ⁻¹ after 10 h of light irradiation, which was a 5.1-fold enhancement in comparison with pure g-C ₃ N ₄ | 57 |
| Potassium Hydroxide (KOH) on g-C ₃ N ₄ . | CO ₂ photocatalytic conversion in the presence of H ₂ O was conducted under visible light (λ > 420 nm). | 3-fold enhancement in CO ₂ photocatalytic reduction is achieved on KOH-decorated g-C ₃ N ₄ | 58 |
| Ultrathin Br-doped g-C ₃ N ₄ nanosheet | Photocatalytic reduction of CO ₂ under visible light (λ > 400 nm) irradiation for | CO ₂ photocatalytic reduction of 0.4 μmolh ⁻¹ g ⁻¹ of CH ₄ and 0.6 μmolh ⁻¹ g ⁻¹ of CH ₃ OH formation, which is 4.0 and 7.5 times higher than the pure g-C ₃ N ₄ NS. | 59 |
| Hybride of cobalt phthalocyanine tetracarboxylic acid and g-C ₃ N ₄ (g-C ₃ N ₄ /CoPc-COOH) | Photocatalytic CO ₂ reduction was performed in DMF/water/triethylamine system saturated with CO ₂ in the presence of visible light. | methanol yield using g-C ₃ N ₄ /CoPc-COOH was found to be 646.5 μmol. g ⁻¹ cat g-C ₃ N ₄ yielded 59.2 μmol (1184 μmol.g ⁻¹ cat) yield of methanol | 60 |
| Carbon nitride (g-C ₃ N ₄) shell encapsulating Cu ₂ O nanowire arrays/Cu mesh (g-C ₃ N ₄ /Cu ₂ O NAs/CM) | Photocatalytic CO ₂ reduction was carried out under visible-light | CH ₃ OH formation rate reached 22.6 ppm cm ⁻² h ⁻¹ | 61 |
| Honeycomb-like morphology (HC-C ₃ N ₄) | Photocatalytic CO ₂ reduction was carried out under visible-light using Ni(OH) ₂ as c cocatalyst | A CO ₂ photocatalytic reduction activity of 1.48 μmolh ⁻¹ g ⁻¹ of CH ₄ and 0.73 μmolh ⁻¹ g ⁻¹ of CH ₃ OH generation which is 3.5 and 4.3 times more than pure g-C ₃ N ₄ | 12 |

Table 8: Summary of some g-C₃N₄ based photocatalysts and hydrogen generation.

| g-C ₃ N ₄ based Photocatalyst | Photocatalytic Experiment | Amount of Hydrogen generated / Enhancement factor | Ref. |
|---|--|---|------|
| porous graphitic carbon nitrides (g-C ₃ N ₄) | The photocatalytic H ₂ evolution experiments were measured in a double-wall inner irradiation-using water and methanol as sacrificial agent | The photocatalytic hydrogen evolution under visible light is around five times higher if urea is used as the g-C ₃ N ₄ precursor instead of thiourea or melamine | 65 |
| Two-dimensional (2D) black phosphorus (BP) quantum dots (BPQDs) onto graphitic carbon nitride (BPQDs/g-C ₃ N ₄) hybrid | The photocatalytic hydrogen evolution was carried under visible light with Pt as a co-catalyst | 5 wt% BPQDs/g-C ₃ N ₄ with cyclability exhibits the greatest hydrogen evolution rate of 271 μmol h ⁻¹ g ⁻¹ that is 5.6 and 4.2 times greater than that of pristine g-C ₃ N ₄ and BPQDs | 66 |
| g-C ₃ N ₄ nanosheet | Photocatalytic water splitting reactions were carried out under visible light visible light (λ > 400 nm) | photocatalytic activity, which is 1.77 times than that of g-C ₃ N ₄ obtained via 3 h thermal treatment under visible irradiation | 67 |
| Hybrid of g-C ₃ N ₄ /free-base meso-tetrakis (carboxyphenyl) porphyrins | The photocatalytic experiments for H ₂ production were carried out at room temperature under visible light, (λ > 400 nm) using EDTA as sacrificial electron donor and Pt co-catalyst. | mTCCP-CN produced the highest amount of H ₂ evolved, 48.4 μmol under 6 h | 68 |
| Zirconium dioxide and graphitic carbon nitride (ZrO ₂ /g-C ₃ N ₄) composite | Photocatalytic hydrogen production was conducted under visible light using 500 W mercury lamp and methanol as sacrificial reagent with Pt as co-catalyst | The highest H ₂ production rate obtained for ZrO ₂ /g-C ₃ N ₄ -5% is 603 μmol/h and it is 20 times higher than pure ZrO ₂ . | 69 |
| WS ₂ /graphitic carbon nitride (CN) 2D/2D nanosheet heterostructure decorated with CdS quantum dots (QDs) (CdS/WS ₂ /CN) | The photocatalytic hydrogen evolution rates were measured under visible-light irradiation (λ =420–780 nm) for 5 h without any cocatalyst using triethanolamine (TEOA) as a sacrificial agent. | The CdS/WS ₂ /CN Photocatalytically evolved H ₂ 1174.5 mmolh ⁻¹ g ⁻¹ under visible-light irradiation which is nearly 67 times higher than that of the pure CN nanosheets | 70 |
| Two-dimensional/one-dimensional molybdenum sulfide (MoS ₂) nanoflake/graphitic carbon nitride (g-C ₃ N ₄) hollow nanotube (MoS ₂ /g-C ₃ N ₄ nanotube) | The photocatalytic test was carried out under visible light in aqueous solution (100 mL) containing 20% triethanolamine | MoS ₂ /g-C ₃ N ₄ nanotube composite with 15 wt% MoS ₂ shows the highest hydrogen (H ₂) production rate (1124 mmolh ⁻¹ g ⁻¹) much higher than bulk g-C ₃ N ₄ (64 mmol h ⁻¹ g ⁻¹) and g-C ₃ N ₄ nanotubes (189 mmol h ⁻¹ g ⁻¹) | 71 |

