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Review

Construction Sector Transition towards Smart Applications of Graphene Oxide in Cement-Based Composites: A Scientometric Review and Bibliometric Analysis

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Abstract: Cement-based composites (CBCs) are essential in the construction sector due to their cost-effectiveness, availability, and versatility, but they struggle with low tensile strength and poor heat resistance. Recent advancements have highlighted the potential of nanomaterials, particularly graphene oxide (GO), in enhancing the mechanical, thermal, and electrical properties of CBCs. This study aims to provide a comprehensive review of the incorporation of GO into cementitious composites, examining its impact on microstructure, mechanical properties, rheology, and durability; thus, a bibliometric review and scientometric analysis were conducted to thoroughly evaluate the existing literature. A total of 263 studies were selected for thorough study. It can be concluded that GO content acts as a pore filler, decreasing porosity by 23% and average pore size by 22%, while boosting compressive strength by up to 15% at a 0.05% concentration. It also enhances workability, stability, and resistance to chloride ingress, sulfate attack, alkali–silica reaction, and carbonation. Incorporating GO reduces cement consumption and carbon footprint, leading to more durable structures and supporting sustainable construction by efficiently utilizing waste materials. The optimal GO concentration for these benefits ranges from 0.03% to 0.1% by weight of cement, as higher concentrations may cause agglomeration. GO-modified cementitious materials are well suited for high-performance and durable applications, particularly in environments with chemical and mechanical stresses.

Keywords: graphene oxide (GO); nanomaterials; cement-based composites (CBCs); construction projects; smart material

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1. Introduction

Cement-based composites (CBCs) are widely used in the construction sector due to their cost-effectiveness, easy availability, and great flexibility. As society has progressed, the architectural requirements for these materials have become increasingly complex. The intrinsic limitations of CBCs, including low tensile strength and poor heat resistance, may no longer match the requirements for structural durability and could jeopardize the overall service performance of the structures in this context [1]. Nanomaterials have attracted considerable interest because of their distinctive properties. Nanoscale phenomena include the small size effect, quantum tunneling effect, and surface effect, among others. Many researchers have studied the integration of nanomaterials with concrete building components to improve their mechanical characteristics, longevity, and adaptability [2].

Cement is the principal adhesive component in concrete, characterized by a low resistance to tension and a brittle quality. Combining fibers and steel reinforcement with CBCs helps to address the above shortcomings to some extent. Nevertheless, the advancement of nanoparticles and their application in CBCs has enhanced the product's strengths, including flexural, toughness, durability, and tensile qualities [3,4]. Nevertheless, scientists are currently examining the efficacy of nanomaterials in concrete, as various nanomaterials have diverse impacts on the characteristics of concrete, including the most cost-effective dosage and other relevant factors [5]. Table 1 provides a comprehensive overview of the nanomaterials used in concrete, including zeolite, nano clay, carbon nanotubes (CNTs), carbon nanofibers (CNFs), graphene oxide (GO), nano fly ash, ferric oxide (Fe_2O_3), aluminum oxide (Al_2O_3), zirconium dioxide (ZrO_2), titanium dioxide (TiO_2), and silicon dioxide (SiO_2). The aforementioned table also includes the reported properties of these nanomaterials. It is evident that the majority of nanomaterials enhance the rheology, mechanical, microstructure, and durability characteristics of concrete.

Table 1. Overview of documented characteristics of nanoparticles in concrete.

Nanomaterial	Rheology	Shrinkage	Mechanical	Heat of Hydration	Microstructure	Durability
Zeolite	√	√	√	-	√	√
SiO_2	√	-	√	√	√	-
Nano clay	√	-	√	-	-	-
CNT	-	√	√	√	√	√
CNF	-	-	√	-	√	-
GO	-	-	√	-	√	-
Zeolite	√	√	√	-	√	√
Nano fly ash	-	-	√	-	-	-
Fe_2O_3	√	-	√	-	-	√
Al_2O_3	√	-	√	√	-	√
ZrO_2	-	-	√	-	-	√
TiO_2	√	-	√	√	-	√

Nanomaterials, such as graphene, CNTs, GO, and graphene-based nanomaterials, have improved various attributes of cement-based composites, including mechanical strength, optical transparency, electrical conductivity, and thermal conductivity [6,7]. GO, which stands for graphene oxide, is a nanomaterial made of carbon that has two dimensions. It has the ability to react with cementitious composites, adding another level of reactivity. GO transforms cementitious composites on a small scale to a larger size, improving their durability, mechanical qualities, and multifunctional characteristics [8].

GO's strong bonding with oxygen functional groups and its heightened reactivity with cement composites have contributed to its increased popularity as a derivative of graphene [9,10]. Nevertheless, the rise in oxygen concentration caused by the existence of oxygen functional groups in GO may lead to an increase in structural flaws, resulting in a decrease in the electrical and thermal conductivity of GO [11]. Thus, in order to mitigate the conductivity limitations noted earlier, GO is converted into reduced graphene oxide (rGO) [12]. rGO is derived from GO by a two-step covalent modification procedure and demonstrates enhanced water dispersion in comparison to GO [13]. The primary oxygen functional groups are mostly identified as epoxy and hydroxyl groups, with lesser quantities of carbonyl, carboxyl, lactone, quinone, and phenol groups [13,14]. Figure 1 illustrates the process of transforming graphene oxide (GO) into reduced graphene oxide (rGO) by structural rearrangement.

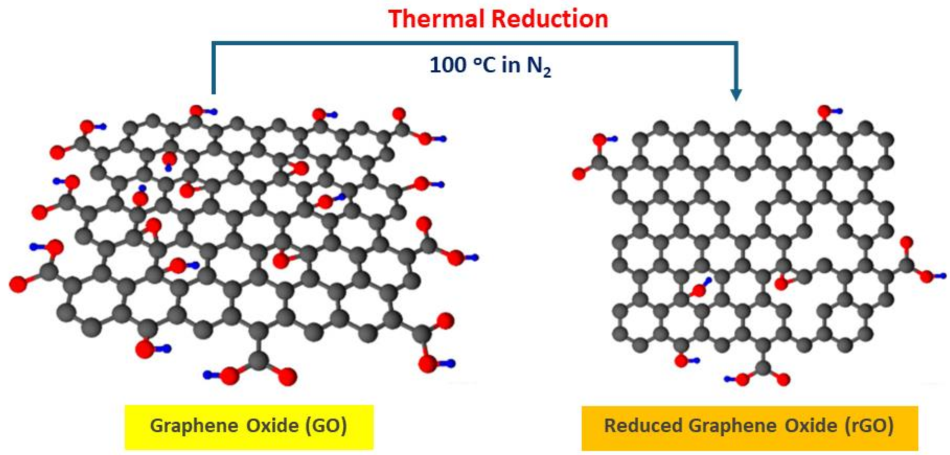

Figure 1. Structural formation of rGO.

In 1859, Brodie [15] created the initial manually produced specimen of graphene oxide by chemically separating graphite. However, this method was hazardous and resulted in a subpar product [16]. Subsequently, Hummers Jr and Offeman [17] presented an enhanced and secure procedure for producing a superior and effective product, commonly referred to as the Hummers method [18,19].

Graphene oxide has a similar atomic structure to other compounds based on graphene. However, its covalent bonds are broken through chemical processes in order to attach functional groups such as epoxy, phenol, carbonyl, and others [20,21]. In addition, the discovery of graphene in 2004 [22] marked a significant milestone in the utilization of nanotechnology in cementitious materials [23].

Graphene is a highly adaptable substance and is regarded as the progenitor of all other kinds of graphene. Graphene, a nano-filler, is considered an optimal choice for cementitious goods. However, it is expensive, and the method of producing it is intricate. Nevertheless, the practicality of utilizing multilayer graphene nanoplatelets (GNPs) is high due to their easy production from GO or graphite [24,25] . Incorporating graphene oxide (GO) with cementitious materials has yielded remarkable results, establishing GO as a widely used nano-filler. Both GO and GNPs are generated from graphene and possess exceptional mechanical characteristics. Nevertheless, the dispersion capacity of GO is superior in composites due to its hydrophilic functional groups, compared to GNPs. Additionally, GO exhibits distinctive electrical and thermal conductivity properties [26,27]. Furthermore, carbon nanotube (CNT)-containing compounds are more costly in comparison to graphene oxide (GO) compounds, which are significantly more affordable. GO is gaining popularity due to the aforementioned advantages over other materials [28]. Figure 2 depicts the chemical structure of graphene derivatives.

