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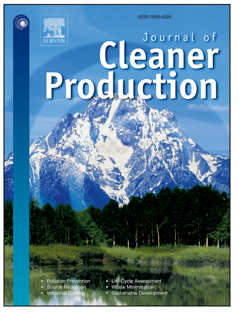


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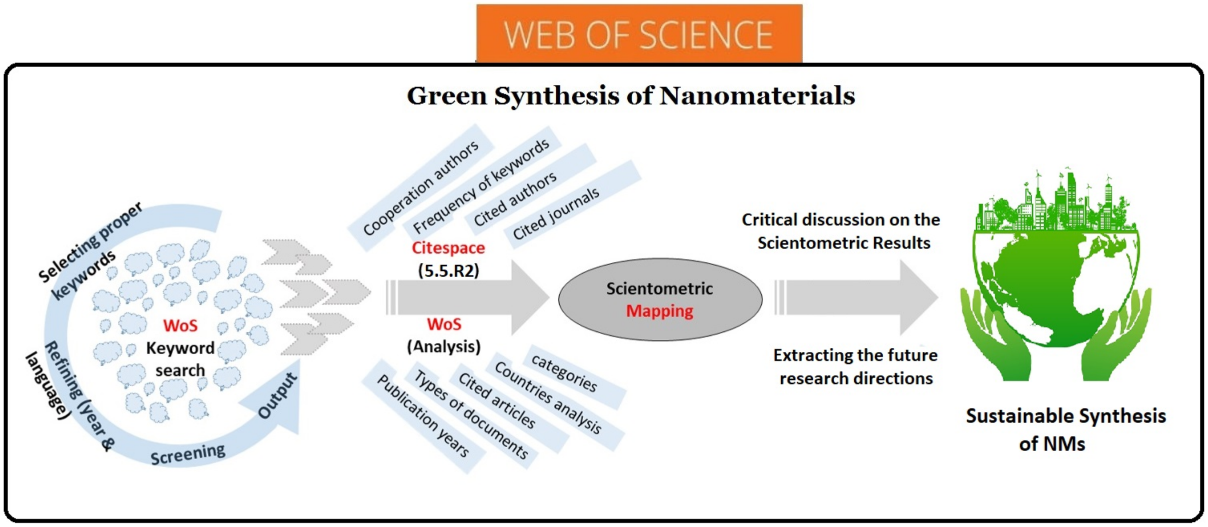
## Credit Author Statement:

**Mohammadreza Khalaj**: Original draft preparation, software and data curation

**Mohammadreza Kamali:** Conceptualization, methodology, writing, reviewing and editing

**M. Elisabete V. Costa:** Supervision**,** Conceptualization, methodology, writing, reviewing and editing

**Isabel Capela:** reviewing and editing


WEB OF SCIENCE
Green Synthesis of Nanomaterials
Selecting proper keywords
Refining (year & language)
Screening
Output
WoS
Keyword search
Cooperation authors
Frequency of keywords
Cited authors
Cited journals
Citespace
(5.5.R2)
WoS
(Analysis)
categories
Countries analysis
Cited articles
Types of documents
Publication years
Scientometric
Mapping
Critical discussion on the Scientometric Results
Extracting the future research directions
Sustainable Synthesis of NMs

# Green Synthesis of Nanomaterials - A Scientometric Assessment

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## Abstract

The green synthesis of engineered nanomaterials (NMs) has deserved an enormous academic interest and huge financial investments during the last decades. However, this prominent position has not been followed by the rapid commercialization of NMs for real applications thus rendering their practical usefulness very doubtful and the appropriateness of novel investments in the field highly questionable. The present manuscript presents the first scientometric study on the green synthesis of NMs aiming to survey the scientific progress in this particular field and identify its main gaps while providing applicable suggestions to facilitate the knowledge transfer from laboratories to real full scale production and applications. The research on green synthesis of nanomaterials published in Web of Science during the period 1991 – 2019 is here carefully analyzed. Overall, 9 scientometric indicators are employed to interpret the results retrieved from the 8761 documents collected. It is found that 107 countries and nearly 22400 authors have contributed to this subject, hence highlighting the relevance of this topic. The keywords spectrum is dominated by the term "nanoparticle" which full adoption takes place at the beginning of the 21st century. Some few years later, a batch of words like "silver nanoparticle", "gold nanoparticle" and "nanocomposite" reaches a significant impact reflecting the emergence of commercial applications for these nanomaterials. It is only in 2009 that the keyword "green synthesis" gains strength, followed then by "biosynthesis" in 2010, making it evident a trend towards environmentally friendly reagents. The number of publications on green synthesis of nanomaterials displays up to now a sigmoidal like growth pattern, which points actually to a decrease on new arrivals, thus suggesting a possible forthcoming decline in this field. However, the analysis carried out in the present work allows identifying various gaps related to sustainability, which, if appropriately addressed, may contribute to a resurgence of

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the research on nanomaterials synthesis while fostering more frugal approaches on material synthesis tendencies.

**Keywords:** Green Synthesis, Scientometric Study, Sustainable Synthesis, Nanomaterials.

## 1. Introduction

Among the green strategies recently reported for the synthesis of nanomaterials (NMs), different approaches are identified, which normally include either the use of green starting raw materials such as natural polymers like chitosan (Benelli, 2019; Choo et al., 2016; Skiba et al., 2020) and plants extracts (Fierascu et al., 2019; Rajendaran et al., 2019; Saha et al., 2017), or the so-called green synthesis routes such as ultrasonic irradiation (Mosaddegh, 2013; Sadjadi et al., 2017; Zheng et al., 2013) and microwave-assisted methods (Mahmoud et al., 2015; Mahmoud and Nabil, 2017), or a combination of methods (Burgaz et al., 2019; Yang et al., 2019). The adoption of green production routes may bring several advantages over conventional methods. For instance, conventional methods generally rely on expensive chemical reagents, tending to generate solid or liquid wastes which imply extra investments for their disposing and treatment purposes (Hwang et al., 2011; Kamali et al., 2019c; Rafique et al., 2017; Singh et al., 2018). Such drawbacks have raised some debate on the efficiency of the methods commonly utilized for producing NMs, as economic considerations are also of high importance when selecting an appropriate synthesis method. In this regard, besides the specifications required for full efficiency such as relatively short reaction times, low temperature and environmental friendly synthesis media, other criteria associated to sustainability, including technical-environmental (like low risk for operators and society), social (public acceptability and contribution for job creation) and economic considerations (initial investment, costs of raw materials and manufacturing), have also to be taken into account (Kamali, 2020; Kamali et al., 2019c)). Nevertheless, studies on nanomaterials synthesis meeting all these criteria are scarce or inexistent. Therefore, green synthesis routes, with better environmental, economic and technical feasibility than conventional methods, are now timely needed.

