

Green Synthesis of Nanomaterials: most cited papers and research trends
Síntese verde de nanomateriais: trabalhos mais citados e tendências de pesquisa
Síntesis verde de nanomateriales: trabajos más citados y tendencias de investigación

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Abstract

A bibliometric analysis using the Web of Science database was performed on the green synthesis of nanomaterials published between 2003 and 2017, obtaining a sample with a total of 159 publications. The number of citations of each paper, thematic areas, sources of publication and countries of origin were quantified. The ten most cited papers were described in further detail, listing the precursor materials, the kind of material produced among other characteristics. There is a variety of precursors listed in the literature, including fungi, bacteria, plants, extracts from plants and fruits. Although United States was a pioneer and is influent in this line of research, India is taking the lead due to government incentives and the presence of natural resources having a total of 44.65% of the analyzed works. Silver oxides is

the material most produced by this method, according to the most cited works and are commonly applied in antibacterial activity. The main areas of discussion at WoS for this topic are the Chemistry and Science Technology categories with 55 papers each. The main sources of publication are the *Colloids and Surfaces B - biointerfaces* and *RSC Advances* with 3,774% in each journal in relation to the study sample. Potentialities of the new nanomaterials production method that could be exploited by other researchers from the use of new precursors, including solid agroindustrial wastes, with the opportunity to add value to the biomass and consequently to reduce environmental impact generated by by-products.

Keywords: Sustainability; Nanomaterials; Green Synthesis; Bibliometrics; Nanoscience and Nanotechnology.

Resumo

Uma análise bibliométrica utilizando o banco de dados *Web of Science* foi realizada sobre a síntese verde de nanomateriais publicados entre 2003 e 2017, obtendo-se uma amostra com um total de 159 publicações. O número de citações de cada artigo, áreas temáticas, fontes de publicação e países de origem foram quantificados. Os dez artigos mais citados foram descritos em maiores detalhes, listando os materiais precursores, o tipo de material produzido dentre outras características. Existe uma variedade de precursores listados na literatura, incluindo fungos, bactérias, plantas, extratos de plantas e frutas. Embora os Estados Unidos tenham sido pioneiros e influentes nessa linha de pesquisa, a Índia está assumindo a liderança devido aos incentivos do governo e à presença de seus recursos naturais, possuindo um total de 44,65% dos trabalhos analisados. Os óxidos de prata configuram-se como o material mais produzido por esse método, de acordo com os trabalhos mais citados e são comumente aplicados em atividades antibacterianas. As principais áreas de discussão na WoS para esse tema são as categorias de Química e Tecnologia & Ciência com 55 trabalhos cada. As principais fontes de publicação são as revistas *Colloids and Surfaces B – biointerfaces* e *RSC Advances* com 3,774% em cada periódico em relação a amostra em estudo. A potencialidade desse método para produção de nanomateriais poderia ser explorada por outros pesquisadores a partir da utilização de novos precursores, incluindo resíduos agroindustriais sólidos, com a oportunidade de agregar valor à biomassa e, conseqüentemente, reduzir o impacto ambiental gerado pelos subprodutos.

Palavras-chave: Sustentabilidade; Nanomateriais; Síntese Verde; Bibliometria; Nanociência e Nanotecnologia.

Resumen

Se realizó un análisis bibliométrico utilizando la base de datos de Web of Science sobre la síntesis verde de nanomateriales publicada entre 2003 y 2017, obteniendo una muestra con un total de 159 publicaciones. Se cuantificó el número de citas de cada artículo, áreas temáticas, fuentes de publicación y países de origen. Los diez artículos más citados se describieron con más detalle, enumerando los materiales precursores, el tipo de material producido entre otras características. Hay una variedad de precursores enumerados en la literatura, incluidos hongos, bacterias, plantas, extractos de plantas y frutas. Aunque Estados Unidos fue un pionero y es influyente en esta línea de investigación, India está tomando la delantera debido a los incentivos gubernamentales y la presencia de recursos naturales teniendo un total de 44,65% de las obras analizadas. Los óxidos de plata son el material más producido por este método, de acuerdo con los trabajos más citados y se aplican comúnmente en actividades antibacterianas. Las principales áreas de discusión en WoS para este tema son las categorías de Química y Tecnología Científica con 55 artículos cada una. Las principales fuentes de publicación son las revistas *Coloides y Superficies B: biointerfaces* y *RSC Advances* con 3.774% en cada revista en relación con la muestra del estudio. Potencialidades del nuevo método de producción de nanomateriales que podrían ser explotados por otros investigadores a partir del uso de nuevos precursores, incluidos los desechos agroindustriales sólidos, con la oportunidad de agregar valor a la biomasa y, en consecuencia, reducir el impacto ambiental generado por los subproductos.