The utilization of GO in cementitious mixtures, such as concrete, is an emerging and swiftly expanding area of research. Although it has gained popularity in recent years, there have been limited research investigations conducted in this area [29]. Recent research has extensively explored the integration of GO into cementitious composites such as mortar and concrete. Scholars have studied the use of graphene oxide in both powdered and water-dispersed forms in cement-based products. The impact of adding GO on the initial setting, final setting, and compressive strength characteristics of cement mortar has been extensively studied. Significant focus has been given to incorporating GO nanoparticles into building materials, representing notable progress in the study of GO-fiber-reinforced concrete [30]. This study aims to gain a comprehensive understanding of the incorporation of graphene oxide in CBCs for its smart applications in the construction sector. This study conducted a bibliometric assessment and scientometric analysis of published data on cementitious composites enhanced with GO. The results of this study will aid in identifying the prominent issues studied, influential sources of publication, institutions, productive scholars, and countries. In addition, a thorough examination of the practical uses of smart GO-CBCs will aid the reader in understanding the current research focus and identifying areas where more study is needed. One of the study goals is to provide insights for readers and researchers regarding the current status of the construction industry in utilizing GO as a composite building material. By elucidating this context, the research will assist the academic community in effectively identifying and planning future research directions in this area. This will also provide recommendations for future prospects in order to fulfill the demands of contemporary construction. Moreover, it analyzes temporal patterns in research activities, highlighting emerging fields and declining areas relevant to GO-CBCs in the domain of the construction industry.

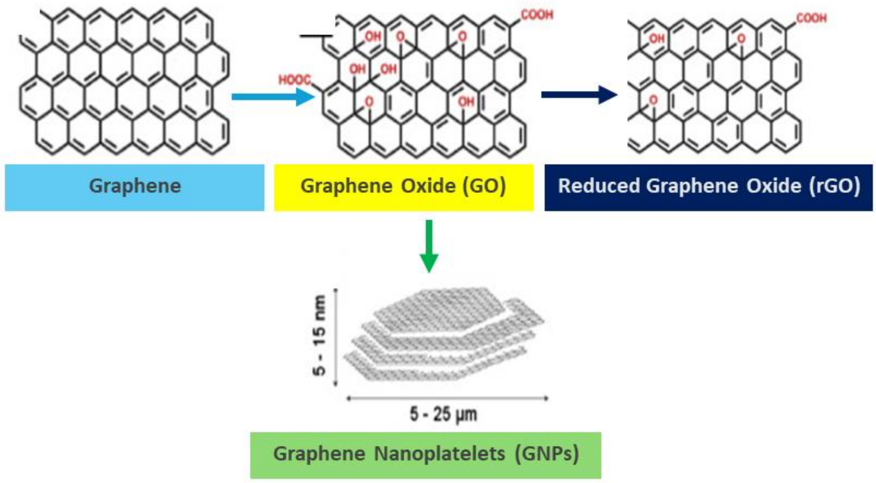

Figure 2. Chemical illustration of graphene derivatives.

2. Methodology

This study utilized bibliometric review and scientometric analysis approaches. Bibliometrics, informatics, and scientometrics are three interconnected metric terminologies that are often misunderstood. Bibliometrics, informatics, and scientometrics share similar approaches, ideas, applications, and technologies. However, they differ in their topic backgrounds [31,32]. The bibliometric technique is a vital statistical instrument used to identify and analyze the current areas of scientific knowledge, as well as to emphasize the relevant information on future prospects and supporting research [33]. The purpose of bibliometric analysis is to visually represent selected data and create scientific networks to analyze themes such as keywords, journals, and authors [34]. The scientometric analysis is used to quantitatively analyze and appraise the contents of publications [35]. Unlike scientometric and bibliometric reviews, which focus on the measurement and analysis of specific data, informetric analysis aims to extract additional information from the literature that is not explicitly stated. This is achieved through the use of quantitative methods in mathematics and statistics [36]. The study employed bibliometric review and scientometric analysis approaches based on the scope and theme of the proposed research. In addition, the research plan was developed in accordance with the existing recommendations for bibliometric and scientometric analysis [33,37].

The objective of this work was to examine intelligent implementations of GO in CBCs. The data were gathered for the research by utilizing the "Web of Science" (WOS) database. WOS is a significant database that contains bibliographic references for scientific investigations conducted since 1945. It includes information on journals, books, and conferences [38]. The articles' information was collected using two keyword strategies for data collection. For the first search, the timespan was set until 2024 and the keywords "Graphene Oxide" and "Concrete". The WOS database's advanced search tool was employed, and the query was constructed using a combination of tags, parentheses, and Boolean operators. Specifically, the query used the following format: TS = ((*graphene* AND oxide) AND concrete), where TS is the abbreviated term for the topic in WOS. The search was restricted to English language papers, specifically technical research, within the indexes of Science Citation Index Expanded (SCIE), Social Science Citation Index (SSCI), and Emerging Sources Citation Index (ESCI). The WOS database identified a total of 206 studies within the stated scope. However, for the second search using the same database and timespan, additional keywords, i.e., smart, intelligent, and engineered, were added to the previous keywords query, modifying the query as TS = ((*graphene* AND oxide) AND concrete AND (smart material* OR intelligent material* OR engineered material*)). This led to the identification of 100 studies under a defined scope by the WOS database. The main purpose was also to assess the commencement of the trend in smart GO applications compared to its regular applications. Figure 3 illustrates the comprehensive framework of the research strategy employed in the present study.

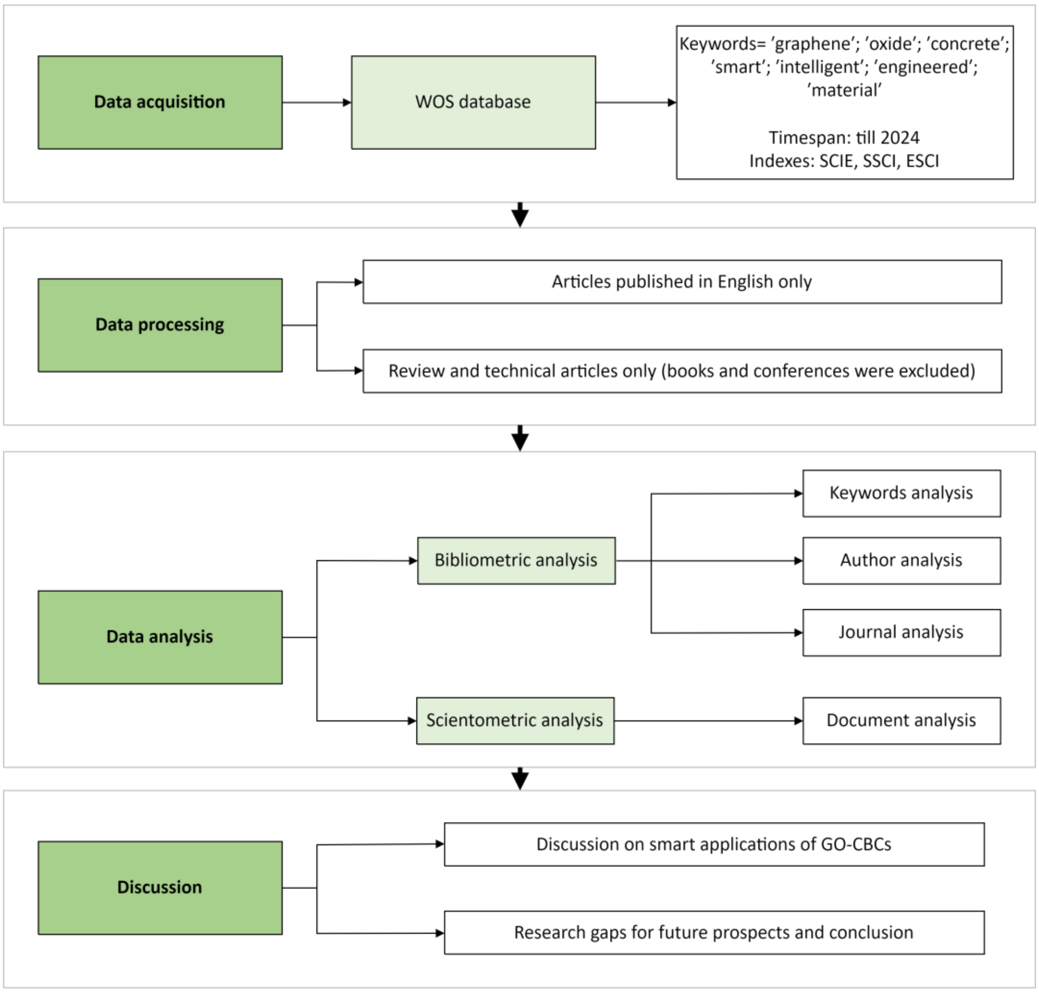

Figure 3. Research strategy framework.

3. Data Analyses

GO, a novel carbon-based nanomaterial [8], has attracted significant attention from researchers because of its unique characteristics when combined with cementitious composites. The purpose of this study was to identify the important themes related to the applications of GO in cementitious composites. Additionally, the study aimed to identify the influential sources of publication, institutions, scholars, and countries that are actively researching in this area. The study also aimed to highlight any research gaps and provide recommendations for future research to meet the requirements of modern construction. The data were gathered for the research by utilizing the WOS database.