The profile of the publication rate in the field of NMs green synthesis evidences a trend for saturation, although a corresponding widespread use of NMs is not yet found, as expected. This gap may suggest a deficit on the knowledge transfer from academic sectors to industry and society, which detrimental effects should not be underestimated. It is commonly known that the application of research results produces a feedback amplification effect on the research dynamics itself, fostering further developments, hence feeding a vicious circle correlation between both (de Wit-de Vries et al., 2019). Conversely, a low rate of knowledge transfer from lab-scale to real-scale applications, with a small fraction of lab-scale technologies being commercialized, will produce negative impacts on the investments necessary to the progress of the research thus slowing down novel scientific achievements (D'Este and Perkmann, 2011). Therefore, it is critically important to address the missing conditions that could render the actual research results in the field of NMs green synthesis totally useful and applicable for industry and the community in general. Scientometrics may provide important guidelines for that purpose.

Scientometric studies have been frequently used in order to study and measure the research progress and efforts from researchers, governments, research institutes, universities, faculties and scientific publishers and journals in a particular scientific subject (Konur, 2012). The scientometric methodology provides a general overview of the science philosophy via the recognition of the general direction followed by the developments in a specific area during a certain period. This is normally performed by analyzing the results of the research carried out using mathematical equations (Ahmad and Thaheem, 2017; Olawumi and Chan, 2018). The number of scientometric studies on various scientific fields has increased considerably in the more recent years, hence demonstrating the importance of surveying the knowledge progress. Sustainability and sustainable development (Olawumi and Chan, 2018), industrial wastewater treatment methods (Jiang et al., 2018), electromagnetic studies (Bernabò et al., 2017), regenerative medicine (Chen et al., 2012), heavy metals release from agricultural watershed to aquatic system (Ouyang et al., 2018), and $CO_2$ underground storage (Davarazar et al., 2019) among others, are few examples of recent scientometric studies. By overviewing the state-of-the-art evolution in a particular scientific field, its main trends and associated gaps can be identified, thus enabling propositions to overcome the barriers to the progress of science and technology in that specific area. In what NMs green synthesis is concerned, its up-scaling to industrial contexts and easy commercialization is far from full attainment (Cai et al., 2019), being this a potential adverse condition for the maintenance of research budgets. In this sense, scientometric studies may provide useful information to clarify this problem and point out possible solutions. However, to the best of our knowledge, there are no literature reports on scientometric studies covering green synthesis methods for NMs production, despite the high number of publications on this field that can provide useful information to promote green fabrication processes. Considering that the technologies already developed at lab-scales are currently mature for transference, it seems crucial to overview the progress made in this field, in order to identify the barriers to the rapid commercialization of those technologies that have proved their efficiency for real applications. Therefore, the present study aims at a scientometric analysis of the green synthesis of NMs in order to assess the efforts already made in the area of knowledge transfer, while addressing its hindering factors. For that purpose, sustainability criteria (i.e. technical, environmental, social and economic ones) are here used to frame the discussion of scientometric data, aiming to promote the real and sustainable application of engineered nanomaterials.

## 2. Methodology

In this study, the Web of Science (WoS) Core Collection database was used to collect data in the span of time 1991–2019, considering all the citations indexed in Science Citation Index Expanded (SCI-EXPANDED). A specific combination of keywords based on an advanced search in the WoS database (see supplementary information), was utilized. In order to identify the appropriate combination of keywords that could be representative of the relevant literature, a search based on a preliminary list of keywords corresponding to the topic of the present study (green, synthesis, and nanomaterials) was performed allowing to extract a primary list of papers published in this area. This list was used to identify and refine the keywords with higher relevance, and then their combination to be used in the final WoS search. The

WoS outputs were saved as “Marked List” and then saved as “text” format, in order to be further used in CiteSpace software (version 5.5.R2) according to the manual provided by CiteSpace (Chen, 2017, 2014, 2005). A schematic of the process implemented in this study is presented in Fig. 1.

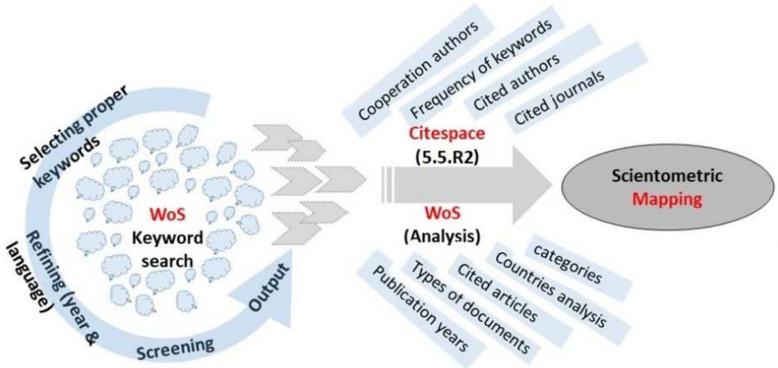


**Fig. 1.**
A schematic of the research design.

Nine scientometric techniques were utilized including analysis of (1) publications, (2) document types, (3) contribution of the countries, (4) authors, (5) keywords, (6) cited authors, (7) cited journals, (8) categories, and (9) cited articles. To proceed with the analysis, the parameters used in this study for the scientometric analysis are further elaborated as follows.

### 2.1 Scientometric parameters

*a) Betweenness Centrality (BC)*

Betweenness Centrality, introduced in 1948, is considered as one of the most critical metric parameters for the specification of every node in a network. BC qualifies the centrality of a node by indicating the extent to which it is located in the shortest line between others (Freeman, 1997). It is a subdivision concept of graph theory demonstrating the alternation of a node in a diagram. This parameter is represented as BC(k) when standing for the node k. It is calculated by Eq. (1).

$$BC\ (k) = \sum_{i\neq j\neq k} \frac{\Delta_{ij}(k)}{\Delta_{ij}} \qquad (1)$$

Where Δij is the number of shortest links between ending nodes i and j, and Δij(k) represents the number of the shortest links that cross the node k. If a node contains high BC, it can be concluded that it is located on a significant fraction of shortest links, meaning that it contains many connections with other nodes. BC of all the nodes are values lying in the range [0,1]. When BC of any intermediate node is maximum, it has the value 1; its value will be zero whenever only one line connects two ending nodes and at the same time, BC of other nodes is minimum. Likewise, if there are n lines connecting two ending points, those two ending points will provide the value of BC/n for all the intermediate nodes (R et al., 2014).

*b) Citation Burst (CB)*

According to Chen (2014) the “citation burst” is an indicator that identifies the most active area (including certain references, authors, etc.) in a specific scientific field according to Kleinberg (2003). Simply, if there are n batches of documents, and the $t^{th}$ batch contains $r_t$ relevant documents out of a total of $d_t$, then citation burst (CB) is defined as:

$$CB(i, r_t, d_t) = -\ln\left[\binom{n}{k} P_i^{r_t}(1-P_i)^{d_t - r_t}\right] \qquad (2)$$

since this is the negative logarithm of the probability that $r_t$ relevant documents would be generated using a binomial distribution with probability $p_i$.

