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Bibliometric Analysis of the Research Progress on Graphene Inks from 2008 to 2018

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Abstract**—**A bibliometric analysis in the Web of Science database was used to identify overall scientific results of graphene inks to date (2008 to 2018). The objective of this study was to evaluate the evolutionary tendency of graphene inks research and to identify its aspects, aiming to provide data that can guide future work. The contributions of different researches, languages, thematic categories, periodicals, place of publication, institutes, funding agencies, articles cited and applications were analyzed. The results revealed a growing number of annual publications, of 258 papers found, 107 were included because they met the inclusion criteria. Three main applications were identified: synthesis and characterization, electronics and surfaces. The most relevant research on graphene inks has been summarized in this article, and graphene inks for electronic devices presented the most incident theme according to the research trends during the studied period. It is estimated that this theme will remain in evidence and will contribute to the direction of future research in this area.

Keywords**—**Bibliometric, coating, nanomaterials, scientometrics.

I. Introduction

THE experimental discovery of the isolation of a single sheet of graphene led to the award of the Nobel Prize to the physicists Geim and Novoselov in 2010. After this discovery, the number of experimental and theoretical researches has increased dramatically [1].

Graphene is a monolayer of carbon atoms with a single atom thickness in a hexagonal crystal structure similar to a two-dimensional honeycomb network with carbon atoms with hybridized sp^2 bonds [2]. Since its experimental discovery, graphene has aroused the interest of the scientific community due to its peculiar configuration, by exhibiting excellent chemical and physical properties [3].

Graphene has been used in many interesting and revolutionary applications. The wide range of applications of graphene includes: nanoelectronic materials, structural compounds, conductive polymers, battery electrodes, supercapacitors, bactericidal papers, biomedical technologies, molecular sensors, electrochemical and biochemical sensors, energy storage, drug delivery, water and wastewater treatment, touch screens and solar cells, printing inks, among others [4].

The great majority of the cited applications utilize graphene as a form of coating. Thermal sensors, electrochemical sensors, solar cells, anti-corrosive materials, and touch screens, all require a coating, which stimulates the development of graphene inks. Therefore, graphene represents the possibility to produce and process it in functional inks, with varied rheological and morphological properties, with different thicknesses, printing processes and coatings, searching for a low cost and reliable industrial scale [5].

Bibliometrics is an effective method that uses quantitative analysis to describe the research trend of a specific field [6]. It describes patterns of distribution of publications according to some categories such as topics, fields, sources, authors, institutions or countries research using a methodology used in librarianship and information science, and is widely applied to analyze scientific production and research trends in several fields [7].

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 better targeting for future research.

II. Materials and Methods

The bibliometric analysis of the literature of graphene inks was conducted in Web of Science database. The contribution of different researches published in the main periodicals up to the present period, including languages, place of publication, institutes, funding agencies, periodicals, articles cited, thematic categories and applications were identified.

The results were analyzed and evaluated according to various criteria and used to determine the quantitative characteristics of graphene inks investigations in the globe and the most relevant trends.

The search was performed in January 2019, for which the following descriptors were used as keywords in the search engine topic field: ("paint*" OR "ink*") and ("graphene" and "coat*"). The search process in the first moment allowed the identification of 258 documents. Then, the scientific papers included in the study were selected through the evaluation of titles and abstracts, following the inclusion criteria of publications with studies of graphene inks for different applications. Because graphene is a relatively new material, an initial time period for the chronological identification of the documents has not been delimited. Documents of the year

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2019 were not considered in order to obtain only full years. After the evaluation of titles and abstracts, 107 documents were selected.

III. Results

The distribution of annual publication output identified by Web of Science database of graphene inks for different applications is shown in Fig. 1.

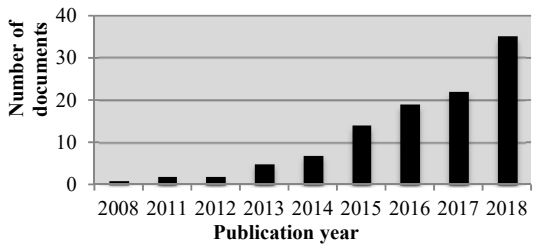

Fig. 1 Annual publication output

Clearly there have been a steadily increasing number of publications each year. The last four years corresponds to 84% of the total publications.