Overall, 306 studies were found. However, these data were scrutinized, and six articles were discarded for being common/repeated. Moreover, collected data were also screened based on the articles' titles and abstracts, and 37 articles were eliminated for being out of this study-defined protocol and scope. Nevertheless, 263 articles were found fit for further analyses and in-depth review. The year-wise distribution of the aforementioned 263 articles is shown in Figure 4.

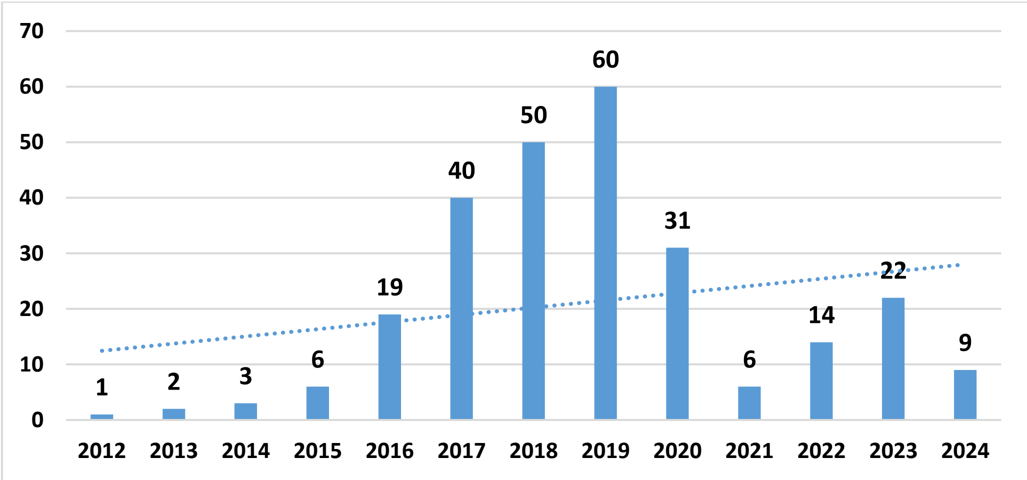

Figure 4. Year-wise distribution of articles.

It was observed that the proper work on GO commenced in 2012, whereas the first article on GO as a smart material was published in 2014 by Saafi et al. [39]. However, in 2016, the concept of GO as a smart material gained interest among researchers. The highest number of publications was found for 2019 (60 articles), and the trend was observed to drop in 2020 and 2021, possibly due to COVID-19 (work-from-home restrictions). However, there has been a noticeable increase in the use of GO in cementitious composites since 2022.

3.1. Keyword Analysis

Keyword analysis provides a concise overview of the scientific literature by emphasizing the main content. This analysis provides a comprehensive overview and visual representation of the research areas within the specified domain [33,37]. For this study, keyword co-occurrence analysis was performed, stated as "The number of co-occurrences of two keywords is the number of publications in which both keywords occur together in the title, abstract, or keyword list" [40]. However, in WOS, other than "author keywords", the database also provides additional keywords against each paper under the category "Keywords Plus", which reflects the additional information about the selected paper. For this analysis, both author keywords and keywords plus were collectively analyzed. The examination of keywords was conducted using the scientometric analytic software VOSviewer 1.6.20 , by setting the minimum threshold for the number of occurrences of a keyword to five. A total of 1276 keywords were found from 263 publications, of which 101 keywords met the criterion. For this analysis, the keywords "graphene oxide", "graphene", "oxide", "performance", "behavior", and "paste" were unselected for better analysis. The purpose was to analyze the data without using generic terms and terms related to graphene. Moreover, the keywords "mortar" and "cement paste" were converted to "cement" for analysis as all three terms reflect the same thing. Figure 5 displays the mapping of keyword co-occurrence for 263 recognized articles. Table 2 provides an overview of the top 10 keywords, including their occurrence rate and the average publication year. The average publication year is the average of the years in which the specific phrase appeared in publications . The keyword mapping network consisted of 101 discovered keywords, which were divided into seven clusters. Each cluster represents an intermediate correlation network. The size of the word cloud and the font size are determined by the frequency at which the phrase appears in the articles [41]. However, it can be seen that "concrete", "mechanical properties", and "microstructure" are the top three keywords with an occurrence rate of 148, 131, and 125, respectively.

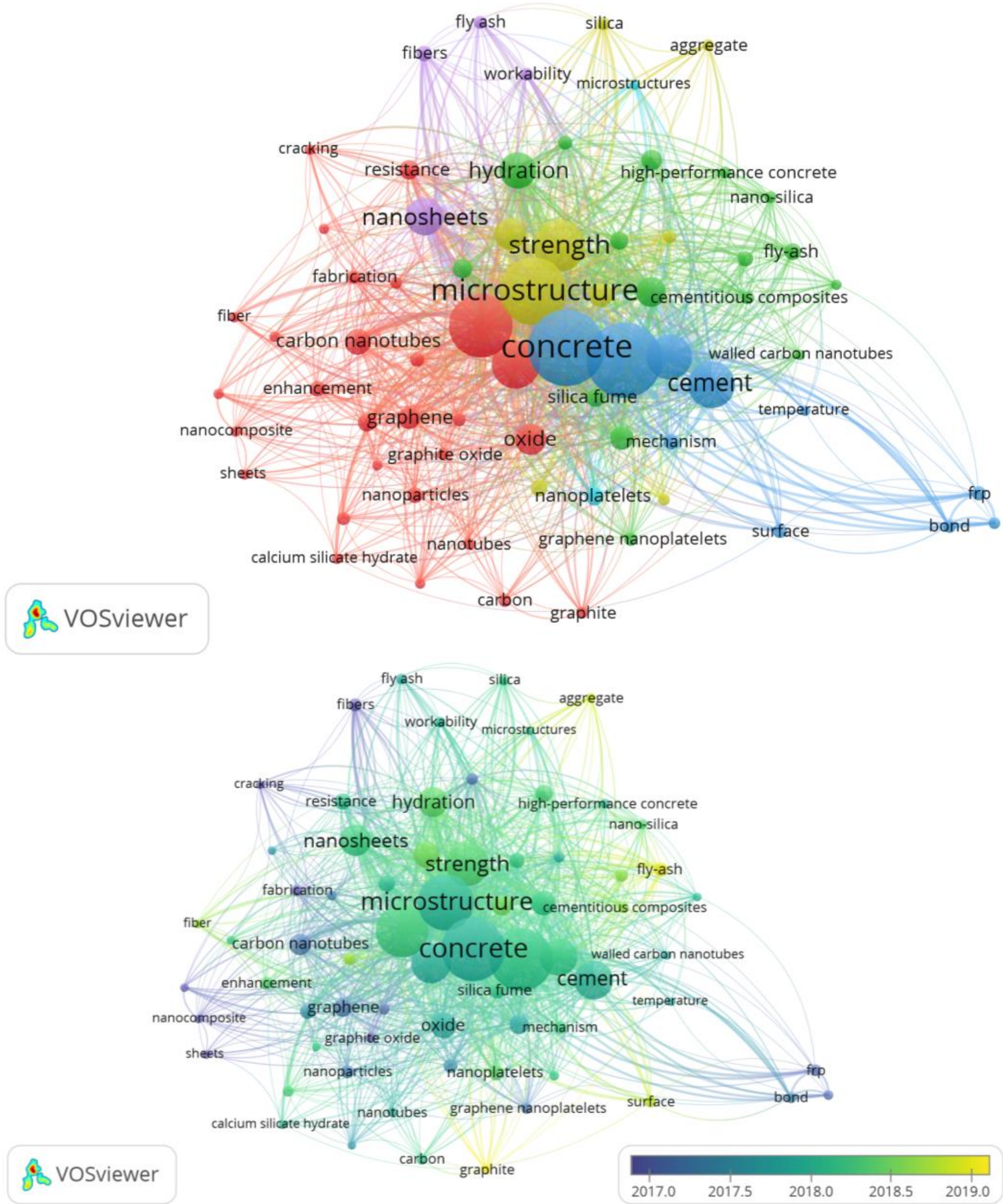

Figure 5. Keyword co-occurrence analysis mapping.

Table 2. Summary of top 10 keywords' co-occurrence analysis.

Keyword	Occurrences	Average Publication Year
concrete	148	2019
mechanical properties	131	2019
microstructure	125	2019
strength	92	2019
composites	72	2019
cement	67	2019
hydration	51	2019
nanosheets	46	2019
dispersion	45	2020
reinforced cement	27	2019

Regarding the properties of cement composites reinforced with GO, the most commonly occurring features were "mechanical properties" (occurrence rate 131), "microstructure" (occurrence rate 125), and "strength" (basically compressive strength) (occurrence rate 92). Moreover, these features of cementitious composites reinforced with GO were followed by "durability" (occurrence rate 25), "rheological properties" (occurrence rate 19), "self-sensing" (occurrence rate 8), "mechanical strength", and "transport properties" with an occurrence rate of 7. However, the least occurring properties were "cracking" and "permeability" with an occurrence rate of 5.