*c) Sigma*

Sigma is an indicator that combines the strength of the structural and temporal properties of a node in scientometric graphs (Chen, 2014). In other words, it combines betweenness centrality and citation burst to measure the scientific novelty of a reference. Sigma is calculated according to Eq. 3 (Chen et al., 2011);

$$\text{Sigma} = (BC + 1)^{CB} \qquad (3)$$

*d) Citation Counts (CC)*

Citation count indicator measures the number of citations that a certain publication has received during a certain period of time. Three main databases can be utilized to provide the CCs including WoS, Scopus, and Google Scholar. CC can be defined for authors, for an individual article, and for a particular journal (Lindner et al., 2010; O’Brien, 2019). A high CC indicates that an author, an article, or a journal has deserved high attention from the scientific community (Leimu and Koricheva, 2005).

*e) Citation Frequency (CF)*

Citation frequency is defined as the total number of citations received by a certain publication during a certain period of time divided by the citation period (years) (United States Environmental Protection Agency, 2004).

*f) Clustering*

When the group under analysis is divided into some sub-categories with certain similarities, this process is called “clustering”. In other words, each cluster contains data possessing similar characteristics and at the same time, these characteristics may differ among clusters. The strength of a cluster is rated as “#x” where x may assume integer values (0, 1, 2, ... etc.), depending on the similarity of the cluster data: data very similar will define a very strong cluster, being the strongest cluster identified as “cluster#0”; clusters #1, #2, ... etc are considered to display a decreasing strength as compared to cluster#0 (Cornish, 2007).

## 3. Results

A combination of keywords including synthesis, fabrication, preparation, green, sustainable, ultrasonic, microwave, sonochemistry, nano, and nanomaterial was used in this scientometric study. The keywords search was also denoted with a “*” (see the supplementary information) refined with the English language and a time period ranging from 1991 to 2019 resulted in 8761 WoS records. The analysis of the main parameters including publication history (section 3.1), document type (section 3.2), countries contribution (section 3.3), authors (section 3.4), keywords (section 3.5), cited authors (section 3.6), cited journals (section 3.7), categories (section 3.8), and cited articles (section 3.9) regarding the extracted WoS records will be next presented.

### 3.1 Publication history

Fig. 2 represents the time evolution of the number of published documents on NMs green synthesis (including scientific articles, review papers, proceeding papers, meeting abstracts and other types of documents) during the studied time period (1991-2019). According to this figure, the largest record belongs to the year 2019 with a total number of 1139 documents published, whereas only one document was published in 1991, thus demonstrating that scientific efforts in this field have started to grow in the 1990s. Also, the cumulative number of published documents follows a sigmoidal like pattern indicating that the growth rate of the number of publications in this field is tending to a slight slowdown**.**

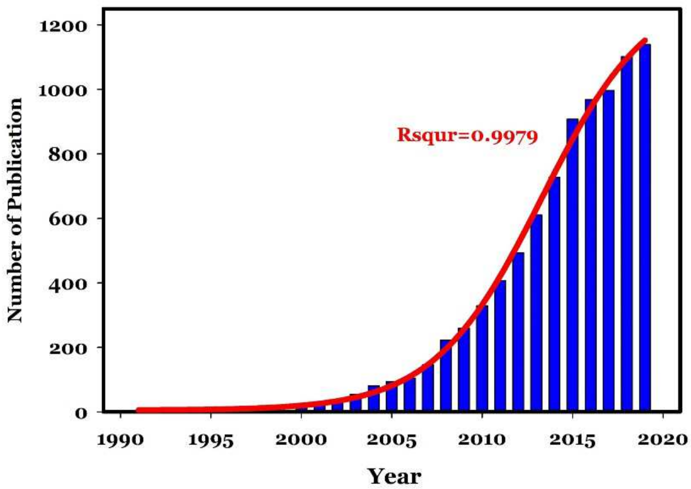


**Fig. 2**
Distribution of the published documents during 1991-2019 and sigmodial pattern of the cumulative number of publications over the studied period of time.

### 3.2 Document type

The contributions (in %) of the various documents published in the field under analysis, including proceeding papers (PP), meeting abstracts (MA), articles (A), reviews (R) and other type of documents (O) such as corrections, letters, editorial material, retracted publications, news items, retractions, data papers and notes based on the results of searching the subject "green synthesis of nanomaterials" in WoS database, are presented in figure 3. As observed, articles share 87.60% of the documents published in this field while review articles and other document types represent a percentage of only 1.54% and 1.41% respectively. Proceeding papers (6.76%) and meeting abstracts (2.69%) share almost 10% of the published documents.

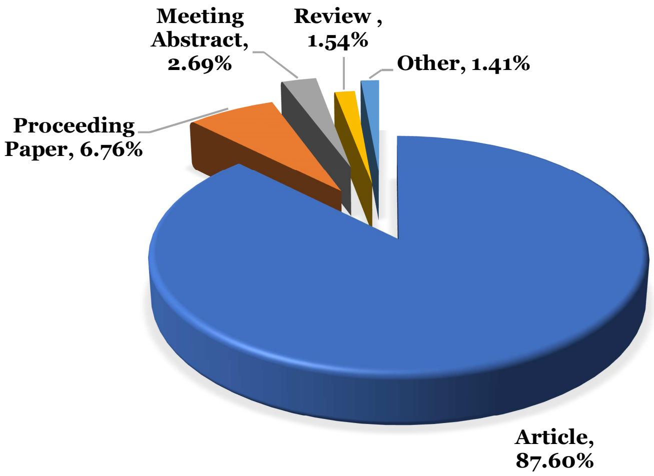


**Fig. 3.**
Shares of the various document types published on the green synthesis of NMs since 1990s. Articles with a share of 87.60% which are by far the documents with the highest share suggest that scientists in this field prefer to publish their works as indexed papers over other kind of scientific documents such as conference papers or meeting abstracts.

### 3.3 Contribution of the countries

Fig. 4. and Table 1. illustrate the outputs of WoS regarding the contribution of various countries for generating scientific publications on green synthesis of NMs. According to these results, 107 countries have so far contributed to the publication of various types of documents in this field. The republic of China with 2151 scientific documents shares the highest number of publications among the contributing countries. India with 1973 scientific documents and Iran with 1644 scientific documents have received the second and third positions, respectively; USA, South Korea, Saudi Arabia, Japan, Malaysia, Taiwan, and Egypt with 685, 556, 260, 247, 205, 187, and 171 documents, respectively, were ranked 4th to 10th place.

