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Research Trends on Magnetic Graphene for Water Treatment: A Bibliometric Analysis

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Abstract**—**Magnetic graphene has received widespread attention for their capability of water and wastewater treatment, which has been attracted many researchers in this field. A bibliometric analysis based on the Web of Science database was employed to analyze the global scientific outputs of magnetic graphene for water treatment until the present time (2012 to 2017), to improve the understanding of the research trends. The publication year, place of publication, institutes, funding agencies, journals, most cited articles, distribution outputs in thematic categories and applications were analyzed. Three major aspects analyzed including type of pollutant, treatment process and composite composition have further contributed to revealing the research trends. The most relevant research aspects of the main technologies using magnetic graphene for water treatment were summarized in this paper. The results showed that research on magnetic graphene for water treatment goes through a period of decline that might be related to a saturated field and a lack of bibliometric studies. Thus, the result of the present work will lead researchers to establish future directions in further studies using magnetic graphene for water treatment.

Keywords**—**Composite, graphene oxide, nanomaterials, scientometrics.

I. Introduction

GRAPHENE is a monolayer of carbon atoms, with a thickness of an atom, assembled in a hexagonal crystalline structure in a two-dimensional honeycomb lattice with carbon atoms with hybridized sp^2 bonds [1]. Due to its peculiar configuration, it may exhibit extraordinary properties such as high specific area [2], high mechanical resistance and elasticity, and is an excellent electric, current and heat conductor, besides being stable at room temperature [3], and is transparent to ultraviolet radiation, visible light, and infrared [4].

An enormous scientific interest in graphene has aroused in the realization of various interesting and revolutionary applications. The wide range of applications of graphene include: nano-electronic materials, structural compounds, conductive polymers, battery electrodes, supercapacitors, transport barriers, printing inks, bactericidal papers, biomedical technologies, molecular sensors, electrochemical and biochemical sensors, energy storage, drug delivery, water and wastewater treatment, touch screens and solar cells, among others [5].

Graphene is an excellent adsorbent for many pollutants. However, graphene nanosheets tend to aggregate and re-stack into graphite when used during adsorption operations and when used in large quantities due to strong interplanar interactions [6]. This disadvantage can be overcome by covalent or non-covalent functionalization of different molecules and other nanomaterials [7]. The functionalization of the surface of graphene materials with nanoparticles or other functional groups increases their sensitivity, selectivity and limit of detection, which opens up new opportunities to further explore the potential of applying these materials to the adsorption treatment of industrial wastewater [8].

Another problem found in the use of graphene on water treatment processes is its separation after the process treatment [9]. Magnetic separation is an alternative that has emerged as a solution to this problem, being an innovative technology that has gained a lot of attention in recent years that offers a significant advantage over other adsorbents, the ability to separate them from an aqueous solution in the application of a magnetic field [10].

Bibliometrics was first introduced in 1969 [11], as an effective method that uses quantitative analysis to describe the field-specific research trend and has been used in many field-specific global studies recently. It refers to the research methodology used in librarianship and information science to describe distribution patterns of publications according to some categories, such as topics, field, author source, institution or country [12].

The bibliometric method has been widely applied to analyze scientific production and research trends in several fields, such as nitrate removal [13], solid waste [14], desalination [15], global biodiversity [16], climate change [17], water resources [18], aerosol research [19], and others. The Science Citations Index Expanded (SCI-EXPANDED), from database of the scientific information institute Web of Science, is the most important and most frequently used for a broad review of the achievements in all fields [20].

In the present study, a bibliometric analysis of the literature related to graphene inks published in the Web of Science database was conducted. The objective was to determine its quantitative characteristics, as well as to identify the most relevant current and future trends, providing a basis for a

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Open Science Index, Chemical and Materials Engineering Vol:13, No:6, 2019 waset.org/Publication/10010518

better targeting for future research.

II. Materials and Methods

Documents used in this study were obtained from the on-line version of SCI-Expanded (SCIE), Thomson Reuters' Web of Science database. This searchable platform of publications gives access to several databases and other sources of technical information that can be relevant for the diffusion and evaluation of the scientific research. The citation databases provide authoritative, multidisciplinary coverage from more than 12,000 high impact research journals worldwide, including open access journals [21].