3.2. Author Analysis

The author's analysis provides insight into the co-citation and collaboration of writers in scientific research, with the aim of achieving a common purpose [42]. This study conducted two primary analyses, namely collaboration analysis and co-citation analysis.

3.2.1. Author Collaboration

Collaboration analysis is a method used to examine the connections and relationships between researchers and institutions. This identification could facilitate access to financial opportunities, expertise, and the exchange of research experiences [29].

In this VOSviewer analysis, the author's minimum document count was set to three. In total, 1003 authors were identified, of which 62 matched the specified criterion. Figure 6 depicts the collaborative network among authors based on specific criteria. In summary, the authors' collaboration network was divided into two clusters. The most active collaborative group consisted of Yuan Gao, Weiqiang Chen, Jiangyu Wu, and Hongwen Jing. Yuan Gao primarily focused on enhancing the performance and characteristics of cemented waste rock backfill [43–46].

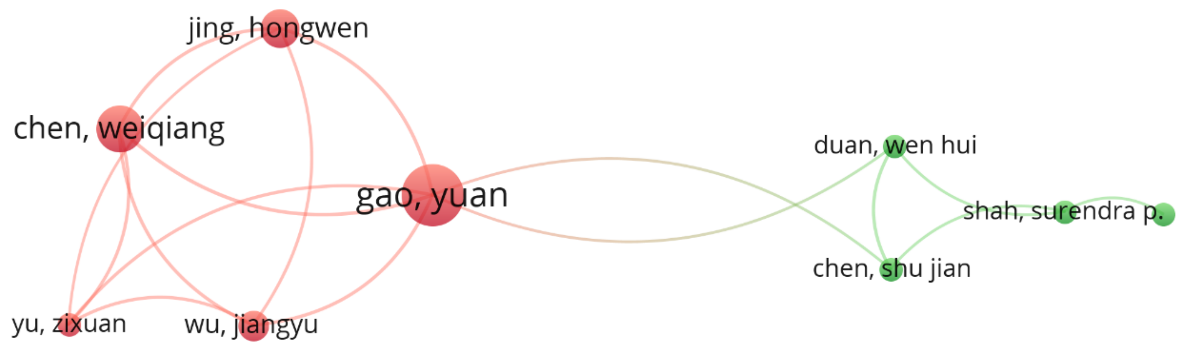

Figure 6. Authors' collaboration analysis.

In addition, the analysis of partnership was conducted for institutions and countries, with a minimum need of five documents. Figure 7 displays a collaboration network across universities, with a total of 339 institutions found, 20 of which met the specified criteria. It is evident that four clusters were created, indicating the presence of collaborative networks within the topic domain. The top five most productive institutes identified are "Southeast University" (with 16 published articles), "Swinburne University of Technology" (with 16 published articles), "Harbin Institute of Technology" (with 12 published articles), "University of Baghdad" (with 12 published articles), and "Monash University" (with 12 published articles).

Furthermore, Figure 8 illustrates the network of countries collaborating, with a total of 34 countries discovered. Out of them, 20 countries satisfied the threshold requirement of having at least three papers. Overall, three clusters were established, with the most robust collaboration network seen for (1) China, England, and the United States of America, and (2) Australia and Iraq were also connected by strong network links . China, the USA, and Australia were the leading countries in terms of contributions to the field of GO applications in cementitious composites, with 157, 36, and 31 published publications, respectively.

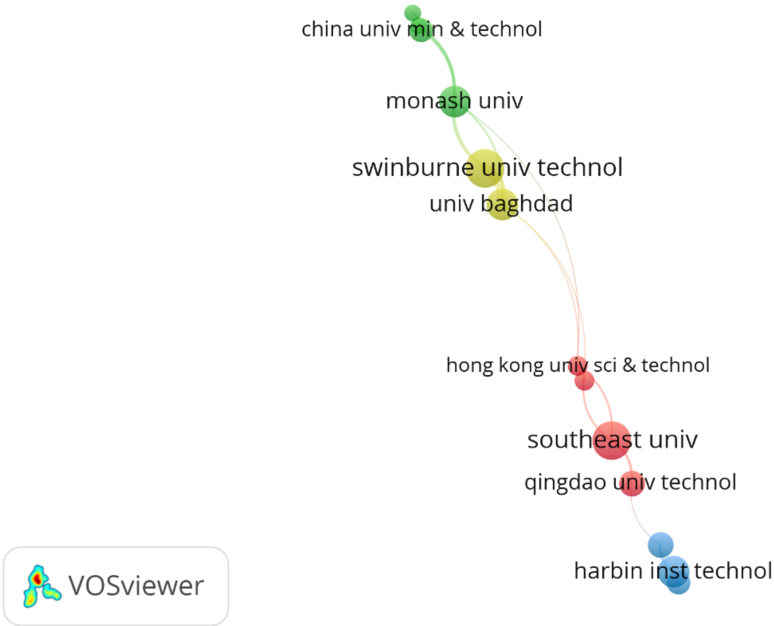

Figure 7. Collaboration network of institutions.

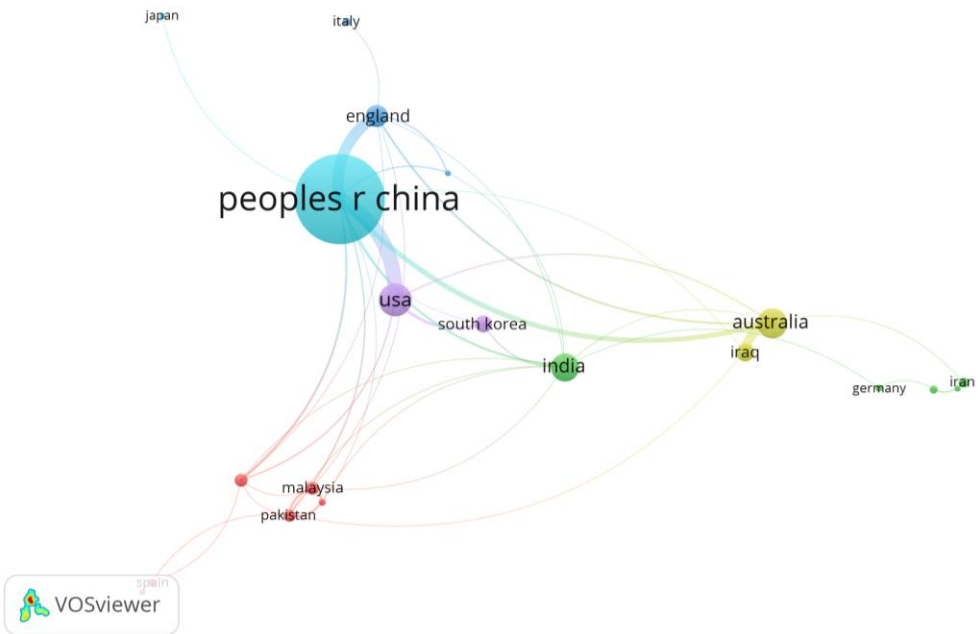

Figure 8. Collaboration network of countries.

Table 3 presents a comprehensive breakdown of the top five authors, including their institutional information, nations, and the highest number of articles they have collaborated on.

Table 3. Top five authors with the most collaborated articles.

Author	Institute	Country	Publications
Feng Xing	Shenzhen University	China	9
Wu-Jian Long	Shenzhen University	China	9
Yuan Gao	Nantong University	China	8
Xianming Shi	Washington State University	USA	8
Alyaa Mohammed	Swinburne University	Australia	8

3.2.2. Author Co-Citation

The technique of author co-citation analysis is used to determine the connections between writers who have been mentioned in the same publication [37]. In this VOSviewer analysis, the minimum number of citations required for an author to be included was set at 20. Out of a total of 6543 writers discovered, only 68 met this requirement. Author co-citation analysis solely considers the primary author of the cited document. Figure 9 displays the co-citation network created by the author.