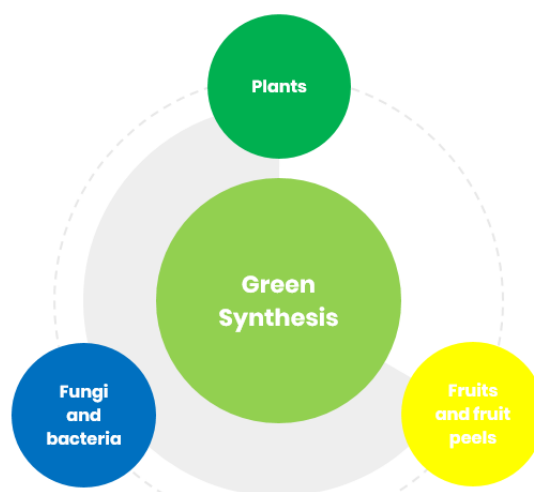
Palabras clave: Sostenibilidad; Nanomateriales; Síntesis verde; Bibliometría; Nanociencia y Nanotecnología.

1. Introduction

Nano-scale materials have been receiving considerable attention due to their unique properties and the large number of applications in various areas such as optics, biomedicine, mechanics, magnetism, and catalysis (Du, Jiang, Liu, & Wang, 2007). Several methodologies are reported for the preparation of these materials, such as sol-Gel (Niederberger, 2007), Grinding (Zhang, Yao, Zhang, Ma, & Lin, 2015), Combustion (Chanadee, 2017), hydrothermal (Debnath, Das, Das, & Sutradhar, 2017), coprecipitation (Benrabaa et al., 2012). In addition to alternative methods that consist of adaptations of the traditional (Majedi, Abbasi, & Davar, 2016).

The chemical processes of green synthesis of nanomaterials (GSN) can be defined as a synthesis using organic precursors extracted directly from nature with no or as little treatment as possible in search of simplicity, reduced production costs, and sustainability, replacing several traditional precursors such as organic solvents with fungi (Mukherjee et al., 2002), bacteria (Wright, Farooqui, White, & Greene, 2016), plants (Sadeghi & Gholamhoseinpoor, 2015), fruits (Proveti et al., 2015) and fruit peels (Bankar, Joshi, Ravi Kumar, & Zinjarde, 2010; R. Kumar, Roopan, Prabhakarn, Khanna, & Chakroborty, 2012) as illustrated in Figure 1.

Figure 1. Precursors for Green Synthesis.



Source: Prepared by the authors.

Within the concept of sustainability, the reuse of residues, mainly from fruit peels for production of nanoparticles is according to the guidelines established by legislation in several countries. In Brazil, Federal Law n. 12305/2010 (Lei, 2010), which establishes the National Policy on Solid Waste - PNRS, determines the application of reverse logistics, characterized as an instrument for waste management, enabling its collection, restitution, recycling or adequate disposal (Rogers & Tibben-Lembke, 2001).

To explain the influence of the “green precursors” during the synthesis stages of the nanostructured compounds, several mechanisms are proposed, such as were reported by the authors Makarov and co-workers (Makarov et al., 2014), who consider plant extracts and their functional groups in the role of ligands to the metallic ions responsible for the chemical reduction of the metal atoms and stabilizing the particles. In other studies, the chemical groups carbonyl, thiol, and Amina are associated with the potential for the reduction of metallic ions and the favoring of the growth of the crystals (He et al., 2007).

Due to the improvement in the resources of freely available databases, the increasing complexity of the research fields and the need for accountability in governmental spending, the use of bibliometric indicators to measure productivity in science and technology is needed. Where bibliometrics is defined as the use of statistical counting of articles, patent documents, citations, words and terms to find trends and patterns in the development of a research field (Milanez, Schiavi, Amaral, Faria, & Gregolin, 2013).

In this paper a bibliometric analysis was used to understand the evolution of the GSN field, including the uses of this technology, which countries are central in its development, if there is any correlation between government policy and a preference for green synthesis, what journals, and papers are the most cited in the area.