**Table 1.**
Contributions of various countries to the production of scientific documents on green synthesis of nanomaterials.

| Rating | Country | Counts |
|---|---|---|
| 1 | China | 2151 |
| 2 | India | 1973 |
| 3 | Iran | 1644 |
| 4 | USA | 685 |
| 5 | South Korea | 556 |
| 6 | Saudi Arabia | 260 |
| 7 | Japan | 247 |
| 8 | Malaysia | 205 |
| 9 | Taiwan | 187 |
| 10 | Egypt | 171 |

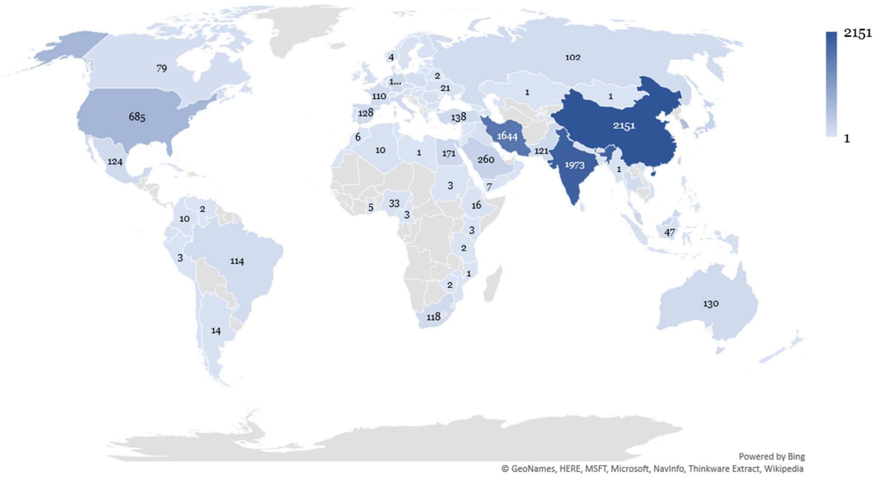


**Fig. 4.**
Worldwide countries contributions for the production of scientific documents on green synthesis of NMs. China, India, and Iran have the highest shares in this scientific area.

### 3.4 Authors

The results extracted from CiteSpace software were used for analyzing the contributions of the authors, which are listed in table 2 and graphically presented in Fig. 5. A total number of 22338 authors have contributed to the field of green synthesis of NMs. As observed, Salavati-Niasari with 107 documents, Morsali with 104 documents, and Gedanken with 55 documents published in WoS have achieved the first three ranks. Collaborations among groups are also clearly identified although some isolated groups lacking interconnection are noticed as well.

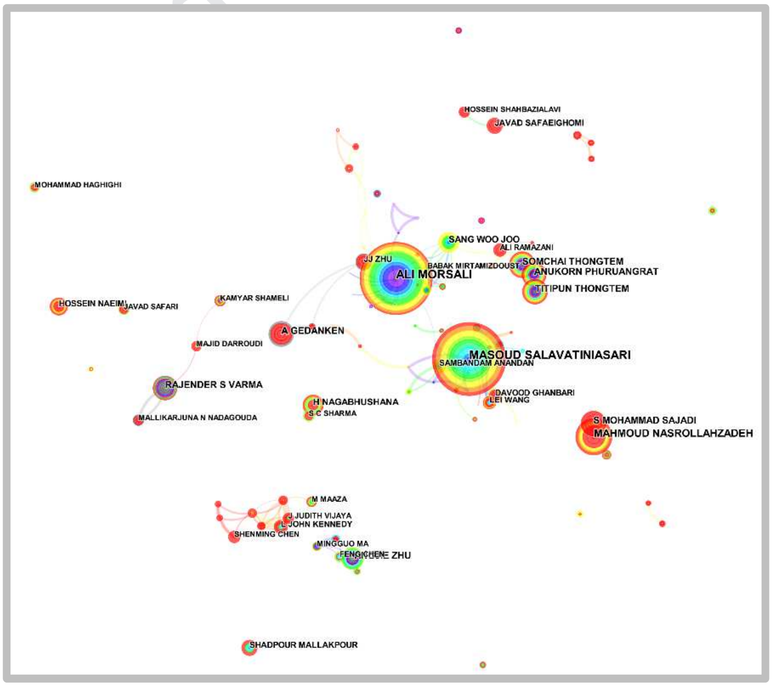


**Fig. 5.**

Graphical outcomes from CiteSpace showing the authors' contributions as a network containing both nodes and links in which each symbolic node refers to an author and each link is used to interpret the pattern of cooperation among the authors (Olawumi and Chan, 2018). Authors like Salavati-Niasari and Morsali, stand out as having the highest contributions.

**Table 2.**

List of authors with relevant contributions for the production of scientific documents on green synthesis of nanomaterials. Salavati-Niasari, and Morsali have the highest contributions among the authors.

| Rating | Authors | Counts |
|---|---|---|
| 1 | Salavati-Niasari M. | 107 |
| 2 | Morsali A. | 104 |
| 3 | Gedanken A. | 55 |
| 4 | Nasrollahzadeh M. | 54 |
| 5 | Varma R.S. | 52 |
| 6 | Zhu Y.J. | 48 |
| 7 | Zhu J.J. | 47 |
| 8 | Wang Y. | 45 |
| 9 | Thongtem S | 43 |
| 10 | Wang J. | 43 |

### 3.5 Keywords

Keywords analysis is considered an essential tool to characterize a scientific domain in a specific field (Shrivastava and Mahajan, 2016; Su and Lee, 2010). The network of keywords can also reveal how they are related to each other and how they can be used for identifying the most suitable document among existing publications in a scientific area (Darko et al., 2019). WoS database covers two types of keywords including "author keywords" which are provided by the authors, and "keywords plus" that are retrieved from journal indexes. In this study, both types of keywords have been utilized (Zhao, 2017). Relying on CiteSpace software outputs, the keywords with the highest frequency were identified as follows: nanoparticle (frequency: 2148), silver nanoparticle (frequency: 1098), and green synthesis (frequency: 973). Table. 3 and Fig. 6 present the results of the keywords analysis and Fig. 7 traces the keywords development during the studied period of time (1991-2019).