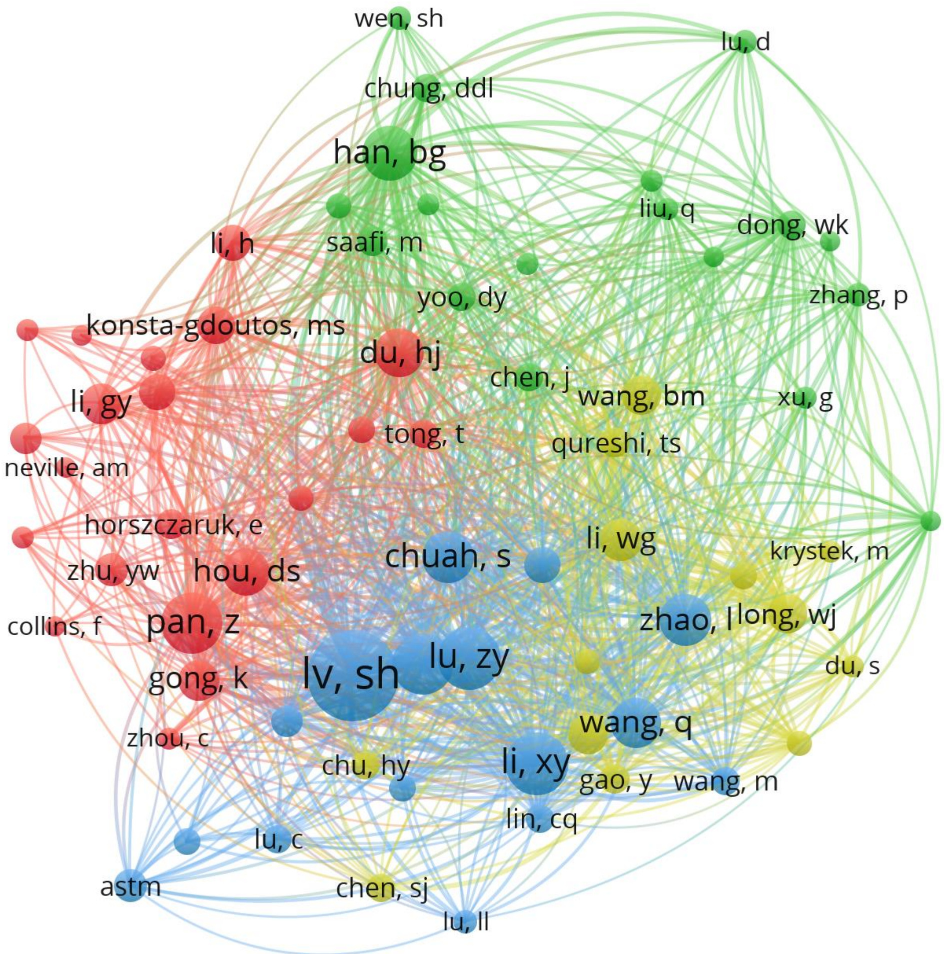

Figure 9. Author co-citation network.

Table 4 displays the 10 writers with the highest number of citations in the obtained data. The gathered research revealed that Shenghua Lv, Xiang-Ying Li, and Zeyu Lu were the three authors with the largest number of citations, with 227, 123, and 121 citations, respectively.

Table 4. Top ten cited authors.

Author	Citations	Country
Shenghua Lv	227	China
Xiang-Ying Li	123	China
Zeyu Lu	121	Hong Kong
Zhu Pan	119	Australia
Alyaa Mohammed	117	Australia
Baoguo Han	101	USA
Samuel Chuah	91	Australia
Li Zhao	89	China
Qian Wang	84	China
Hongjian Du	80	Singapore

3.3. Journal Analysis

The journal analysis facilitates the discovery of leading journals that publish top academic studies, enabling scholars to comprehend the latest growing trends and advancements in the field [47]. Journal analysis was conducted based on two specific criteria: (1) Journal contribution refers to the journal that is making the greatest contribution in terms of research output or influence. (2) Journal co-citation refers to the journal that is being cited the most by other journals. The previous analysis involved examining data from 263 publications to identify the top journals that are making the greatest contributions within the specific topic of this study. The subsequent co-citation analysis was performed utilizing the comprehensive data of references acquired from these 263 articles.

Table 5 presents a summary of the study of journal contributions, specifically focusing on the top five journals. Among the 109 journals discovered in the collection of 263 articles, *Construction and Building Materials* stands out as the most influential journal, with a total of 66 articles. Furthermore, the citation number of these journals has been specified, indicating the frequency at which these 109 journals were cited among the 263 papers collected.

Table 5. Contribution of journals.

Journal	Publisher	Citations	Documents	Percentage
Construction and Building Materials	Elsevier	1001	66	25%
Cement & Concrete Composites	Elsevier	136	14	5%
Materials	MDPI	46	11	4%
Nanomaterials	MDPI	175	10	3%
Journal of Building Engineering	Elsevier	103	9	3%

A journal co-citation study was conducted, with a minimum citation requirement of 40. Out of a total of 2320 journals, only 55 journals satisfied this criteria. Figure 10 displays the results of the journal co-citation analysis conducted on the 263 collected publication references.

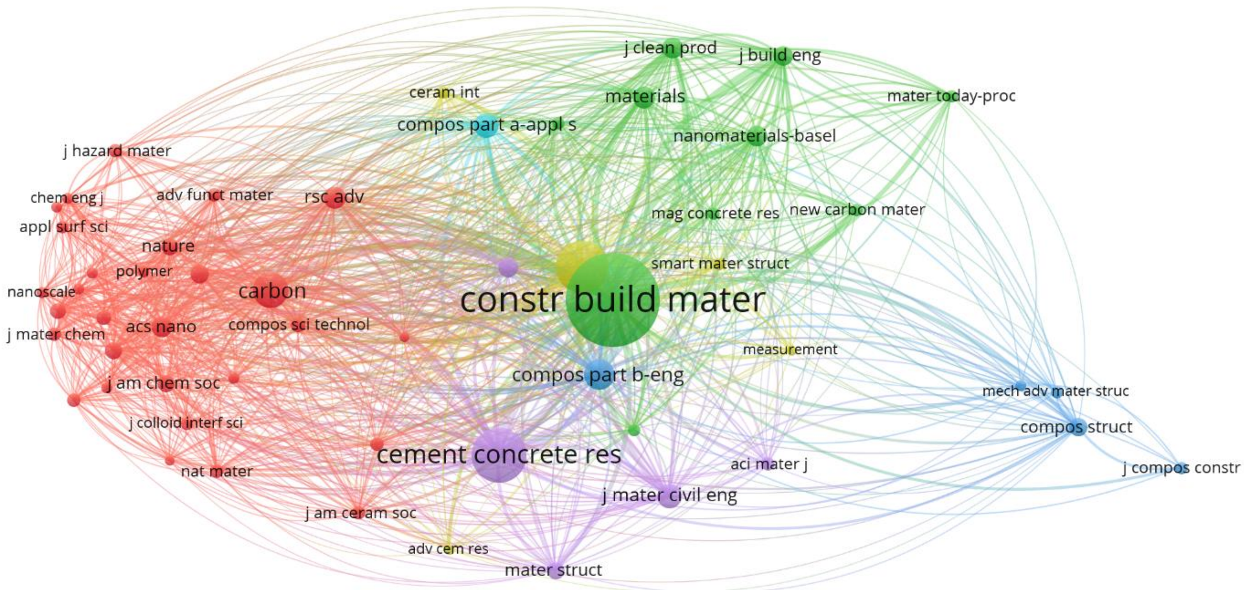

Figure 10. Journal co-citation analysis mapping.

The analysis determined that the journal *Construction and Building Materials* had the highest number of citations, with a total of 2609. It was followed by *Cement and Concrete Research* with 931 citations, *Cement & Concrete Composites* with 855 citations, *Carbon* with 411 citations, and *Composites Part B: Engineering* with 300 citations.

3.4. Document Analysis

Document co-citation is a technique used in document analysis to measure how often two articles are cited together within other documents [48]. In VOSviewer, the minimum criterion for the number of citations for a referenced document was established as 25. This led to a total of 9708 cited references, out of which 28 met the established criteria. Figure 11 displays the co-citation mapping of the documents, while Table 6 provides an overview of the top five most-cited articles. It can be observed that the papers by Pan et al. [7], Lv et al. [49], and Chuah et al. [6] were the three most frequently referenced.