2. Methodology

2.1 Data source

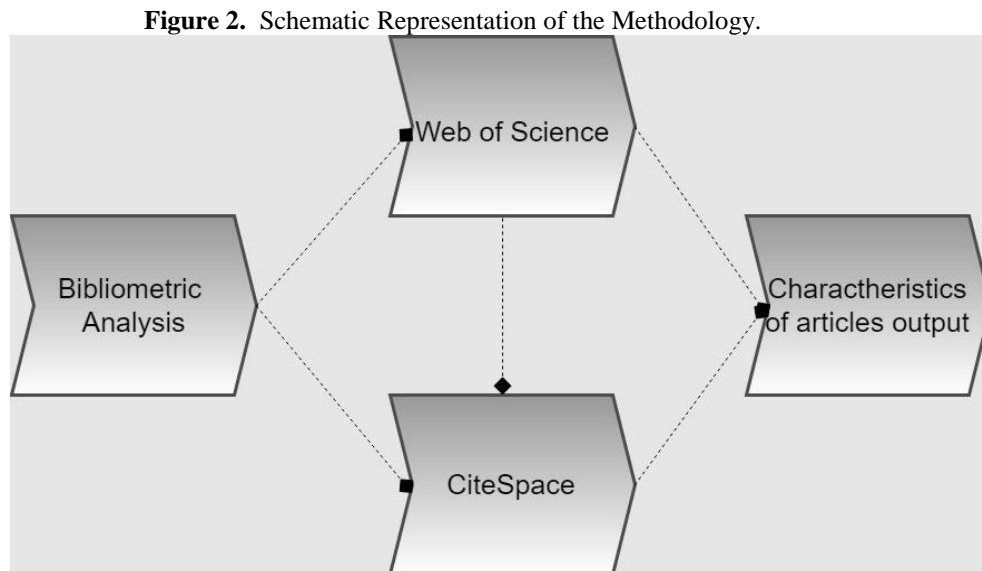
A bibliometric survey was carried out through the Web of Science (WoS) database. The WoS database was chosen, because of its academic recognition on the world stage, the leading publications of various areas are listed on it. WoS has more than 12000 journals indexed. The Keywords used were “green synthesis” AND “biosynthesis” AND “nanomaterials,” the data was checked up to 2017. The reported journal impact factor was taken from the JCR Science Edition 2017.

The empirical parameter Betweenness Centrality (BC) was obtained by CiteSpace (Chen, 2017). High values of BC indicate a dominant influence on the transfer of items through networks, if item transfer follows the shortest paths. Robust BC nodes tend to be part of joints bridging the tightly connected sub-graphs that represent discovered knowledge communities (Zhu & Hua, 2017).

2.2 Data analysis

With the mapping done, a set of data was obtained and treated according to the following topics: (I) evolution of the number of citations and publications; (II) search areas and categories of WoS that are linked to the works; (III) kinds of documents; (IV) major published periodicals and the analysis of the impact of each journal through the Journal Citation Reports (JCR) basis of Thomas Reuters; (V) countries of origin; (VI) the relationship of publications between the countries of origin, thematic areas and the keywords were treated

in series using the software CiteSpace a freely available Java application ([http:// Cluster. CIs. Drexel. edu/* Cchen/Citespace](http://Cluster.CIs.Drexel.edu/~Cchen/Citespace)) developed by Chaomei Chen at Drexel University (USA) (Chen, 2017). The methodology of the research is sketched in Figure 2.



Source: Prepared by the authors.

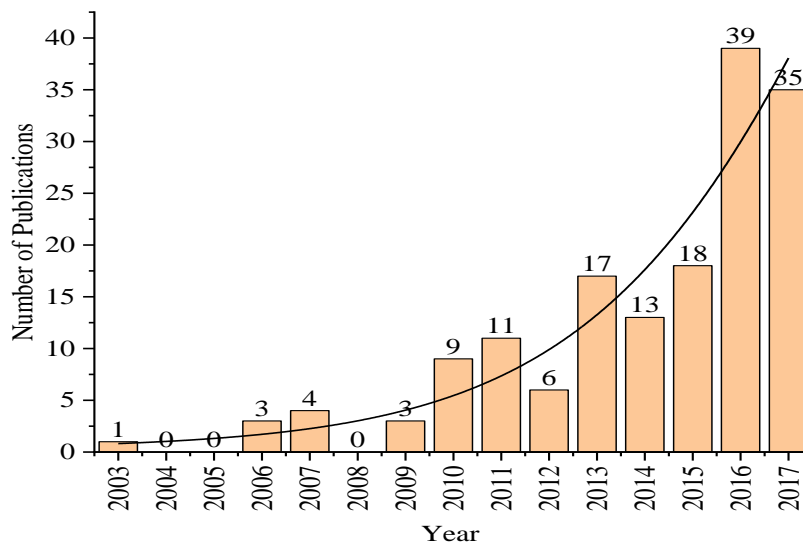
The top ten articles classified by the number of citations were studied in further detail. It is understood that these works are the source of reference for the researchers of the area and will allow the greater understanding of the subject under study.