The publications analysis identified that the first paper published was in 2008 titled "Graphene-stabilized copper nanoparticles as an air-stable substitute for silver and gold in low-cost ink-jet printable electronics" [8], the authors applied graphene as an ink, for the first time, in an inkjet printer for electronic devices. In this study, the electrical properties, high conductivity and stability of graphene, were used in a new application for graphene.

The distribution of the document types was analyzed. It was found 93 documents of scientific article type, two review articles, and 14 proceedings papers, totalizing 107 publications selected during the period of study. Please note that review articles are also included in scientific articles.

Most of the publications have been published in English, except one that was published in Chinese. English is undoubtedly the main language of scientific research and has become the best option for all fields [9].

Results with the most productive countries are shown in Fig. 2.

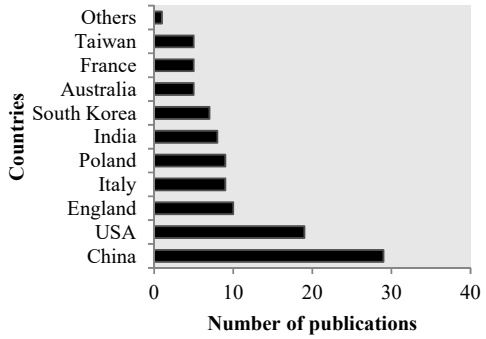

Fig. 2 The most productive countries

China was the most productive country, with 35 articles, and a percentage of 32.7% of all documents, followed by the United States (USA) and England, with 20.6% and 17.8%, respectively.

The most productive institutes and funding agencies were classified by the number of articles published and the results are presented in Tables I and II.

TABLE I
Research Institutes with the Largest Number of Documents on Graphene Inks

Rank	Institutes	Country	Documents	Percentage (%)
1	Chinese Academy of Sciences	China	11	10.28
2	Warsaw University of Technology	Poland	8	7.48
3	Institute of Electronic Materials Technology	Poland	5	4.67
4	University of Cambridge	England	5	4.67
5	Istituto Italiano di Tecnologia IIT	Italy	4	3.74
6	Jeju National University	China	4	3.74

TABLE II
Major Funding Agencies for Research on Graphene Inks

Rank	Institution	Country	Documents	Percentage (%)
1	National Natural Science Foundation of China	China	17	15.89
2	National Science Foundation	USA	5	4.67
3	Engineering and Physical Sciences Research Council	England	3	2.80
4	National Basic Research Program of China	China	3	2.80

The results for the most productive countries, institutions and funding agencies were in agreement, since, the three most productive funding agencies (China, USA and England) agree with the most productive countries. Regarding the most productive institutes, the institutes are from countries that appear on the list of the top five most productive countries, except USA that are not listed in the most productive research institutes. This result may be an indicative that USA do not have the graphene inks research concentrated in a single institution, but in several institutions.

The distribution of the documents in periodicals is presented in Table III. The corresponding impact factors of the most productive periodicals were also included.

TABLE III
Journals with the Largest Number of Articles Publications on Graphene Inks and Scientific Impact Factor

Rank	Journal	Documents	Percentage (%)	Impact factor
1	ACS Applied Materials & Interfaces	5	4,68%	8.097
2	Carbon	5	4,68%	7.088
3	Journal of Materials Chemistry C	5	4,68%	5.976
4	Advanced Materials	4	3,74%	21.950
5	ACS Nano	3	2,80%	13.709

It can be observed that ACS Applied Materials & Interfaces, Carbon and Journal of Materials Chemistry C were the most productive journal, with five publications each, followed by Advanced Materials and ACS Nano, both with

four and three publications, respectively. The other studies were distributed in different journals.

The most relevant periodicals according to the impact factor, among the most productive journals, was Advanced Materials with impact factor of 21,950. It is noticeable that there was a good distribution in different journals, and there was no concentration of documents of the studied topic in a single journal.

It is worth mentioning that the study on graphene inks is a very comprehensive subject, with different applications, so that it is published in journals from different areas.

Table IV lists the 10 most cited articles in the scientific literature.