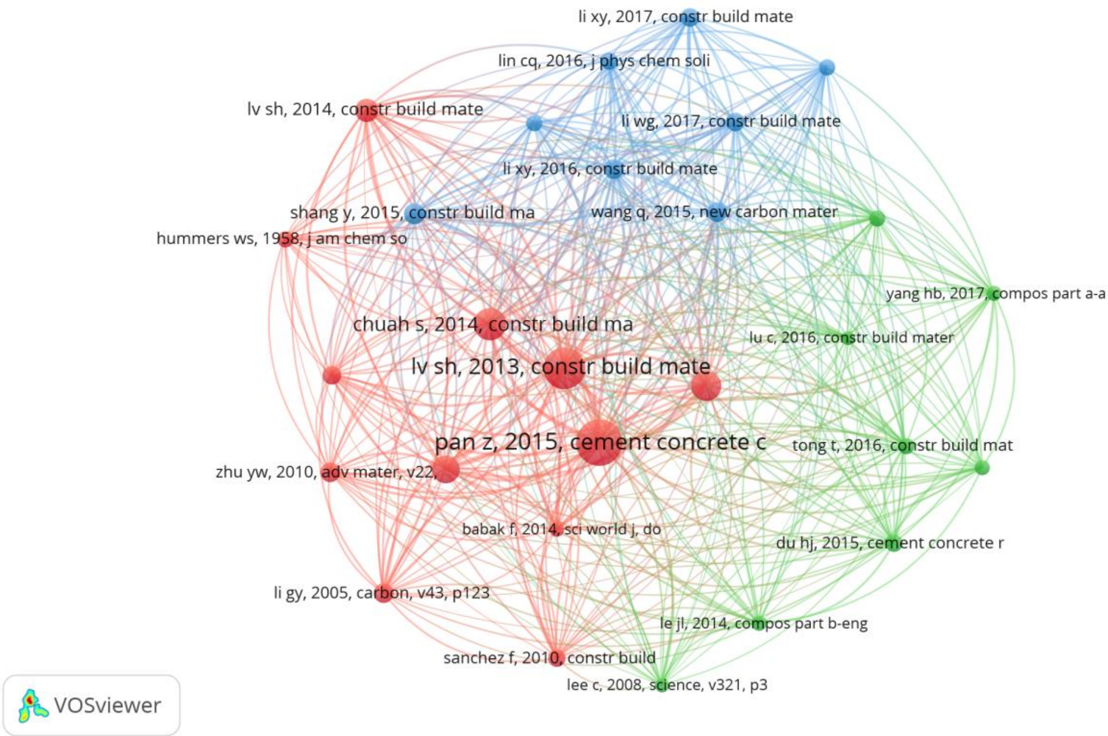

Figure 11. Document co-citation analysis.

Table 6. Top five most cited publications.

Title	Citations	Reference
"Mechanical properties and microstructure of a graphene oxide–cement composite"	107	[7]
"Effect of graphene oxide nanosheets of microstructure and mechanical properties of cement composites"	94	[49]
"Nano-reinforced cement and concrete composites and new perspective from graphene oxide"	70	[6]
"Incorporating graphene oxide in cement composites: A study of transport properties"	63	[50]
"Reinforcing effects of graphene oxide on portland cement paste"	58	[51]

4. Results and Discussion

This study sought to comprehend the current research state regarding the utilization of GO in cementitious composites. Additionally, it tried to identify the prevailing research theme in this field in order to highlight areas where further information is needed. The integration of GO into cementitious composites has garnered significant attention in scientific research, owing to its potential to augment diverse properties of these materials. Cementitious composites, notably concrete, play a pivotal role in construction due to their robustness, resilience, and adaptability. The incorporation of nanomaterials like GO holds promise for bolstering the mechanical, thermal, and electrical characteristics of these composites. Studies incorporating the interaction of GO within cementitious composites have explored several key aspects.

4.1. Environmental Perspective of Using Graphene Oxide (GO)

Numerous studies have shown that GO can act as an eco-friendly material in the formation of different cement composites. GO is produced by oxidizing natural graphite, using methods such as Brodie's [15] and those in Staudenmaier [17,52,53]. Although Hummers' method is commonly used, it has limitations like low yield, toxic gas emissions (NO_2 and N_2O_4), and residual $N_a{}^+$ and $NO_3{}^-$ ions in wastewater. To lower costs and environmental impact, more sustainable GO synthesis methods have been developed. For example, J. Chen et al. [54] found that graphite can be oxidized to GO using concentrated H_2SO_4 with $KMnO_4$, an improved version of Hummers' method that avoids using $NaNO_3$, thus reducing toxic gas emissions and simplifying waste liquid purification. Pei et al. [55] introduced an electrochemical method for synthesizing clean GO sheets via water electrolytic oxidation of graphite, a scalable, safe, and ultrafast green process that is significantly faster than traditional methods. Nasreen et al. [56] proposed an economical and straightforward room-temperature method, replacing sodium nitrate ($NaNO_3$) with phosphoric acid (H_3PO_4) to eliminate toxic gas production. These new methods retain the GO's properties, such as thickness, dispersibility, lateral dimensions, and chemical structure, similar to those produced by conventional methods.

The use of GO in cement composites (GO/CCSs) as a sustainable building material has also gained attention. Muthu et al. [57] found that incorporating GO reduces the penetration of citric and sulfuric acids into concrete, enhancing its lifespan. Zeng et al. [58] showed that GO significantly improves the sulfate resistance of cement-based materials by refining pores and preventing microcracks, with 0.03 wt% GO extending structural life by 2.3 times. Ho et al. [59] used industrial byproducts like fly ash, ground granulated blast furnace slag, lead smelter slag, and GO to create alkali-activated binder mortars (AABs), which exhibited better mechanical and durability properties than AABs without GO. Given the low dosage required, the cost of GO in cement composites remains manageable, presenting a promising solution for sustainable construction by reducing environmental impact.

4.2. Mechanical Properties of Graphene Oxide

Graphene oxide (GO) possesses impressive mechanical properties that make it an ideal material for various applications, including composites and construction. For example, GO features a high Young's modulus, indicating its stiffness, with typical values ranging from 200 to 250 GPa [60]. Additionally, GO showcases a remarkable tensile strength, reaching up to 130 MPa [61]. Its elastic modulus ranges between 200 and 350 GPa, depending on the quality and preparation methods [62]. Moreover, GO's fracture toughness, which reflects its resistance to crack propagation, is significant, ranging from 10 to 20 $MPa \cdot m^{0.5}$ [63]. These attributes underscore GO's potential to enhance the mechanical performance of various materials, making it a promising candidate for sustainable advancements in the fields of construction and materials science.

The integration of GO into cementitious composites has garnered significant attention in scientific research, owing to its potential to augment diverse properties of these materials. Cementitious composites, notably concrete, play a pivotal role in construction due to their robustness, resilience, and adaptability. The incorporation of nanomaterials like GO holds promise for bolstering the mechanical, thermal, and electrical characteristics of these composites.

4.3. Microstructure and Properties of Cement Paste Composites

The study by Yang et al. [64] investigated how GO influences the microstructure and properties of cement paste composites. Their research revealed substantial improvements in microstructure attributed to GO acting as a pore filler. This pore-filling capability resulted in a more compact and uniform composition within the cement paste, significantly reducing porosity and refining the pore structure. The literature reveals that that the addition of GO enhances the microstructure of concrete and a decrease in the amount of porosity that exists inside concrete is brought about by the transformation of hydration crystals into a structure resembling flow. The development of calcium carbonate crystals in concrete may be aided by the incorporation of nucleation sites throughout the material. These crystals have an important part to play in the process of pore filling, which eventually results in the formation of self-healing properties in the concrete [65]. Moreover, scanning electron microscopy (SEM) images of GO-modified samples clearly showed fewer visible pores compared to the control samples. Additionally, pore size distribution analysis indicated a decrease in both the number and size of pores, highlighting the enhanced compactness of the composite material.

In terms of mechanical properties, the study demonstrated significant enhancements in compressive strength with the addition of GO. Even a small addition of 0.05% GO by weight of cement led to a notable 15% increase in compressive strength, from 40 MPa in the control sample to 46 MPa. This improvement was attributed to the refined microstructure and improved bonding between the cement matrix and GO. GO's role as a filler material also contributed to the overall density enhancement of the composite, further bolstering its mechanical properties.

Furthermore, detailed pore structure analysis provided quantitative evidence of the microstructural improvements. The total porosity of the cement paste composite decreased from 22% in the control sample to 17% with 0.05% GO, indicating a 23% reduction in porosity. Moreover, the average pore size decreased from 180 nm to 140 nm, demonstrating a more refined and compact pore structure in the GO-modified samples (summary in Table 7).

Table 7. Key findings of research on microstructure properties of GO.

Property	Control (0% GO)	0.05% GO	Improvement
Microstructure	-	-	More compact and uniform composition
Total Porosity (%)	22	17	23% reduction
Average Pore Size (nm)	180	140	22% reduction
Compressive Strength (MPa)	40	46	15% increase

This underscores GO's pivotal role in enhancing both the microstructure and mechanical properties of cement paste composites. By filling pores and refining the structure, GO significantly improves the strength, durability, and overall performance of these materials. These findings underscore the potential of GO as a valuable additive for developing high-performance and durable cementitious materials suitable for various construction applications.

The effect of 0.05% GO on porosity is likely due to its interactions with calcium hydrosilicates and the resulting microstructural changes rather than its physical presence in the pores. GO's high reactivity, surface interactions, and ability to alter the microstructure at the nanoscale can significantly reduce porosity without needing to fill the voids entirely. In a system with 17–22% porosity, the ability of a small amount of GO (0.05%) to influence pore structure is likely not due to physically filling the pores, given its minimal volume. Instead, the impact is more likely explained by several other factors.