3. Results and discussion

3.1 Number of publications and citations.

There were 159 publications in the WoS database that match the search criteria. The first publication was in 2003, but there was only a small number up to 2009. In 2010, the number of papers triplicates, and a growth behavior were seen until 2017. The greatest impulse of published work was verified between 2015-2016, reaching a peak of production in 2016, with 39, as shown in Figure 3.

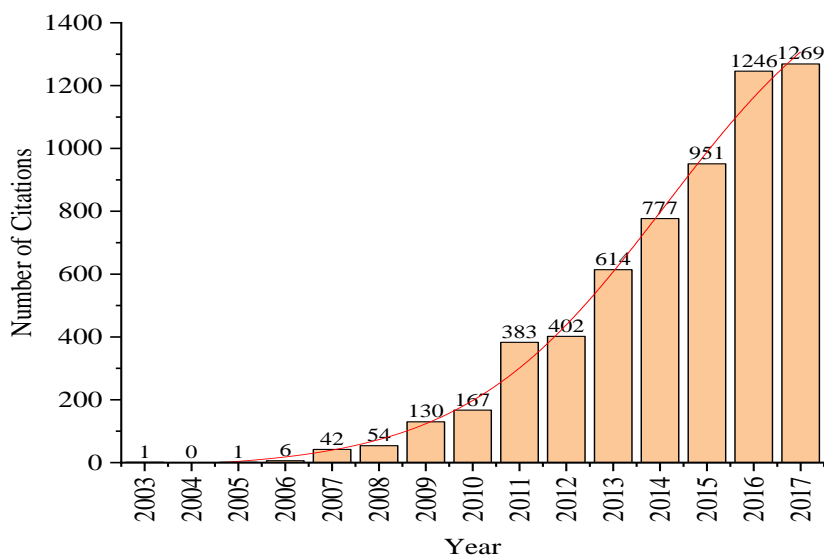
Figure 3. Evolution of the number of publications. The line is a guide to the eyes that show a tendency for growth.



Source: Prepared by the authors.

The number of citations followed that of publications (Figure 4), since 2010 there was significant growth, with a percent increase of 660 % from 2010 to 2017.

Figure 4. List of citations per year with search terms.



Source: Prepared by the authors.

3.2 WoS categories

The areas with greater relevance to the production of nanomaterials by the methodology of green synthesis or biosynthesis are chemistry, science and technology, and material sciences, as illustrated in Table 1. These results illustrate the striking multidisciplinary characteristic of the theme. The Chemistry category (55) and Science Technology (55) have the highest number of papers on the green synthesis methodology, but Chemistry has a higher parameter BC, and this value can be related to its presence into a more substantial number of journals in WoS. For the entire study sample, the Biotechnology Applied Microbiology is the category with the highest BC parameter (0.43) due to the significant number of occurrences of works on gold and silver oxides for applications in molecular biochemistry. High BC values are also associated with the categories of Life Sciences Biomedicine Other Topics, Material Science, Pharmacology Pharmacy and Research Experimental Medicine.

Table 1. Top 20 WoS.