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

Rank	Document	Citations
1	Title: Graphene as a Long-Term Metal Oxidation Barrier: Worse Than Nothing Authors: [10] Source: ACS Nano	236
2	Title: Graphene-stabilized copper nanoparticles as an air-stable substitute for silver and gold in low-cost ink-jet printable electronics Author(s): [8] Source: Nanotechnology	166
3	Title: 2D-Crystal-Based Functional inks Author(s): [5] Source: Advanced Materials	111
4	Title: Transparent Conductive Electrodes from Graphene/PEDOT:PSS Hybrid Inks for Ultrathin Organic Photodetectors Author(s): [11] Source: Advanced Materials	111
5	Title: Conductive Inks Based on a Lithium Titanate Nanotube Gel for High-Rate Lithium-Ion Batteries with Customized Configuration Author(s): [12] Source: Advanced Materials	81
6	Title: Graphene oxide nanopaint Author(s): [13] Source: Carbon	71
7	Title: Robust Superhydrophobic Graphene-Based Composite Coatings with Self-Cleaning and Corrosion Barrier Properties Author(s): [14] Source: ACS Applied Materials and Interfaces	66
8	Title: Sun-Believable Solar Paint. A Transformative One-Step Approach for Designing Nanocrystalline Solar Cells Author(s): [15] Source: ACS Nano	58
9	Title: Synthesis of Fluorinated Graphene Oxide and its Amphiphobic Properties Author(s): [16] Source: Particle & Particle Systems Characterization	56
10	Title: Graphene-based large area dye-sensitized solar cell modules Author(s): [17] Source: Nanoscale	50

According to the list of the 10 most cited articles (Table IV), it was noted that, although China is the most productive country, with 32.7% of the total work published, any document was found in the list of the most cited articles. This indicates that while China has a significant amount of published work, they do not have studies that have great impact and were effectively used for the development of other researches. The most cited articles were results of research developed mainly in developed countries, among them the United States, Italy, Canada, Germany, Australia and Switzerland.

The most quoted article, "Graphene as a Long-Term Metal Oxidation Barrier: Worse Than Nothing" [10], the authors present graphene as a bad alternative for application in anti-corrosive coatings, which is very controversial, since many authors prove that the use of graphene is excellent as a protective layer of the action of sea water and acidic substances. In this context, the study of Krishnamoorthy and collaborators [13] is also among the most cited works.

Also among the most cited articles, is the first article published using graphene for inks in an inkjet printer with electronic applications [8] and the review article: "2D-Crystal-Based Functional inks" [5]. In this review study, the authors approached different researches that used graphene ink in different solvents with various applications.

The distribution of the research areas is shown in Fig. 3. The documents were compiled by thematic categories of the Web of Science database. It was considered that some publications were included in more than one category.

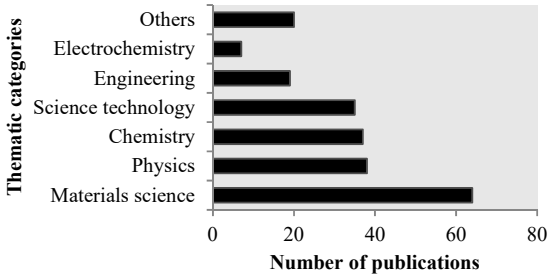

Fig. 3 The most popular thematic categories of Web of Science

The ranking indicates that materials science (59.81%), physics (35.51%), chemistry (34.58%), science technology (32.71%), engineering (17.76%) and electrochemistry (6.54%)

are the most common research areas. This result may indicate that the research on graphene inks is still new and is being developed as a material and not so much for applications in engineering fields and technologies.

The applications discussed in the publications selected were also compiled and are presented in Fig. 4.

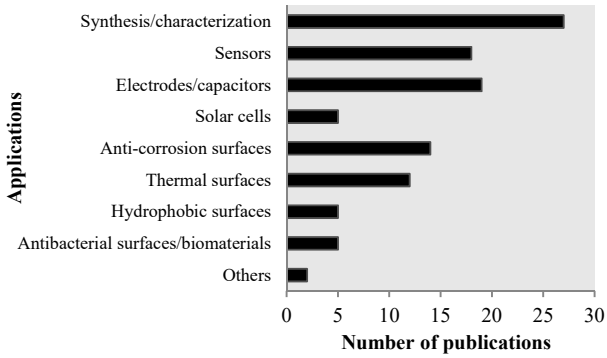

Fig. 4 Applications covered in the publications

Different applications have been identified and organized. A higher concentration of publications of synthesis/characterization of graphene inks was already expected (27.2%), since almost 60% of the publications fall into the thematic category of material science of the Web of Science. It is noteworthy that studies with other applications also include synthesis and characterization, but in this application investigation it was included the main objective of each document.