4.3.1. Surface Chemistry and Interactions

GO Adsorption on Pore Walls: With its large surface area and functional groups (like hydroxyl, carboxyl, and epoxide groups), GO can adsorb onto pore surfaces, especially in materials such as calcium hydrosilicates. This chemical adsorption alters surface properties and effectively reduces pore space without directly filling the pores.

4.3.2. Role of Calcium Hydrosilicates

Interaction with Calcium Hydrosilicates: Weakly crystallized calcium silicate hydrates (C-S-H), which have a gel-like structure, tend to retain water. GO's high reactivity can lead to structural changes through interaction with these hydrosilicates.

Nucleation Sites: GO can act as a nucleation site for forming additional C-S-H phases or other hydrated products, leading to finer crystals or denser gel-like structures, which can reduce overall porosity.

4.3.3. Microstructural Changes

Microstructure Refinement: GO can enhance matrix packing, resulting in a more compact and interconnected microstructure. Even in small amounts, it can promote the formation of fine, dense crystals or alter the morphology of existing phases, reducing pore space.

Pore Blockage by GO-C-S-H Interaction: The interaction between GO and C-S-H may create gel phases that block larger pores, thus decreasing effective porosity.

4.3.4. GO in Nanopores

Dispersion in Nanopores: Despite its low concentration, GO could disperse at the nanoscale, occupying the smallest pores or interlayer spaces within the calcium hydrosilicate structure. This would lead to reduced porosity, even without traditional pore filling.

4.4. Rheological Properties

The research proposed by Shang et al. [66] delved into the influence of GO on the rheological properties of cement pastes. Their study provided valuable insights into how the inclusion of GO affects the workability and stability of cement pastes, ultimately shaping their flow behavior. The results revealed a significant enhancement in the workability and stability of cement pastes with the incorporation of GO. Specifically, GO contributed to a reduction in the viscosity of the cement paste, thereby improving its flowability. This reduction in viscosity plays a crucial role in enhancing the ease of handling and application of cement pastes, particularly in construction scenarios where optimal workability is essential. The observed improvements in rheological properties can be attributed to the dispersing effect of GO within the cement paste. By effectively dispersing throughout the paste matrix, GO prevented the agglomeration of cement particles and other additives, facilitating a more uniform mixture. This uniform dispersion promotes homogeneity within the cement paste, facilitating smoother flow and improved consistency.

Furthermore, the presence of GO optimizes the packing density of particles within the cement paste, further enhancing its flow characteristics. This optimization reduces internal friction and resistance to flow, resulting in improved workability and stability.

Table 8 succinctly summarizes the key findings regarding the rheological properties of cement pastes incorporating GO. At a content of 0.05%, the addition of GO resulted in notable improvements in workability and stability, alongside reduced viscosity and increased flowability of the cement pastes.

Table 8. Rheological properties of cement pastes with GO.

Rheological Properties	GO Content
Enhanced workability and stability, reduced viscosity, increased flowability	0.05%

4.5. Enhancement of Mechanical Properties with Graphene Oxide (GO) in Cementitious Materials

Incorporating graphene oxide (GO) into cementitious materials has been shown to significantly enhance their mechanical properties. This enhancement is primarily due to the unique characteristics of GO, such as high surface area, excellent mechanical strength, and functional groups that facilitate strong interactions with the cement matrix. This summary details findings from two key studies: Long et al. [67] and Peng et al. [68].

Long et al. [67] conducted a thorough investigation into the effects of GO on the mechanical properties of cementitious composites, focusing on compressive strength, flexural strength, and modulus of elasticity. By using varying concentrations of GO, they aimed to determine the optimal dosage for maximum mechanical enhancement. Their study revealed that a 0.05 wt% addition of GO resulted in a 24% increase in compressive strength compared to the control sample, while a 0.1 wt% addition led to a 35% improvement. However, higher GO concentrations (above 0.1 wt%) did not yield significant additional improvements and sometimes caused a slight decrease in strength due to agglomeration of GO sheets. Similarly, a 0.05 wt% GO addition increased flexural strength by 22%, with the maximum improvement (28%) at 0.1 wt% GO. The modulus of elasticity also saw an 18% increase with a 0.05 wt% GO addition, and a 25% increase at 0.1 wt% GO.

Peng et al. [68] expanded on this research by exploring the mechanisms behind the mechanical improvements observed. They identified key mechanisms through which GO enhances the mechanical properties of cement composites: enhanced interfacial bonding, crack bridging effect, and nucleation of hydration products. The functional groups on GO sheets, such as carboxyl and hydroxyl, form strong chemical bonds with the cement matrix, leading to improved interfacial bonding. GO sheets also act as bridges across microcracks, preventing crack propagation and thus enhancing the material's toughness. Additionally, GO provides nucleation sites for the formation of hydration products, resulting in a denser and more homogeneous microstructure.

Quantitative findings from Peng et al. [68] showed that a 0.03 wt% addition of GO increased compressive strength by 20%, while a 0.06 wt% addition led to a 30% improvement. Similarly, a 0.03 wt% GO addition increased flexural strength by 19%, with the maximum improvement (27%) at 0.06 wt% GO. The modulus of elasticity increased by 15% with a 0.03 wt% GO addition and by 22% at 0.06 wt% GO (summary in Table 9).

Table 9. Summary of mechanical properties of cementitious composites with GO.

GO Content (wt%)	Compressive Strength (MPa)	Improvement (%)	Flexural Strength (MPa)	Improvement (%)	Modulus of Elasticity (GPa)	Improvement (%)
0.00	38	-	6.5	-	24	-
0.03	45.6	20	7.74	19	27.6	15
0.06	49.4	30	8.26	27	29.3	22
0.09	48	26	8.1	25	28.7	20

4.6. Durability Enhancement with Graphene Oxide (GO) in Cementitious Materials

The incorporation of graphene oxide (GO) into cementitious materials has demonstrated considerable potential for enhancing their durability. Studies by Qureshi and Panesar [69] and Ghazizadeh et al [70] have explored various durability aspects of GO-modified cement composites, noting improvements in resistance to chloride ingress, sulfate attack, alkali–silica reaction (ASR), and carbonation. The enhanced impermeability of the cement matrix, due to the barrier effect of GO sheets, plays a significant role in reducing the diffusion of aggressive ions and enhancing the long-term performance of concrete structures.

Qureshi and Panesar [69] investigated the impact of GO on the resistance of cement composites to chloride ingress and sulfate attack. They tested cement composites with different GO concentrations to determine the optimal dosage for durability enhancement. Their findings indicated that a 0.05 wt% addition of GO reduced chloride ion diffusion by 25% compared to the control sample, while a 0.1 wt% addition reduced chloride ion diffusion by 40%. In terms of sulfate attack, a 0.05 wt% GO addition resulted in a 20% decrease in mass loss, with the maximum improvement (35% reduction in mass loss) observed at 0.1 wt% GO.

Conversely, Ghazizadeh et al. [70] extended their investigation to assess the effects of GO on ASR and carbonation resistance, as well as impermeability. Their study found that the expansion due to ASR was reduced by 18% with a 0.03 wt% GO addition, while a 0.06 wt% addition led to a 30% reduction in ASR-induced expansion. For carbonation resistance, a 0.03 wt% GO addition reduced the carbonation depth by 22%, and a 0.06 wt% GO addition achieved a 35% reduction in carbonation depth. Additionally, the water permeability coefficient decreased by 20% with a 0.03 wt% GO addition and further decreased by 28% with a 0.06 wt% GO addition (summary in Table 10).

Table 10. Comparison of studies on durability enhancement of GO.

Durability Aspect	References	GO Content (wt%)	Improvement/Reduction
Chloride Ingress	[69]	0.05	25% reduction in chloride ion diffusion
		0.10	40% reduction in chloride ion diffusion
Sulfate Attack	[69]	0.05	20% reduction in mass loss due to sulfate attack
		0.10	35% reduction in mass loss due to sulfate attack
Alkali–Silica Reaction (ASR)	[70]	0.03	18% reduction in ASR-induced expansion
		0.06	30% reduction in ASR-induced expansion
Carbonation Resistance	[70]	0.03	22% reduction in carbonation depth
		0.06	35% reduction in carbonation depth
Impermeability	[70]	0.03	20% reduction in water permeability coefficient
		0.06	28% reduction in water permeability coefficient

Both studies highlight the effectiveness of GO in enhancing the durability of cementitious materials, though they show some differences in focus and findings. Qureshi and Panesar [69] concentrated on chloride ingress and sulfate attack, demonstrating significant improvements at relatively higher GO concentrations (up to 0.1 wt%). In contrast, Ghazizadeh et al. [70] provided a broader assessment that included ASR, carbonation resistance, and impermeability, with notable enhancements even at lower GO concentrations (up to 0.06 wt%). Collectively, these findings suggest that GO is a valuable additive for enhancing the durability and long-term performance of cement-based materials in various construction applications.