Bibliometric analysis was carried out in January 2018 with the online search within Web of Science database by the insertion of "water treatment", "graphene" and "magnetic" as keywords in the topic field of the search-engine in order to compile a complete bibliography with all the articles related to the research on magnetic graphenic materials in drinking water treatment published so far. As graphene is a relatively new material [1], a period of time for the chronological identification of the articles was not delimited; however, documents of the year 2018 were not considered in order to obtain only full years.

The initial search process identified 105 documents. Then, the documents were evaluated through their titles and abstracts using an inclusion criterion of studies on magnetic graphene and graphene derivatives (graphene oxide, reduced graphene oxide, functionalized graphene oxide, etc) materials for water and wastewater treatment. After the evaluation of titles and abstracts, 55 documents were selected. Graphene and graphene derivatives were not differentiated in this study, all of which were simply referred to as "graphene" for simplification purposes.

The research trend information of all the compiled articles were assessed with the following aspects: publication year, place of publication, institutes, funding agencies, journals, most cited articles, distribution of output in thematic categories and applications.

III. Results

The distribution of annual publication output results is shown in Fig. 1.

It is evident that there was a continuous increasing trend until 2015 and after 2015 the number of publications was decreasing, although the publications of the last 3 years, account for almost three-quarters (73%) of the total found publication. This result might indicate that the research on magnetic graphene for water treatment is becoming saturated.

The analysis of the documents identified that the first article published was in 2012, titled "Efficient removal of arsenate by versatile magnetic graphene oxide composites" [22]. Magnetic graphene emerged for the first time as a new composite for water treatment. In this study, the authors studied graphene oxide and magnetite to remove arsenate from water.

The countries ranked by the number of publications are shown in Fig. 2.

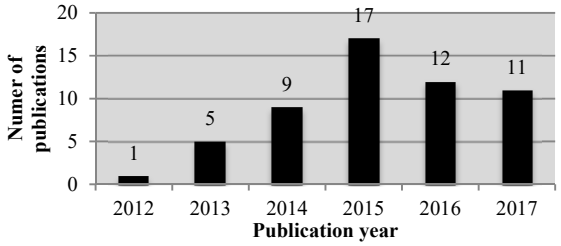

Fig. 1 Annual publication output

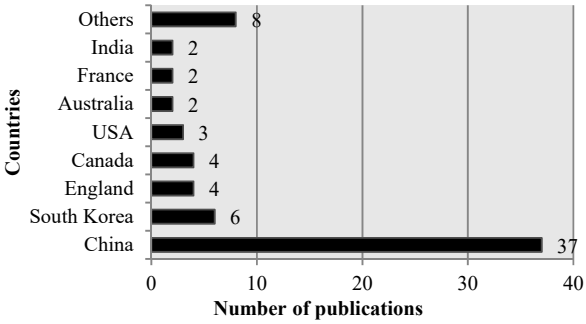

Fig. 2 The most productive countries

The existence of a small number of countries that dominate the publication of scientific documents was expected, as this pattern occurs in most scientific fields [23].

China presented as the most productive country, with 67% publication of total documents, followed by South Korea, England and Canada, with percentages of 11%, 7%, and 7%, respectively.

The most productive institutions and funding agencies of publish documents of magnetic graphene for water treatment were ranked (Tables I and II).

TABLE I
The Most Productive Research Institutes

Rank	Institutes	Country	Documents	Percentage (%)
1	University of Jinan	China	4	7.14
2	Chinese Academy of Sciences	China	3	5.36
3	Hunan University	China	3	5.36
4	Lanzhou University	China	3	5.36
5	Nankai University	China	3	5.36
6	Tongji University	China	3	5.36

The results of institutions and funding agencies showed complete agreement with the most productive country, as all top ranked institutions and funding agencies were Chinese.

The National Natural Science Foundation of China only has supported 65% of the Chinese research regarding magnetic graphene for water treatment.

The distribution output in journals is shown in Table III. The corresponding impact factors, of the most productive journals were also included.