Applications for graphene inks have been gaining greater prominence, especially in the area of electronics, where graphene inks are generally applied on flexible substrates and then applied with high performance in electronics due to high conductivity [18]. Considering sensors, electrodes/capacitors and solar cells as a general application of electronics, a major application is found with 36.4% of the total documents. This result may represent that the research related to synthesis and characterization may be no longer the main focus of studies, and that researchers are focusing on applications, especially in the electronic area.

The physical properties of graphene, such as high thermal conductivity, high strength, were also exploited using graphene inks. Anti-corrosive surfaces [19], super-hydrophobic [20], and thermal surfaces [21] were developed. Furthermore, graphene has also used as an antibacterial material [22] and been studied for use in biomaterials [23]. Therefore, a broader class of applications may include all documents of surfaces treatments. As result, 33.6% of the documents are studies concerning to surfaces and also surpass the characterization documents (27.2%). This result again indicates that graphene inks research is more consolidated, and studies of synthesis and characterization are no longer the main focus of research, opening new possibilities for studies for different applications.

IV. Conclusion

This bibliometric study provided an overview of current research and identified some significant factors. The analysis of the applications revealed that the research on magnetic graphene could be considered under three aspects: synthesis and characterization, electronics and surface treatment. The researches of synthesis and characterization are still numerous, however the application in electronics and surface treatment have been increasing and has become the majority. Publications related to graphene inks have increased significantly in the last 10 years, since the first publication was in 2008, and are expected to grow in the coming years. A good distribution in different periodicals was found with no concentration of documents in few journals. China was the largest contributor of the research of graphene inks, but the most relevant studies were the research in developed countries such as USA, Italy and England.

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References

[1] O. C. Compton, S. T. Nguyen, "Graphene Oxide, Highly Reduced Graphene Oxide, and Graphene: Versatile Building Blocks for Carbon-Based Materials", *Small*, vol. 6, pp. 711-723, 2010.

[2] A. K. Geim, K. S. Novoselov, "The rise of graphene", *Nat Mater*, vol. 6, pp. 183-191, 2007.

[3] A. Martín, A. Escarpa, "Graphene: The cutting–edge interaction between chemistry and electrochemistry", *TrAC Trends in Analytical Chemistry*, vol. 56, pp. 13-26, 2014.

[4] N. A. A. Ghany, S. A. Elsherif, H. T. Handal, "Revolution of Graphene for different applications: State-of-the-art", *Surfaces and Interfaces*, vol. 9, pp. 93-106, 2017.

[5] F. Bonaccorso, A. Bartolotta, J. N. Coleman, C. Backes, "2D-Crystal-Based Functional Inks", *Advanced Materials*, vol. 28, pp. 6136-6166, 2016.

[6] A. Pritchard, "Statistical bibliography or bibliometrics", *Journal of Documentation*, vol. 25, pp. 1, 1969.

[7] H.-Z. Fu, M.-H. Wang, Y.-S. Ho, "Mapping of drinking water research: A bibliometric analysis of research output during 1992–2011", *Science of The Total Environment*, vol. 443, pp. 757-765, 2013.

[8] N. A. Luechinger, E. K. Athanassiou, W. J. Stark, "Graphene-stabilized copper nanoparticles as an air-stable substitute for silver and gold in low-cost ink-jet printable electronics", *Nanotechnology*, vol. 19, pp. 445201, 2008.

[9] R. Abejón, A. Garea, "A bibliometric analysis of research on arsenic in drinking water during the 1992–2012 period: An outlook to treatment alternatives for arsenic removal", *Journal of Water Process Engineering*, vol. 6, pp. 105-119, 2015.

[10] M. Schriver, W. Regan, W. J. Gannett, A. M. Zaniewski, M. F. Crommie, A. Zettl, "Graphene as a Long-Term Metal Oxidation Barrier: Worse Than Nothing", *ACS Nano*, vol. 7, pp. 5763-5768, 2013.

[11] Z. Liu, K. Parvez, R. Li, R. Dong, X. Feng, K. Müllen, "Transparent Conductive Electrodes from Graphene/PEDOT:PSS Hybrid Inks for Ultrathin Organic Photodetectors", *Advanced Materials*, vol. 27, pp. 669-675, 2015.