To ensure uniform dispersion of small amounts of GO in cement composites (0.05–0.1%), techniques such as ultrasonication, mechanical stirring, chemical functionalization, and the use of surfactants are frequently used. Ultrasonication is particularly efficient at breaking up GO clusters, while surfactants help maintain stability and improve workability. Often, a combination of these methods is applied to achieve optimal GO distribution, leading to enhanced mechanical strength and durability of the cement composites [71]. Moreover, even at low concentrations of 0.05–0.1%, graphene oxide (GO) can reduce cement consumption by improving the mechanical properties and microstructure of cement-based materials. This enhancement enables the use of less cement while maintaining or even increasing the strength and durability of concrete composites. GO strengthens the internal bonding within the cement matrix, potentially reducing the required cement by 5–15%, depending on the application and mix design. Its ability to enhance hydration, densify the microstructure, and fill voids plays a key role in achieving similar compressive strength with less cement in GO-enhanced cement formulations [64,72].

4.7. Sustainability Considerations of Graphene Oxide (GO) in Cementitious Materials

The research by Wang et al. [73] and Wang et al. [72] underscores significant sustainability benefits associated with integrating GO into cementitious materials. Their studies emphasize how GO enhances environmental performance and supports sustainable practices in concrete production. Key benefits include the potential to reduce cement consumption by improving the properties of cement paste, which could lead to lower overall cement content in concrete mixes. This reduction is crucial given the substantial carbon footprint of cement production.

Furthermore, GO-modified concrete exhibits enhanced mechanical properties and durability, enabling the construction of thinner and longer-lasting structures. This improvement not only reduces carbon emissions from construction activities over the lifecycle of buildings and infrastructure but also extends the service life of structures, minimizing the need for frequent maintenance and replacement. Such durability enhancements contribute to sustainability by conserving resources and reducing material waste.

In addition to environmental advantages, the incorporation of GO supports greener construction practices by facilitating more efficient use of waste materials in concrete production. This aligns with principles of the circular economy, promoting sustainable resource management and reducing waste generation. By improving concrete performance without increasing material consumption, GO promotes sustainability in construction, mitigating environmental impacts associated with building materials and methods.

Overall, Wang et al.'s research [72,73] underscores GO's potential to address sustainability challenges in cementitious materials by reducing cement use, lowering carbon emissions, extending structure lifespan, and fostering greener construction practices. These findings highlight GO as a promising additive for advancing environmentally friendly and economically viable solutions in construction and infrastructure development.

5. Challenges and Future Directions

There is a scarcity of studies on the durability of cementitious composites enhanced with GO in comparison to other criteria. Several factors influence the durability of cement composites, including alkali–aggregate interaction, corrosion of reinforcement, and freeze–thaw cycles due to chloride or carbonation exposure. The corrosion resistance of graphene-reinforced concrete is directly related to the presence of GO [21]. The presence of linked, sponge-shaped graphene oxide (GO) layers can decrease the depth to which chloride ions can penetrate [50]. Typically, the anti-freezing performance of GO-reinforced masonry is worse than that of ordinary mortar due to the desorption and absorption of nano-level pores. Nevertheless, the durability of GO-reinforced composites can be enhanced through the incorporation of GO nanosheets. These nanosheets facilitate the formation of a well-organized crystal structure, hence minimizing the presence of harmful pores and microstructural cracking [74]. However, further analysis and research are necessary to enhance the durability of goods through the use of GO-based cement composites. The durability and mobility of liquids in concrete are directly correlated. The durability of concrete is determined by its transport qualities, which affect the permeability of gases and liquids into the material [50,75]. The durability of cement composites is enhanced by the improvement in transport properties, such as water permeability, gas permeability, and water sorptivity, when reinforced with GO . The incorporation of graphene oxide (GO) into the cement matrix improves the durability of composites against corrosive elements by forming a robust barrier that restricts the passage of these hostile chemicals. The transport qualities are influenced by the overall porosity, the distribution of pore sizes, the tortuosity, and the connection of the pores [76,77]. It is believed that GO effectively occupies the empty spaces in cement composites, including both small and large pores at different levels [78]. Only a small number of studies have investigated the self-sensing and piezoresistive capabilities of GO-reinforced cement composites (electrical resistivity qualities) [79]. Self-sensing composites have the ability to detect cracks and defects in materials without the need for additional sensors [80]. The self-sensing qualities and damage detection capabilities of GO-reinforced cement composites have been identified by researchers through the analysis of electrical properties [81]. The researchers utilized the piezoresistive properties of cement composites to assess their structural health. They found that the addition of graphene nanoplatelets improves the piezoresistive qualities of the composite [82]. Limited research studies have been conducted to evaluate the volume stability of cement-based composites containing GO. Nevertheless, the inclusion of GO has a significant impact on the pore structure of cementitious composites, resulting in changes to the hydration process and subsequent modifications to the volume [29]. Furthermore, when compared to concrete, the majority of investigations on GO have been conducted using cement mortars. Concrete is a widely utilized construction material that consists of cementitious composites [83,84]. It is advisable to enhance research methods in GO-reinforced cement-based composites, specifically focusing on material properties at the structural level, in order to facilitate its implementation in construction practices. It is imperative to refine research methodologies concerning graphene oxide-reinforced cement-based composites. This refinement should specifically emphasize the investigation of material properties at the structural level, including mechanical strength, durability, thermal stability, and microstructural characteristics. By employing advanced characterization techniques such as scanning electron microscopy (SEM), X-ray diffraction (XRD), and mechanical testing under various loading conditions, researchers can gain deeper insights into the effects of GO on interfacial bonding and overall composite behavior.

Additionally, it is crucial to explore the scaling effects of graphene oxide incorporation, as well as its interaction with other composite constituents, to understand how these factors influence performance in real-world applications. This focused approach will facilitate the development of guidelines for the effective integration of graphene oxide in cement-based materials, ultimately promoting its adoption in construction practices and enhancing the sustainability and resilience of built environments.

This paper provides an overview and identifies areas that academics could study in future research. A summary of the key aims follows:

1. Enhance the quality of GO-reinforced cementitious composites by investigating the microstructure features and hydration process.
2. Research efficient techniques to enhance the rheological characteristics of cement composites supplemented with GO.
3. Enhance the long-lasting nature and its assessment for GO-reinforced concrete or cement composites.
4. Improve the long-term mechanical qualities of cement-based products with the use of GO.
5. Alleviate issues related to the stability of volume in GO-reinforced concrete or cement composites.
6. Identify utilizations of GO in conjunction with concrete and its potential impact on material and structural characteristics.

6. Conclusions

The study sought to comprehend the application of graphene oxide (GO) in cement-based composites and products, and to assess the present status of research to identify future avenues. In order to achieve the intended objective, the study was structured employing a bibliometric review and scientometric analysis to comprehensively assess the available literature. The research gathered data and information from the WOS database by conducting a search from 2005 to the present. A total of 263 studies were identified, all of which were initially published in 2012. Since 2016, there has been a noticeable increase in the level of interest among researchers in studying GO-reinforced cementitious composites, resulting in its popularity as a research topic. The keyword analysis revealed that, apart from "graphene oxide", the terms "cement" and "microstructure" were the most frequently used keywords in the papers. However, the analysis of keywords also revealed that a substantial amount of research was concentrated on examining the mechanical properties of cementitious composites reinforced with graphene oxide (GO). Overall, GO functions as a pore filler in cement paste composites, significantly reducing porosity and refining pore structure. GO improves the workability and stability of cement pastes by reducing viscosity and enhancing flowability. GO disperses within the paste matrix, preventing the agglomeration of cement particles and promoting a uniform mixture and consistency. GO enhances resistance to chloride ingress, sulfate attack, alkali–silica reaction (ASR), and carbonation. On the other hand, key findings include a 25–40% reduction in chloride ion diffusion, 20–35% decrease in mass loss due to sulfate attack, 18–30% reduction in ASR-induced expansion, and 22–35% reduction in carbonation depth. Incorporating GO into cementitious materials can lower cement consumption, reducing the carbon footprint of concrete production. Improved mechanical properties and durability of GO-modified concrete lead to longer-lasting structures, reducing the need for frequent maintenance and replacement. GO promotes efficient use of waste materials in concrete production, aligning with circular economy principles and supporting sustainable construction practices. GO-modified cementitious materials are suitable for various construction applications requiring high performance and durable materials. These materials are especially beneficial in environments needing enhanced resistance to chemical attacks and mechanical stresses. While the use of GO in cementitious composites is currently a popular and rapidly growing area of research, it is still in its early stages. This finding demonstrates the need for more study and efforts in the field of GO-reinforced cementitious composites to achieve better outcomes in terms of mechanical characteristics, rheological properties, durability, and other factors.

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