**Table 3.**

Analysis of keywords occurrence. "Nanoparticle" stands out as the keyword with the highest frequency.

| Rating | Keyword | Sigma | Centrality | Burst | Frequency |
|---|---|---|---|---|---|
| 1 | Nanoparticle | 1 | 0.1 | | 2148 |
| 2 | Silver nanoparticle | 1 | 0.03 | | 1098 |
| 3 | Green synthesis | 1 | 0.02 | | 973 |
| 4 | Nanostructure | 1 | 0.07 | | 785 |
| 5 | Nanocomposite | 1 | 0.11 | | 701 |
| 6 | Biosynthesis | 1 | 0 | | 699 |
| 7 | Gold nanoparticle | 1 | 0.04 | | 657 |
| 8 | Growth | 117.82 | 0.14 | 35.63 | 637 |
| 9 | Nanocrystal | 1.46 | 0.04 | 8.86 | 627 |
| 10 | Composite | 1 | 0.06 | | 574 |
| 11 | Reduction | 1 | 0.06 | | 560 |
| 12 | Particle | 18.47 | 0.11 | 29.2 | 501 |
| 13 | Performance | 1 | 0 | | 474 |
| 14 | Size | 1 | 0.05 | | 470 |
| 15 | Catalyst | 1 | 0.08 | | 462 |
| 16 | Fabrication | 1 | 0.02 | | 455 |
| 17 | Optical property | 1 | 0.05 | | 448 |
| 18 | Microwave | 1.07 | 0.02 | 3.86 | 431 |

| | | | | | |
|---|---|---|---|---|---|
| 19 | Oxide | 1 | 0.07 | | 409 |
| 20 | Nanorod | 3.11 | 0.04 | 25.89 | 407 |

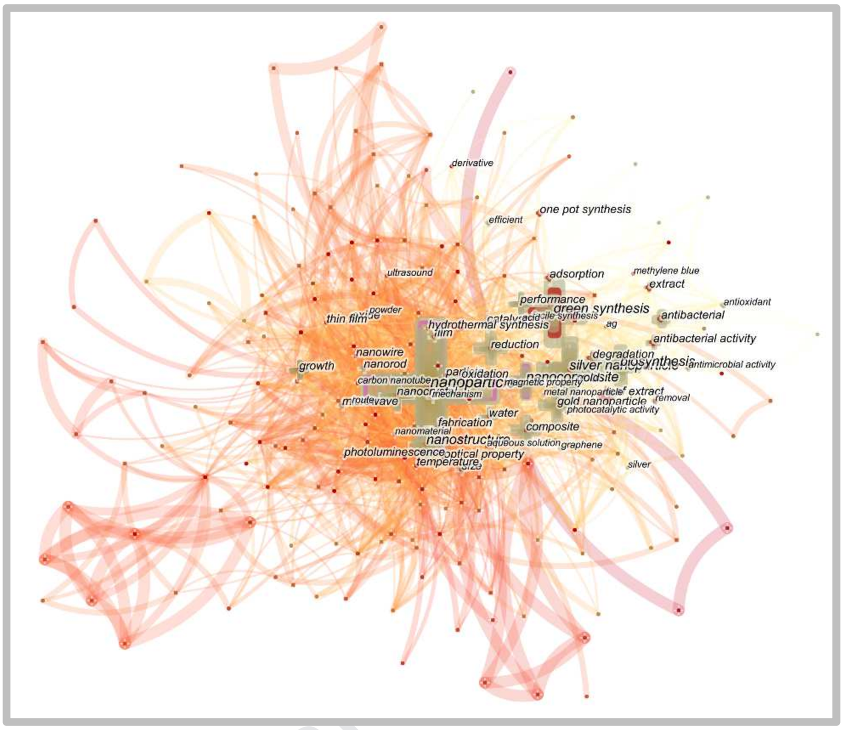


**Fig. 6.**
Keywords occurrence pattern. The size of nods reflects the frequency of each keyword (Olawumi and Chan, 2018). The keywords "Nanoparticles", "Silver nanoparticle", and "Green synthesis" display the highest frequencies.

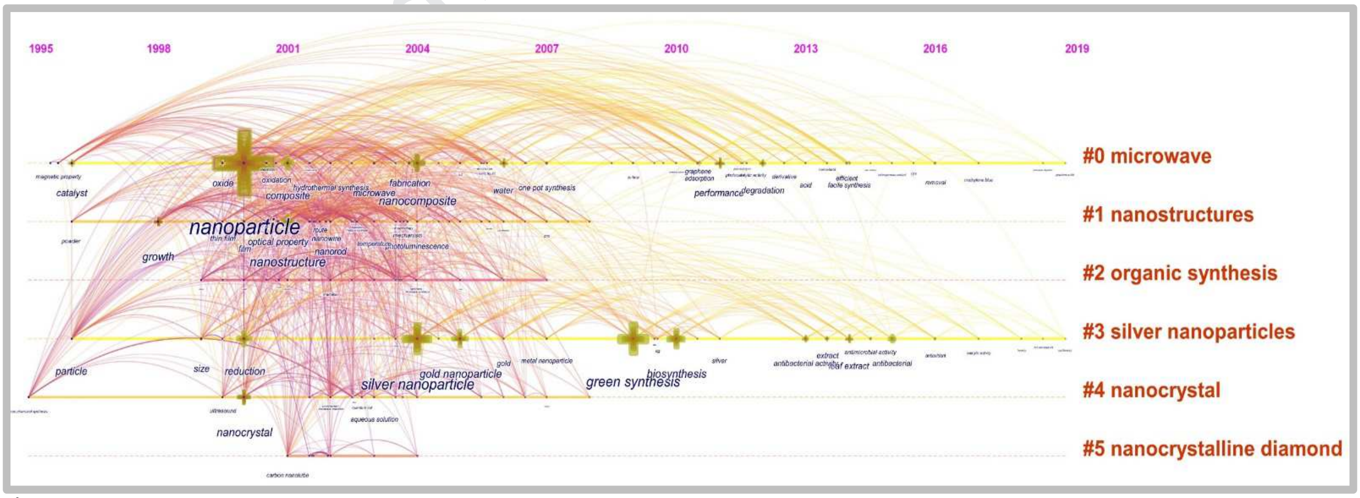


**Fig. 7.**
Evolution trend of keywords during the studied period (1991-2019). The size of nods indicates the frequency of keywords occurrence in the studied documents during the mentioned period of time. The keywords "nanoparticle" in the cluster #0, "silver nanoparticle", and "Green synthesis" in the cluster #3 display the highest frequencies that are found in the first decade of the 21st century.

### 3.6 Cited authors

Identifying the authors' collaboration networks in a specific scientific area allows distinguishing both the experts in the field with the highest contributions and the main funding organizations (Ding, 2011;

Hosseini et al., 2018). In addition, this tool can reveal the collaborative efforts among the authors, and the author's impact on the scientific area under consideration. These outcomes can be identified using variables such as citation quantity, citation burst, betweenness centrality, and citation sigma. Table 4, and Figure 8 presents the results achieved for cited authors' analysis using CiteSpace program. According to the authors' citation frequency, the top-ranked author is Suslick K.S. (frequency = 549). The second place belongs to Wang Y. (frequency = 460), Shankar S.S. (frequency = 441), and Raveendran P. (frequency = 335) owned the next positions. Furthermore, M. Nasrolazadeh (2017-2019) in cluster #3 with burst of 58.89, S. Komarneni (2003-2012) in cluster #8 with the burst of 54.25, and S. Ahmed (2017-2019) in cluster #3 with the burst of 53.95 have been identified the top ranked authors in terms of citation burst strengths. In addition, the centralities for M. Nasrolazadeh in cluster #3, S. Komarneni in cluster #8, and S. Ahmed in cluster #3 the centralities were measured 0.00. Finally, the top-ranked authors according to the calculated sigma values are M. Nasrolazadeh in cluster#3, S. Komarneni in cluster #2, and S. Ahmed in cluster #3 with a sigma of 1.00. The largest cluster #0 is labeled as "ionic liquid" with 80 members. The second position belongs to cluster #1 labeled as "coordination polymer" with 66 members. Finally, cluster # 2 is located in the third position and labeled as "microwave-polyol method" with 51 members.