[12] Y. Tang, Y. Zhang, X. Rui, D. Qi, Y. Luo, W. R. Leow, S. Chen, J. Guo, J. Wei, W. Li, J. Deng, Y. Lai, B. Ma, X. Chen, "Conductive Inks Based on a Lithium Titanate Nanotube Gel for High-Rate Lithium-Ion Batteries with Customized Configuration", *Advanced Materials*, vol. 28, pp. 1567-1576, 2016.

[13] K. Krishnamoorthy, K. Jeyasubramanian, M. Premanathan, G. Subbiah, H. S. Shin, S. J. Kim, "Graphene oxide nanopaint", *Carbon*, vol. 72, pp.

328-337, 2014.
[14] M. J. Nine, M. A. Cole, L. Johnson, D. N. H. Tran, D. Losic, "Robust Superhydrophobic Graphene-Based Composite Coatings with Self-Cleaning and Corrosion Barrier Properties", *ACS Applied Materials & Interfaces*, vol. 7, pp. 28482-28493, 2015.
[15] M. P. Genovese, I. V. Lightcap, P. V. Kamat, "Sun-Believable Solar Paint. A Transformative One-Step Approach for Designing Nanocrystalline Solar Cells", *ACS Nano*, vol. 6, pp. 865-872, 2012.
[16] A. Mathkar, T. N. Narayanan, L. B. Alemany, P. Cox, P. Nguyen, G. Gao, P. Chang, R. Romero-Aburto, S. A. Mani, P. M. Ajayan, "Synthesis of Fluorinated Graphene Oxide and its Amphiphobic Properties", *Particle & Particle Systems Characterization*, vol. 30, pp. 266-272, 2013.
[17] S. Casaluci, M. Gemmi, V. Pellegrini, A. Di Carlo, F. Bonaccorso, "Graphene-based large area dye-sensitized solar cell modules", *Nanoscale*, vol. 8, pp. 5368-5378, 2016.
[18] K. Karimi, E. Jabari, E. Toyserkani, P. Lee-Sullivan, "Highly conductive graphene paper for flexible electronics applications", *Journal of Materials Science: Materials in Electronics*, vol. 29, pp. 2537-2549, 2018.
[19] S. De, J. L. Lutkenhaus, "Corrosion behaviour of eco-friendly airbrushed reduced graphene oxide-poly(vinyl alcohol) coatings", *Green Chemistry*, vol. 20, pp. 506-514, 2018, W. Sun, L. Wang, Z. Yang, T. Zhu, T. Wu, C. Dong, G. Liu, "A facile method for the modification of graphene nanosheets as promising anticorrosion pigments", *Materials Letters*, vol. 228, pp. 152-156, 2018.
[20] L.-B. Lv, T.-L. Cui, B. Zhang, H.-H. Wang, X.-H. Li, J.-S. Chen, "Wrinkled Graphene Monoliths as Superabsorbing Building Blocks for Superhydrophobic and Superhydrophilic Surfaces", *Angewandte Chemie International Edition*, vol. 54, pp. 15165-15169, 2015.
[21] N. Karim, M. Zhang, S. Afroj, V. Koncherry, P. Potluri, K. S. Novoselov, "Graphene-based surface heater for de-icing applications", *RSC Advances*, vol. 8, pp. 16815-16823, 2018, S. Giaveri, P. Gronchi, A. Barzoni, "IPN Polysiloxane-Epoxy Resin for High Temperature Coatings: Structure Effects on Layer Performance after 450 °C Treatment", *Coatings*, vol. 7, pp. 213, 2017.
[22] Dybowska-Sarapuk, A. Kotelab, J. Krzemińskia, D. Janczaka, M. Wróblewska, H. Marcheld, P. Łęgorzb, M. Jakubowskaa, "Antibacterial activity of graphene layers," *Photonics Applications in Astronomy, Communications, Industry, and High-Energy Physics Experiments*, Wilga, 2016.
[23] J.-W. Yang, M.-L. Tseng, Y.-M. Fu, C.-H. Kang, Y.-T. Cheng, P.-H. Kuo, C.-K. Tzeng, S.-H. Chiou, C.-Y. Wu, G.-Y. Chen, "Printable Graphene Oxide Micropatterns for a Bio-Subretinal Chip", *Advanced Healthcare Materials*, vol. 7, pp. 1800365, 2018.