small size, covalent bonding which is non-interfering with the drug, low level of toxicity, and excellent optical property.⁴³ According to DFT evaluation carried out, g-C₃N₄ is an effective drug delivery material for cisplatin and carboplatin drugs for treatment of various cancers.³²⁻³³ In practice, g-C₃N₄ quantum dots (g-CNQDs) was synthesized and incorporated into diamine-terminated oligomeric poly(ethylene glycol) (PEG) resulting in a high intensity blue-photoluminescent PEGylated g-CNQDs. The g-CNQDs-PEG was assessed to be a perfect drug delivery system for cancer (especially anti-cancer drug doxorubin).⁷² Furthermore, a study on interaction of pyrazinamide and 5-fluorouracil anticancer drugs and bulk C₃N₄ with folic acid conjugation as shows a suitable combination for anticancer drug delivery.⁷³ Similarly, it has been shown that g-C₃N₄ can effectively mediate cancer therapy; both is *in vitro* and *in vivo* using Photodynamic therapy (PDT).⁷⁴

In the mathematical science, g-C₃N₄ was used as an ideal material to compute topological index of the molecular structure. This has enhance the researcher's knowledge and understanding of the physical science and bioorganic attributes of the g-C₃N₄ for further application in graph theory.⁷⁵ Similar, the M-polynomial and NM-polynomial with graphical representations of g-C₃N₄ was computed as topological indices to be used in determination of pharmaceutical properties of drugs.⁷⁶

Currently, other disciplines where in-depth researching is being carried out and utilizing g-C₃N₄ application included disease and toxin treatment, computer application systems, food safety monitoring, drug and biosensor detection.

Conclusion and Future Research Outlook

This study attempted to outline the progress and hotspots areas using a scientometric assessment in the field of photocatalysis and g-C₃N₄ for CO₂ photocatalytic reduction. A lot of articles and reviews have been reported on g-C₃N₄ showing its uniqueness and potential applications. However, this scientometric study explore the current trends and status revealing the emerging nature of g-C₃N₄ over relatively short period. In all, a total of 504 articles were collected from the Scopus database of Web of Science (WOS) spanning from 2010 to May, 2021. The scientometric techniques employed in the assessment are documents types and their numbers, research subject areas, trend analysis of publications, keyword co-occurrence analysis and clustering, co-occurrence analysis (i.e. for countries, institutions, and authors), authors and citations and documents and co-citations of journals. In this study, VOSviewer software was employed to map and visual the various scientometric techniques. Research articles (i.e. 390) dominated the documents recorded with Chemistry being the main area. There is a progressive increase in published

documents over the years with the keyword "Photocatalysis" having the highest holding frequency of 240 suggesting it is a current hotspot area.

Regarding contributions in the field, Zhang X. from China is identified to be the lead productive researcher with 27 articles. There are some levels of international collaborations between the researchers and China emanating as the lead productive country in the field. Ong W. J. obtained the top researcher with the highest citation frequency of 2986 and Applied Catalysis B: Environmental journal ranked top for publishing in this area of study.

The g-C₃N₄ photocatalytic materials have been applied in a lot of areas including pollutant degradation, renewable solar fuels generation and water splitting into hydrogen and oxygen. This scientometric assessment is an evidence that there is a lot of scientific progress being made in the field of photocatalysis and g-C₃N₄ semiconductor material through the visualization of the trends and hotspots. In addition, the study provides important information for the scientific community, researchers, policy makers and practitioners in the photocatalysis field to develop more excellent materials from g-C₃N₄ for various fields of application.

Consequently, a lot of efforts have been directed to the fabrication of various g-C₃N₄ based photocatalysts and their applications in environmental remediation, energy generation, waste disinfection, water splitting among others. However, this scientometric assessment shows that there are other potential areas for application of g-C₃N₄ photocatalyst based materials including medicine, biomedical systems, pharmacy and toxicological industries. In addition, other multidisciplinary fields such as computer science, mathematics, earth science etc. could also derive benefits from the application of a g-C₃N₄ material. Thus, further research in these areas must be strengthened by the scientific community.

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CONFLICT OF INTEREST

The authors declare no conflict of interest.

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