**Table 4.**
CiteSpace results regarding the analysis of cited authors. Suslick K.S. is the author with the highest citation frequency.

| Rating | Author | Year | Sigma | BC | CB | CF |
|---|---|---|---|---|---|---|
| 1 | Suslick K.S. | 1995 | 1 | 0 | 1.29 | 549 |
| 2 | Wang Y. | 2000 | 1 | 0 | | 460 |
| 3 | Shankar S.S. | 2009 | 1 | 0 | 14.67 | 441 |
| 4 | Raveendran P. | 2006 | 1 | 0 | 1.19 | 335 |
| 5 | Wang J. | 2007 | 1 | 0 | 1.37 | 303 |
| 6 | Zhang Y. | 2006 | 1 | 0 | 20.02 | 302 |
| 7 | Zhang J. | 2005 | 1 | 0 | 0.47 | 299 |
| 8 | Wang X. | 2005 | 1 | 0 | 0.47 | 290 |
| 9 | Zhang H. | 2007 | 1 | 0 | 2.07 | 290 |
| 10 | Wang H. | 2002 | 1 | 0 | 1.26 | 288 |

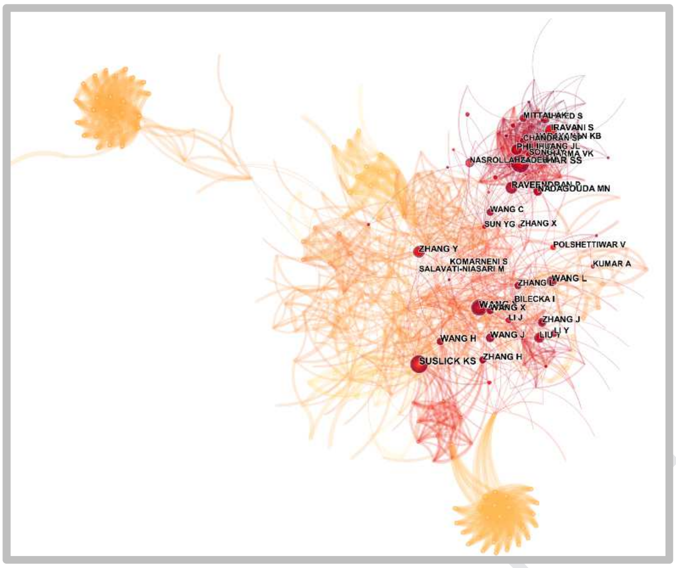


**Fig. 8.**
Cited authors schematic where the citation frequency for each author is shown by the node size (Olawumi and Chan, 2018). As observed, the authors “Suslick K.S.”, “Wang Y.”, and “Shankar S.S.” have the highest citation frequencies.

### 3.7 Cited journals

Table 5 and Figures 9 and 11 present the outputs of CiteSpace regarding the analysis of cited journals based on various parameters such as the number of journal citations, citation burst, betweenness centrality, and finally the citation sigma. According to the obtained results respecting journal citations, the top-ranked journal is “Journal of the American Chemical Society” with a frequency of 3728. The second rank journal is “Materials Letters” with a frequency of 3048, followed by “Chemistry of Materials” in the third rank with a frequency of 3016. In terms of the citation bursts strength, RSC Advances (2017-2019) in cluster #2 with a burst of 223.64, Journal of Crystal Growth (2001-2013) in cluster #0 with a burst of 167.04, and Chemical Physics Letters (2000-2012) in cluster #0 with a burst of 154.96 are the top-ranked journals in this scientific field. In addition, when considering the betweenness centrality, Materials Letters in cluster #1 with a centrality of 0.13, Angewandte Chemie International Edition in cluster #2 with a centrality of 0.13, and Advanced Materials in cluster #2 with a centrality of 0.12 are the top-ranked journals. Regarding citation sigma, Journal of the American Ceramic Society in cluster#1 with a sigma of 18930, RSC Advances in cluster #2 with a sigma of 12323, and The Journal of Physical chemistry in cluster# 0 with a sigma of 4092 are ranked in the top. The largest cluster (#0) is labeled as “nanocrystalline diamond film” with 35 members. Cluster #1 is labeled as “nanoparticles synthesis” with 33 members and is in second place.

**Table 5.**
CiteSpace results regarding the analysis of cited journals. Journal of the American Chemical Society has the highest citation frequency.

| Rating | Journal | Year | Sigma | BC | CB | CF |
|---|---|---|---|---|---|---|
| 1 | Journal of the American Chemical Society | 1996 | 1.06 | 0.04 | 1.38 | 3728 |
| 2 | Materials Letters | 2001 | 1 | 0.13 | | 3048 |
| 3 | Chemistry of Materials | 1996 | 2.26 | 0.09 | 9.76 | 3016 |
| 4 | Journal of Materials Chemistry | 1999 | 1.29 | 0.05 | 5.09 | 2799 |

| | | | | | | |
|---|---|---|---|---|---|---|
| 5 | The Journal of Physical Chemistry C | 2008 | 1 | 0.03 | | 2771 |
| 6 | Langmuir | 1997 | 1.26 | 0.08 | 2.85 | 2655 |
| 7 | The Journal of Physical Chemistry B | 2000 | 5.61 | 0.04 | 44.32 | 2645 |
| 8 | Advanced Materials | 1995 | 1.36 | 0.12 | 2.62 | 2571 |
| 9 | Angewandte Chemie International Edition | 1996 | 1 | 0.13 | | 2392 |
| 10 | Chemical Communications | 1999 | 1.11 | 0.11 | 0.98 | 2269 |