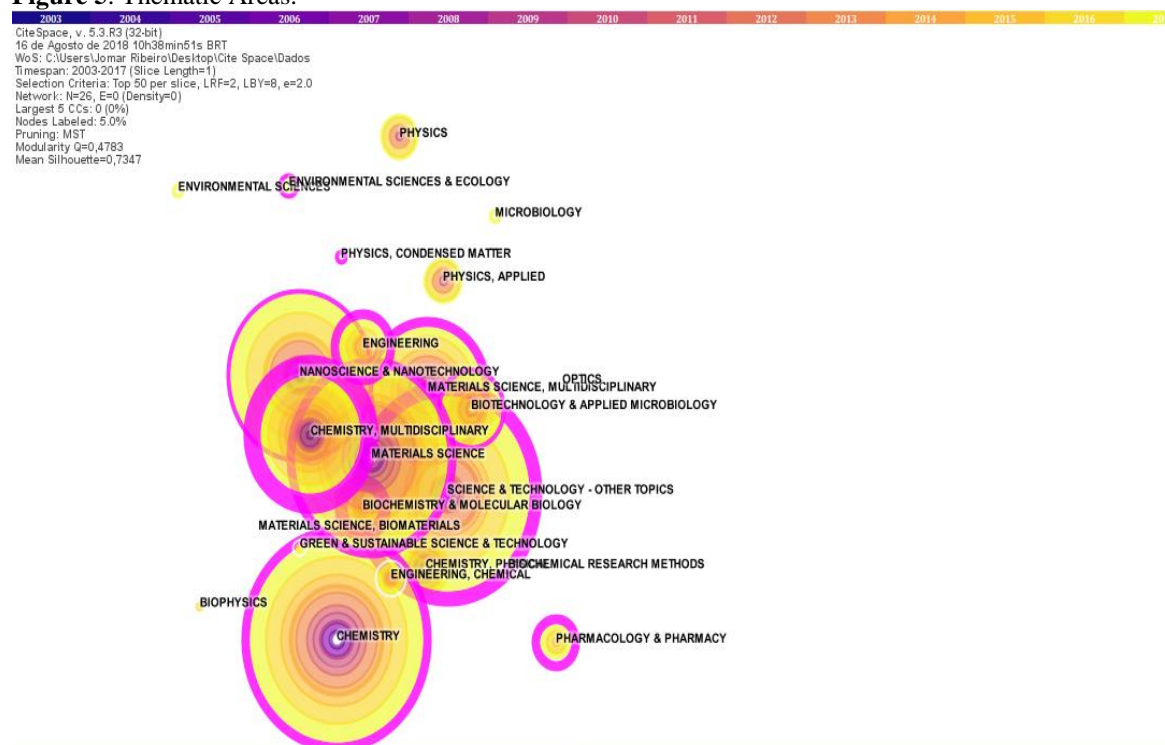
Number of Publications	BC	Start Year	Categories
55	0.4	2003	Chemistry
55	0.08	2006	Science Technology
47	0.21	2007	Materials Science
46	0.1	2007	Biotechnology Applied Microbiology
37	0.11	2007	Engineering
34	0.3	2006	Physics
24	0.43	2006	Biochemistry Molecular Biology
20	0.25	2007	Pharmacology Pharmacy
18	0.18	2010	Environmental Sciences Ecology
17	0.05	2007	Biophysics
17	0.01	2007	Microbiology
13	0.21	2011	Research Experimental Medicine
12	0.08	2012	Optics
11	0.06	2013	Spectroscopy
9	0.18	2010	Electrochemistry
9	0	2010	Immunology
8	0.04	2006	Energy Fuels
8	0.01	2009	Entomology
7	0	2011	Food Science Technology
7	0.3	2009	Life Sciences Biomedicine Other Topics

Source: Prepared by the authors.

Overlay maps using CiteSpace shows to the evolutionary relationship of the thematic areas on Green Synthesis Nanoparticles (Figure 5). In the period evaluated 2013-2017 were

found 26 nodes, 5% nodes labeled. The results for modularity Q and the mean silhouette scores were 0.4783 and 0.7347, respectively. These two metrics were used to evaluate the overall structural properties of the network. Q value was minor than 50%, meaning that the network is low when divided into loosely coupled clusters. Since the mean silhouette score was 0.7347, suggesting that the homogeneity of these clusters on average are high. It is possible to note that the silhouette plot displays a measure of how close each point in one cluster is to points in the neighboring clusters. Another important fact is that two or more specific areas showed interfaces indicating possible networking.

Figure 5. Thematic Areas.



Source: Prepared by the authors.

3.3 Sources of publication

The Top 20 journals with more publications in the area under study are listed in Table 2. To classify the journals the Journal Impact Factor (JCR) representing citation activity in 2017 was taken from the Journal Citation Reports (Thomas Reuters). There are only two journals that published on GSN and are not listed in this classification: Advanced Materials Research and International Journal of Nano Dimension.

From Table 2 it is possible to see that the two journals with higher JCR, “Colloids and

Surfaces B-biointerfaces” e “RSC Advances,” are also the two with a higher number of publications in the area a strong indication that citations follow publication. There is no predominance of a given journal tough.

Comparing the list in Table 2 with the correlation between areas seen in Figure 5 it is possible to see that at least the seven journals with more publications in the area have words related to the central hub in their titles. They fit in the categories of chemistry, biochemistry and material sciences that appear strongly interrelated in the figure.

Table 2. TOP 20 sources of publication.