It can be observed that the journal ACS Applied Materials Interfaces is the most productive journal with seven

publications, followed by RSC Advances and Chemical Engineering Journal both with six publications. The other publications were distributed in several journals. The most relevant journal according to the impact factor among the most productive journal was ACS Applied Materials Interfaces with impact factor of 8.097.

The 10 most cited papers of the found bibliography are presented in Table IV.

TABLE II
THE MOST PRODUCTIVE FUNDING AGENCIES

Rank	Institution	Country	Documents	Percentage (%)
1	National Natural Science Foundation of China	China	24	44.64
2	Program for Changjiang Scholars and Innovative Research Team in University	China	4	7.14
3	Program for New Century Excellent Talents in University	China	3	5.36
4	Science and Technology Development Plan of Shandong Province	China	3	5.36
5	Science and Technology Plan Project of Jinan	China	3	5.36

TABLE III
THE MOST PRODUCTIVE JOURNALS AND THE CORRESPONDING SCIENTIFIC IMPACT FACTOR

Rank	Journal	Documents	Percentage (%)	Impact factor
1	ACS Applied Materials Interfaces	7	12.7%	8.097
2	RSC Advances	6	10.9%	2.936
3	Chemical Engineering Journal	6	10.9%	6.735
4	Applied Surface Science	5	9.1%	4.439
5	Ceramics International	3	5.5%	3.057
6	Journal of Colloid and Interface Science	3	5.5%	5.091

TABLE IV
THE MOST CITED GRAPHENE INKS ARTICLES IN THE SCIENTIFIC LITERATURE

Rank	Document	Times cited
1	Title: EDTA functionalized magnetic graphene oxide for removal of Pb(II), Hg(II) and Cu(II) in water treatment: Adsorption mechanism and separation property Author(s): [24] Source: Chemical Engineering Journal	126
2	Graphene Oxide-$MnFe_2O_4$ Magnetic Nanohybrids for Efficient Removal of Lead and Arsenic from Water Author(s): [9] Source: ACS Applied Materials Interfaces	122
3	Facile synthesis of polypyrrole decorated reduced graphene oxide-Fe_3O_4 magnetic composites and its application for the Cr(VI) removal Author(s): [25] Source: Chemical Engineering Journal	117
4	Efficient removal of arsenate by versatile magnetic graphene oxide composites Author(s): [22] Source: RSC Advances	99
5	Synthesis of reduced graphene oxide/magnetite composites and investigation of their adsorption performance of fluoroquinolone antibiotics Author(s): [26] Source: Colloids and Surfaces A-Physicochemical and Engineering	70
6	One-Step Fabrication of Graphene Oxide Enhanced Magnetic Composite Gel for Highly Efficient Dye Adsorption and Catalysis Author(s): [27] Source: ACS Sustainable Chemistry & Interfaces	64
7	Highly Efficient Antibacterial and Pb (II) Removal Effects of Ag-$CoFe_2O4$-GO Nanocomposite Author(s): [28] Source: ACS Applied Materials and Interfaces	64
8	Water-dispersible magnetic nanoparticle-graphene oxide composites for selenium removal Author(s): [29] Source: Carbon	63
9	Facile Aerosol Synthesis and Characterization of Ternary Crumpled Graphene-TiO_2-Magnetite Nanocomposites for Advanced Water Treatment Author(s): [30] Source: ACS Applied Materials Interfaces	50
10	Magnetic nanomaterial derived from graphene oxide/layered double hydroxide hybrid for efficient removal of methyl orange from aqueous solution Author(s): [31] Source: Journal of Colloid and Interface Science	50

According to the list of top 10 most cited articles, 7 were from Chinese research centers. EUA, Singapore, Canada, India and UK presented also studies in the top 10 cited articles. Few studies with international collaboration were observed, only an article with Singapore and China [31] and another with India and UK [9].

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Open Science Index, Chemical and Materials Engineering Vol:13, No:6, 2019 waset.org/Publication/10010518

The results in Table IV are in agreement with the most productive journals (Table III) and the most relevant journal that was ACS Applied Materials Interfaces according with its highest impact factor, resulting in the most cited articles.