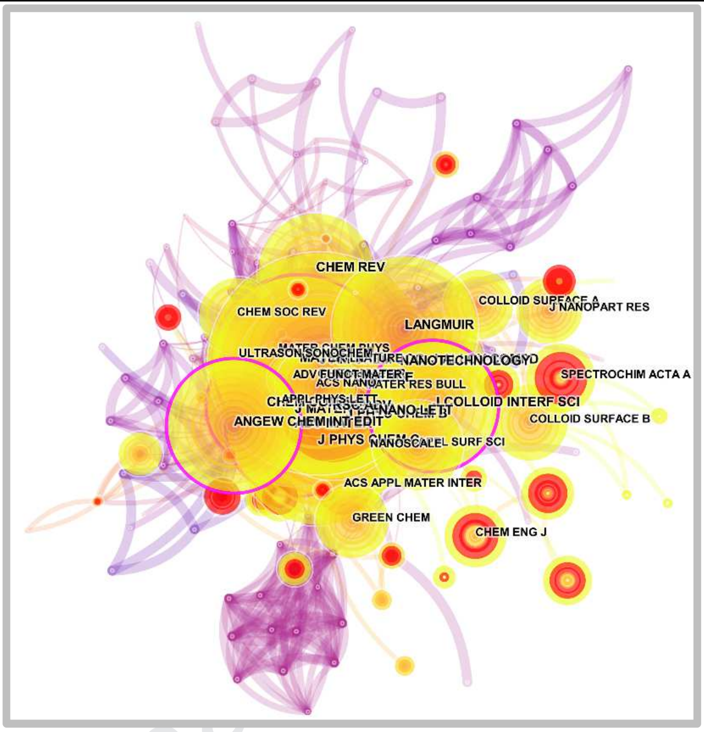


**Fig. 9.**
Outputs of CiteSpace regarding cited journals. The journals “American Chemical Society “and “Materials Letters” to which correspond the largest nodes are the most cited journals.

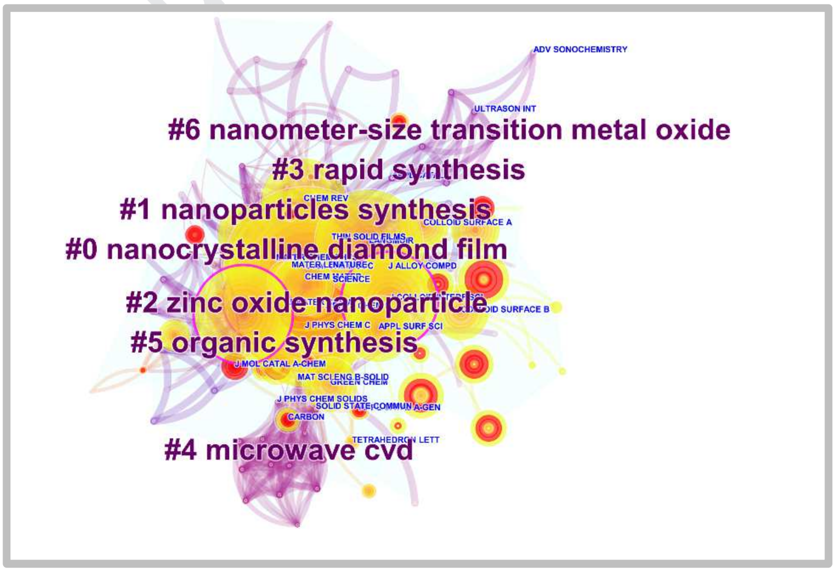


**Fig. 10.**
CiteSpace software outcomes on clusters of cited journals. In this figure, cluster#0 (nanocrystalline diamond film) contains the highest number of journals that have published documents analyzed in this study.

## 3.8 Categories

The documents published in WoS are classified, at least, in one specific category. The top ten categories based on the number of documents are listed in Table 6.

**Table 6.**
Rating of documents categories based on the documents number. As shown "Materials Science, Multidisciplinary" is the category with the highest number of documents.

| Rating | Categories | Number of documents |
|---|---|---|
| 1 | Materials Science, Multidisciplinary | 2776 |
| 2 | Chemistry, Multidisciplinary | 2371 |
| 3 | Chemistry, Physical | 1308 |
| 4 | Physics, Applied | 1302 |
| 5 | Nanoscience & Nanotechnology | 1202 |
| 6 | Physics, Condensed Matter | 680 |
| 7 | Engineering, Chemical | 614 |
| 8 | Chemistry, Inorganic & Nuclear | 433 |
| 9 | Acoustics | 328 |
| 10 | Materials Science, Ceramics | 312 |

## 3.9 Cited articles

The results achieved via WoS in terms of the top 10 highly cited articles during the period ranging from 1991 to 2019 are presented in Table 7 and Fig. 11.

**Table 7.**
The more highly cited articles in the field under analysis. The article entitled " Silver nanoparticles: Green synthesis and their antimicrobial activities" stands out as the most cited one (Sharma et al., 2009).

| Rank | Title of the article | Total Citations | Ref. |
|---|---|---|---|
| 1 | Silver nanoparticles: Green synthesis and their antimicrobial activities | 1996 | (Sharma et al., 2009) |
| 2 | A Green Approach to the Synthesis of Graphene Nanosheets | 1614 | (Guo et al., 2009) |
| 3 | Completely green synthesis and stabilization of metal nanoparticles | 1413 | (Raveendran et al., 2003) |
| 4 | Green synthesis of metal nanoparticles using plants | 1008 | (Iravani, 2011) |
| 5 | Reducing Sugar: New Functional Molecules for the Green Synthesis of Graphene Nanosheets | 982 | (Zhu et al., 2010) |
| 6 | Applications of Ultrasound to the Synthesis of Nanostructured Materials | 968 | (Jin et al., 2010) |
| 7 | Microwave synthesis of fluorescent carbon nanoparticles with electrochemiluminescence properties | 778 | (Zhu et al., 2009) |
| 8 | Microwave chemistry for inorganic nanomaterials synthesis | 660 | (Bilecka and Niederberger, 2010) |
| 9 | Photochemical green synthesis of calcium-alginate-stabilized Ag and Au nanoparticles and their catalytic application to 4-nitrophenol reduction | 641 | (Saha et al., 2010) |
| 10 | Economical, green synthesis of fluorescent carbon nanoparticles and their use as probes for sensitive and selective detection of mercury(II) ions | 615 | (Lu et al., 2012) |

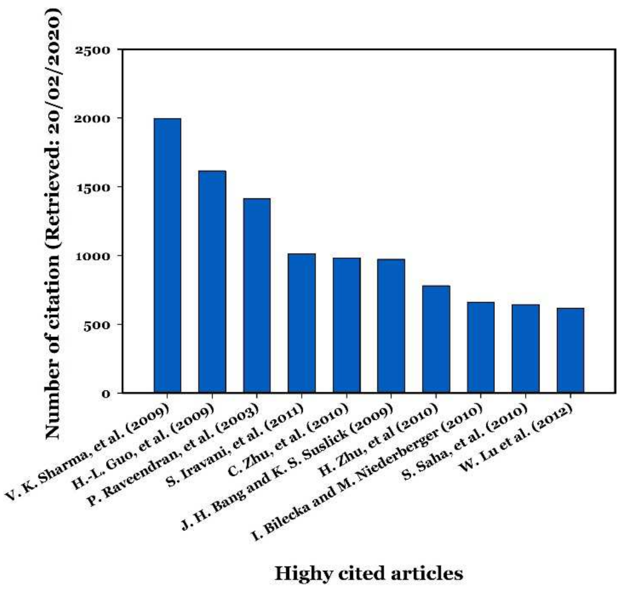


**Fig. 11.**
Graphical presentation of WoS results in terms of the top 10 highly cited articles during the period ranging from 1991 to 2019.