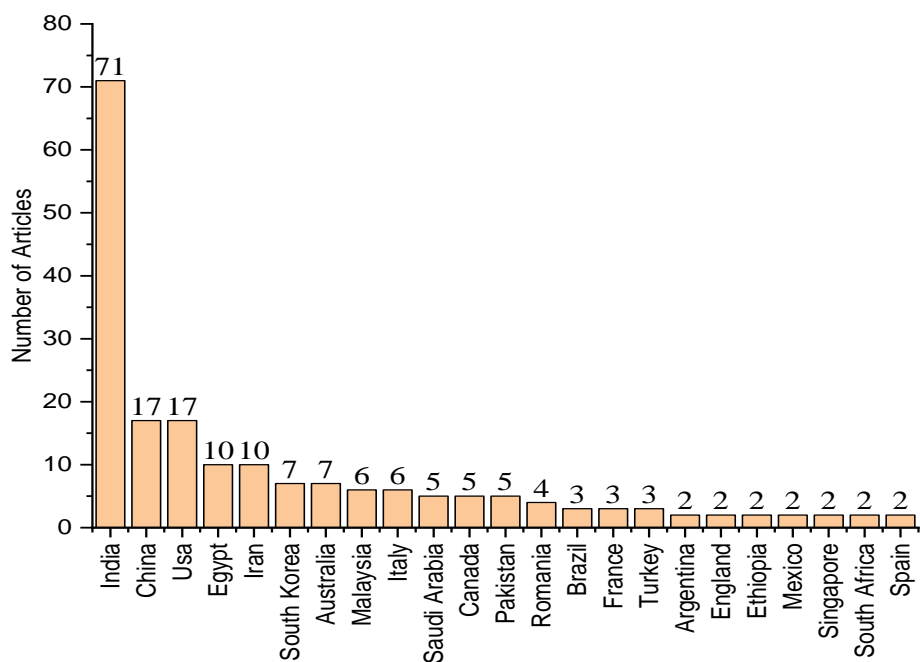
Main sources of publication	Registration Count	Articles % *	JCR
Colloids and Surfaces B - biointerfaces	6	3.774	3.997
RSC Advances	6	3.774	2.936
Journal of Cluster Science	5	3.145	1.715
Digest Journal of nanomaterials and biostructures	4	2.516	0.673
IET Nanobiotechnology	4	2.516	2.059
Acs Sustainable Chemistry Engineering	3	1.887	6.140
Advances in Colloid and Interface Science	3	1.887	7.346
Applied Microbiology and Biotechnology	3	1.887	3.340
Applied Nanoscience	3	1.887	2.951
Biomedicine Pharmacotherapy	3	1.887	3.457
Environmental Science and Pollution research	3	1.887	2.800
Green Chemistry	3	1.887	8.586
International Journal of Nano Dimension	3	1.887	-
International Journal of Nanomedicine	3	1.887	4.370
Langmuir	3	1.887	3.789
Nanoscale research letters	3	1.887	3.125
Advanced materials research	2	1.258	-
Advanced Powder Technology	2	1.258	2.943
Journal of Applied Biomedicine	2	1.258	1.783
Journal of colloid and interface science	2	1.258	5.091

* Percentage of a total number of articles published.

3.4 Countries

The division of publication by country of origin is represented in Table 3 and Figure 6. The country that is most prominent in the production of nanomaterials by this new methodology of synthesis is India that was the origin of 44.65% of the articles of the sample studied, corresponding to a total of 71 works. Followed by China and the USA, with a total of 17 publications each.

Figure 6. Countries with two or more publications.



Source: Prepared by the authors.

India presents the node with the highest centrality parameter (BC) this characteristic means influence on the researchers of the sample because it connects two or more large groups of nodes. The USA, despite the smallest number of published works, has a high BC value that may be associated with the early start of the researches in the area (2006).

Table 3. Top 20 keywords.

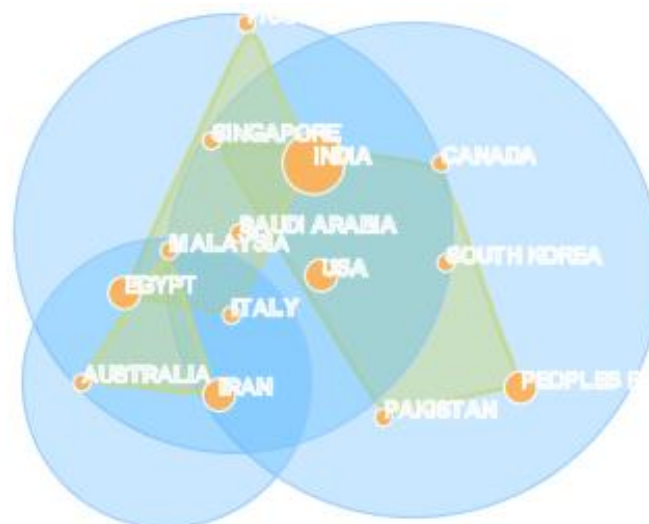
Freq	BC	Start Year	Countries
71	0.39	2003	India
17	0.06	2007	China
17	0.33	2006	USA
10	0.06	2011	Egypt
10	0.02	2011	Iran
7	0.07	2013	South Korea
7	0.04	2007	Australia
6	0.01	2013	Malaysia
6	0.00	2014	Italy
5	0.14	2012	Canada
5	0.02	2015	Pakistan