Among the articles on list, the 4th most cited article is also the first article published. Overall the most cited articles are those that follow the same line of the first article. In general, they study magnetic graphene without modification or minor modifications for the adsorption removal of simple contaminants such as dyes and metals.

The distribution of the research areas is shown in Fig. 3, where the 5 most popular thematic categories were compiled. It was considered that some publications were included in more than one category.

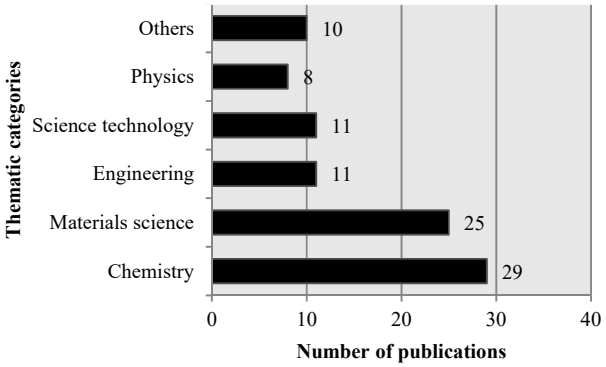

Fig. 3 The most popular thematic categories of Web of Science

The ranking indicates that chemistry (53%), materials science (45%), engineering (20%), science technology (20%), and physics (15%) were the most common research areas, and might indicate that the research trends is in basics sciences, like chemistry, physics, and materials sciences, and less focused in industrial and engineering applications.

The documents were analyzed for the type of pollutants treated in the studies. The results are presented in Fig. 4.

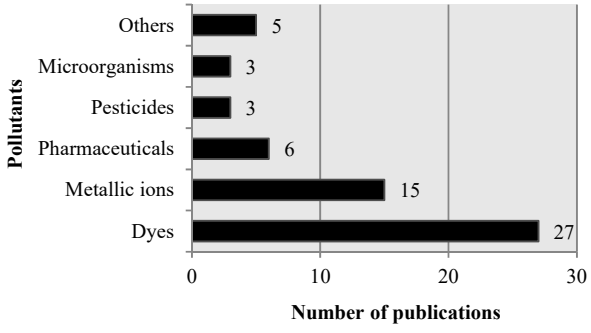

Fig. 4 The most popular pollutants studied

The most popular pollutants studied were dyes (49%) and metallic ions (27%). In others micro pollutants were included, as endocrine disrupting compounds (bisphenol A) and microcystin. It is noteworthy that some studies evaluated more than one pollutant.

A wide range of dyes was studied. Fig. 5 presents the most studied dyes.

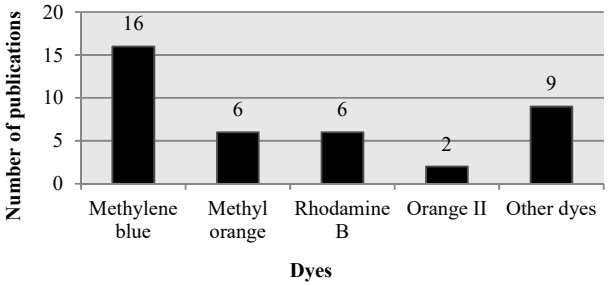

Fig. 5 The most popular dyes studied using magnetic graphene in water treatment

A large number of studies comprise the removal of methylene blue dye, with 29% of the total documents found. This result suggests that researchers may be conducting similar experiments. This result is possibly indicating a lack of review studies and shows the importance of bibliometric studies.

The second most studied pollutant, the metallic ions, was also compiled as the most metallic ions studied and the results are presented in Fig. 6.

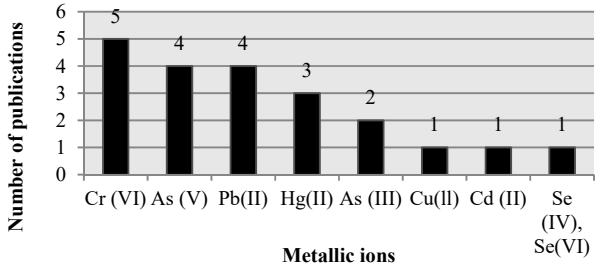

Fig. 6 The most popular metallic ions studied

Among the publications that treat metallic ions, eight different metal ions were studied. The most studied metal ions were chromium, arsenic, and lead.

Heavy metal ions have high solubility in aquatic environments, are bioaccumulative, toxic and nonbiodegradable, can cause severe health problems in animals and humans. Chemical intensive industries discharge large amounts of metal-contaminated wastewater. Thus, it is necessary intensive research on metal-contaminated wastewater treatment.

The results were also analyzed in respect to the composition and/or modification of the composite, the magnetic graphene. The results are shown in Fig. 7.

The majority of studies published have graphene oxide or reduced graphene oxide with ferrite and another metallic compound (such as Mn, Co, Zn, and others) corresponding to 42% of the total published work. Some of these studies can be observed in the most cited articles: nanohybrids of graphene oxide and $MnFe_2O_4$ [9], graphene oxide and Ag-$CoFe_2O_4$ [28], graphene and TiO_2-Magnetite [30].

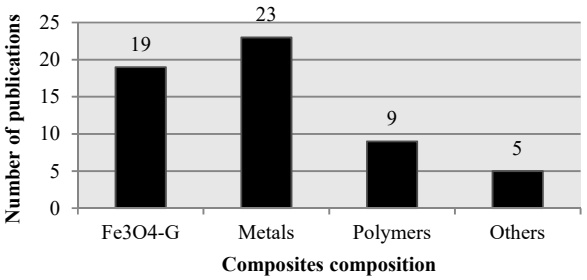

Fig. 7 The most popular composite composition

Magnetic graphene was also synthesized using modification with polymers, and others, in order to obtain different properties for the removal of different pollutants.

The processes used for water treatment with magnetic graphene were also compiled. The results are presented in Fig. 8.

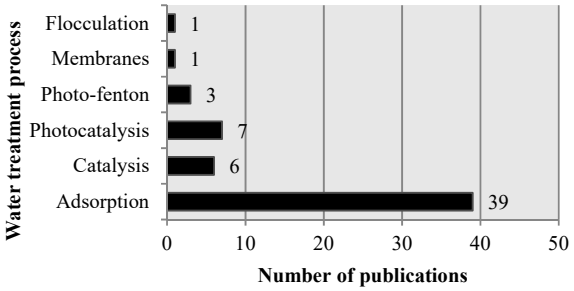

Fig. 8 Water treatment processes used with magnetic graphene

The most popular water treatment process was adsorption with 70% of total publication. This result was expected, as adsorption is the most versatile and widely used method of water treatment because of its low cost, ease of operation, and efficiency in treatment [32].

Advanced oxidation processes, that include photocatalysis and photo-fenton degradation, were also representative, totalizing 18% of the publications. Advanced oxidation processes have received increasing attention in the research and development of wastewater treatment technologies. These processes have been applied successfully for the removal or degradation of toxic pollutants or used as pretreatment to convert recalcitrant pollutants into biodegradable compounds that can then be treated by conventional biological methods [33].

Few studies were found using alternative processes, as membrane and flocculation. Magnetic graphene is a particulate material, and is easily separable from the aqueous solution with the application of a magnetic field that enables the reuse of the nanomaterial in adsorption, catalysis and advanced processes. In this way, the process may become more sustainable and less costly when compared to other processes.

IV. Conclusion

The bibliometric analysis revealed that research on magnetic graphene could be separated into three aspects: pollutants removal, composite composition, and water treatment processes. The composite composition is variable, but the most popular was graphene with ferrite and its combination with another metallic compound. Adsorption presented as the most popular water treatment process, and the removal of pollutants dyes and metallic ions are the most studied application. ACS Applied Materials Interfaces published the greatest number of articles. China is the most productive country; it has the most productive institutes and funding agencies, and also 7 of 10 top cited articles are from Chinese research centers. In this way, China showed to be a dominant contributor to magnetic graphene for water treatment publications. Magnetic graphene for water treatment publications has increased during the past 5 years, but a tendency to decrease was evident. It may be due to the large amount of similar publication that suggests a lack of bibliometric research. Thus, the research trends presented in this study will lead researchers to establish future directions in new studies using magnetic graphene for water treatment.

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