5	0.00	2014	Saudi Arabia
4	0.00	2010	Romania
3	0.00	2017	Turkey
3	0.21	2016	France
3	0.00	2013	Brazil
2	0.00	2016	South Africa
2	0.00	2016	Argentina
2	0.00	2011	England
2	0.00	2015	Spain

Source: Prepared by the authors.

India, at least up to 2015 was not a leading country in nanoscience and nanotechnology as a whole. According to Zibareva (Zibareva, 2015), China, United States, France, Germany, South Korea, and Japan are the leading countries in the broader nano research field. The prominence of India in the specific area of green synthesis shows a local preference, probably due to natural resources. The growth of research in India in the field of nanotechnology is justified, since, from the year 2000, significant investments were fomented with the Program on Nanomaterials: Science and Devices by the Department of Science and Technology (DST), with initiatives of support to projects related to nanomaterials and their technologies (A. Kumar, 2014). Figure 7 shows the co-authorship between the countries represented by the nodes, the orange circle's size is related to the number of papers.

Figure 7. Network among countries—based on the number of publications relationship between the authors of the countries.



Source: Prepared by the authors.

In continental terms, there is a predominance of the Asian continent, as the origin of the works developed and published, in addition to the interaction of researchers from different countries divided into six main clusters.

3.5 Top 10 WoS research papers as classified by the number of citations

The ten most cited articles are displayed in Table 4. As might be expected, the four papers with more citations are reviews that report the experimental procedures and the products obtained by green synthesis of nanomaterials. Most of the ten main Works deal with silver oxide synthesis.

Table 4. Top 10 cited papers in the Web of Science database.

Paper Title	Authors	Nanomaterial produced or review/ precursor	Citations
A review of the antibacterial effects of silver nanomaterials and potential implications for human health and the environment	Marambio-Jones e Hoek (2010)	Review/ Polysaccharides and Polyphenols	1013
Biological synthesis of metal nanoparticles by microbes	Narayanan e Sakthivel (2010)	Review/ Microorganisms	537
The use of microorganisms for the formation of metal nanoparticles and their application	Mandal et al. (2006)	Review/ Microorganisms	477
Biosynthesis of metal nanoparticles using fungi and actinomycete	Sastry et al. (2003)	Review / Microorganisms	429
Biogenic synthesis of silver nanoparticles and their synergistic effect with antibiotics: a study against gram-positive and gram-negative bacteria	Fayaz et al. (2010)	Silver nanoparticle / Fungus <i>Trichoderma viride</i>	382
Green synthesis of silver nanoparticles using <i>Capsicum annuum</i> L. extract	Li et al. (2007)	Silver Nanoparticles / Plant	356
Silver nanoplates: From biological to biomimetic synthesis	Xie et al. (2007)	Silver Nanoparticles / Algae	274
Identification of active biomolecules in the high-yield synthesis of single-crystalline gold nanoplates in algal solutions	Xie et al. (2007b)	Gold Nanoparticles / Algae- <i>Chlorella vulgaris</i>	172
Green synthesis of biogenic metal nanoparticles by terrestrial and aquatic phototrophic and heterotrophic eukaryotes and biocompatible agents	Narayanan e Sakthivel (2011)	Review / Plants, diatoms and algae	152
Biogenic Synthesis of Metallic Nanoparticles by Plant Extracts	Akhtar, Panwar e Yun (2013)	Review / Plant Extracts	121

Source: Prepared by the authors.

Marambio-Jones e Hoek (2010), wrote a review of the antibacterial properties of silver

nanocomposites. Their text discusses the methodologies of production of the material, including one method of green synthesis where polysaccharides and polyphenols are used as a reactional medium, showing their capacity to reduce the silver ions. The authors note that the reaction mechanism is not understood.

Narayanan e Sakthivel (2010), Mandal et al. (2006) and Sastry et al. (2003) wrote reviews on nanomaterials from microorganisms, including bacteria, actinomycetes, yeasts, fungi and viruses producing materials with a wide variety of sizes and shapes. They emphasize the importance of developing research in which they explore this type of compound preparation, integrated with the concepts of sustainability, reducing the excessive costs of production and the use of hazardous chemicals.

Fayaz et al. (2010) use the fungus *Trichoderma viride*. In the production of silver nanoparticles, which combined with antibiotics combat gram-positive and gram-negative bacteria. The nanomaterials produced have a spherical shape and grain size between 4-40 nm. Through the technique of infrared spectroscopy with Fourier transform (FTIR), the solution composed of the silver nitrate and *Trichoderma viride* was characterized and suggested that the bands between 1425 cm^{-1} , which correspond to the proteins of the fungus, assist in the formation and stabilization of silver nanomaterial.

Li et al. (2007) used the extract of the plant *Capsicum annuum* L. to produce silver nanoparticles with a spherical shape and grain sizes between 30-70 nm. Analyses by FTIR of the extract and the solution with silver nitrate have noted the increase in the peak associated with proteins about Amide I C=O at around 1635 cm^{-1} after the reaction of the water with the nitrate, which indicates its involvement in the stabilization of nanomaterials by proteins.

Xie et al. (2007a) produced silver nanocrystals using algae of the *Chlorella vulgaris* species, obtaining nanoparticles with 20 nm in diameter. They observed that carboxylic and hydroxylic groups from the algae help to reduce the metallic ions. In a later work (Xie et al. (2007b) they used the same precursor to synthesize gold nanoparticles.

In the review written by Narayanan e Sakthivel (2011) nanomaterial synthesis using plants, algae, and diatoms. The high diffusion in the scientific community of the production of silver oxide nanomaterials by the green synthesis technique is observed from the revised articles, being reported in almost all the works referenced by the authors.

Another well-cited paper is the review written by Akhtar, Panwar e Yun (2013) on the use of plant extracts as precursors to produce silver, gold, platinum and palladium nanoparticles. The pH, incubation time and temperature are the main factors in the control of the shape and size of the particles.

3.6 Main keywords

The keywords used in this research (biosynthesis, green synthesis, and nanomaterial) are often followed by ones related to gold and silver nanoparticles (Table 5) and Figure 7. The application of these materials in antibacterial activity has a large number of occurrences in the study sample. The same behavior was observed in the parameter of centrality for the corresponding category of WoS (Biochemistry Molecular Biology, see Table 1).

Table 5. Top 20 Keywords

Keywords	Registration Count
Biosynthesis	93
Silver nanoparticle	78
Green synthesis	57
Gold nanoparticle	50
Nanoparticle	38
Nanomaterial	35
Antibacterial activity	28
Metal nanoparticle	23
Extracellular biosynthesis	22
Extract	20
Biological synthesis	20
Leaf extract	19
Bacteria	19
Escherichia coli	17
Gold	14
Green synthesis	13
Nanotechnology	11
Antibacterial	10
Plant	10
Antimicrobial activity	10

Source: Prepared by the authors.

4. Conclusions

It was observed with the bibliometric analysis that the research related to the subject in the study, green* synthesis and biosynthesis of nanomaterials, has evolved significantly in recent years, the result expressed by the most significant number of published papers and by

the interest of the researchers evidenced in the significant increase in the number of quotes.

The multidisciplinary of the subject under study is seen from the diversity of areas where it appears, including chemistry, materials science and the environment, and applications focused on the medical area. As for the leading periodicals in which the works are published, the same multidisciplinary behavior is observed, characterized by documents in journals of an environmental nature, as well as with sources of the discussion focused on the area of chemistry and materials.

About 82.0 % of the published material in this subject is comprised of papers. More than forty percent of these papers come from India (44.65 %), proving that this country is the current leader in this production methodology.

Six of the ten most cited Works in this subject review, in which a diversity of precursors is for green synthesis and biosynthesis are presented, including plants, bacteria, fungi, plant extracts, and fruits, illustrating the search for new materials and substrates. The most common papers though are on the synthesis of silver oxides due to their large area of application, mainly in biomedicine.

Finally, it is hoped that through the results presented here, the interest and motivation of new researchers to explore different residues for the synthesis of materials, especially the agroindustrial ones, that need a suitable destination according to the directives of the PNRS. Furthermore, in new review studies, it is necessary to expand the databases during refinement to cover a larger number of articles.

5. Acknowledgments

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