## 4. Discussion

In the present section an historical background covering the synthesis of nanomaterials will be firstly provided in order to frame a comprehensive definition of the term "green synthesis" whose evolution over the years will be subsequently traced and discussed.

Synthesis of nanomaterials is thought to have been introduced by Michael Faraday (1850s) while preparing activated gold nanoparticles (Edwards and Thomas, 2007; Heiligtag and Niederberger, 2013; Jeevanandam et al., 2018; Tweney, 2006). Faraday demonstrate the differences in optical properties between bulk and nano-sized colloidal gold particles (Heiligtag and Niederberger, 2013). At that time Faraday was not able to prove by direct methods the nanometric size of the particles he had synthesized. It was only a century later that an innovative technique (electron microscopy) allowed disclosing the nanometric size of Faraday colloidal gold which average particle size ranged from 3 to 30 nm (Edwards and Thomas, 2007). Richard Zsigmondy, which research in colloids awarded him a Nobel Prize in Chemistry in 1925, introduced the concept of "nanometer" in 1925 by. The modern nanotechnology, however, was founded later in 1959 by Feynman with his famous words: "there is plenty of room at the bottom" (Hulla et al., 2015). Later, in the 1970s, Norio Taniguchi used the word "nanotechnology" to describe semiconductors processing with a nanometer-scale precision level. These achievements were precursor milestones of Nanotechnology which flourishing period took place in the 1980s. Kroto, Smalley, and Curl in the 1980s discovered the fullerenes and Eric Drexler wrote the book entitled "Engines of Creation: The Coming Era of Nanotechnology" published in 1986 (Hulla et al., 2015). The last decade of the twentieth century, driven by a growing awareness of the need for pollution prevention, seeded the first steps in the "green synthesis" of NMs. The evolution of NMs green synthesis until the present days is mapped and analyzed in the discussion that follows, and its important milestones presented in Table 8 and Figure 12.

**Table. 8.** Milestones on the evolution of nanomaterials green synthesis over the period 1991-2019.

| Time Period | Remarks | Number of publications | Leading Country [a.] |
|---|---|---|---|
| 1991-2000 | - First document published in WoS on NMs green synthesis. | 54 | USA |
| 2001-2005 | - Introduction of the "Green Synthesis" term to the scientific community | 293 | China |
| 2006-2010 | - Remarkable advances in NMs green synthesis methods mostly due to green methods such as biosynthesis<br>- Manipulation of NMs green synthesis conditions to improve the properties of NMs | 1062 | China |
| 2011-2014 | - Promoting NMs synthesized by green methodologies for various applications such as environmental clean-up, drug delivery, and energy<br>- Studies on the mechanisms of green synthesis methods. | 2238 | China |
| 2015-2019 | - Promoting the biosynthesis of NMs<br>- Green synthesis of NMs in compliance with environmental protection agency (EPA) green chemistry's 12 principles. | 5114 | India |

a. The countries with the highest number of Publication in the period under study.

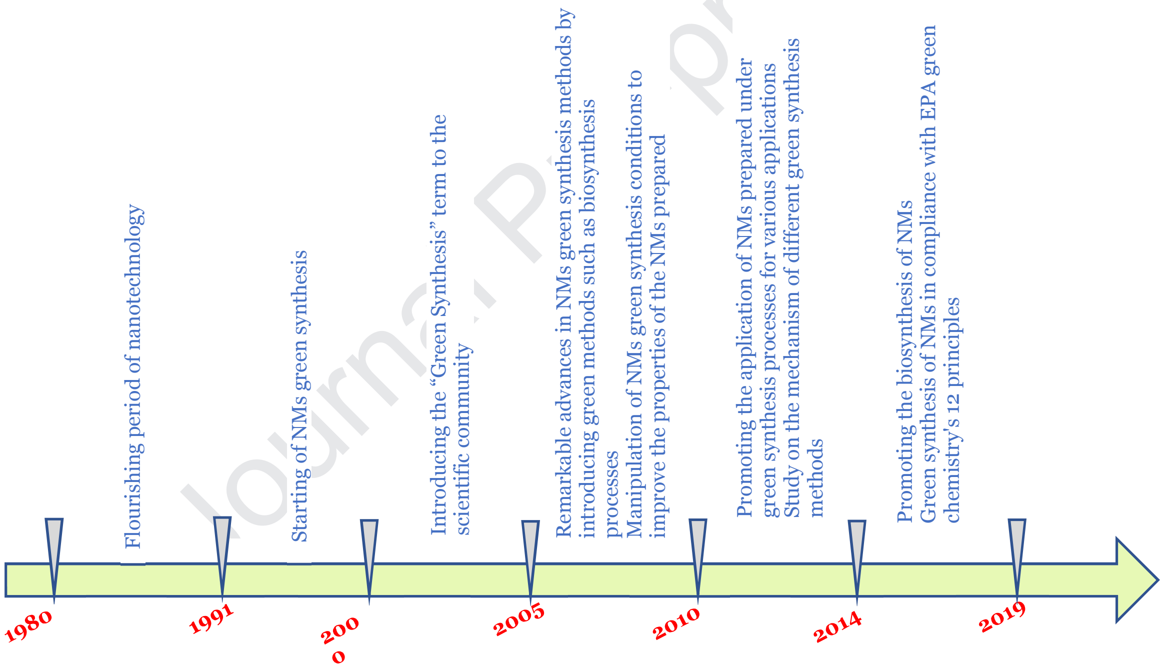


**Fig. 12.**
Milestones in the progress on green synthesis of NMs during the period 1991-2019.

The first attempts on the green synthesis of NMs go back to the period 1991-2000 when ultrasonic irradiation (UI) was introduced as a green production technique for the synthesis of NMs, being the synthesis of $Si_3N_4$/SiC ceramic nanocomposite by Gonsalves, K.E. et al (1991) one of the first studies reporting on material synthesis assisted by UI (Gonsalves et al., 1991). In this decade, 54 documents were published on green synthesis of NMs (Fig. 2), most of them relying on UI. However, the number of publications in this period is relatively low, indicating that the scientific knowledge at that time was still at in its infancy. At that time, UI was even not considered as a green production method but rather a tool to prepare NMs with desired properties. Microwave-assisted methods underwent a similar approach in this period of time. For instance, Yu et al (1998) fabricated polymer-stabilized platinum colloids (with an average diameter of 2-4 nm) using a household microwave oven as a heating device. They found that microwaves could heat a material evenly in a glass or plastic reactor. In fact, microwave irradiation favors homogeneous nucleation while allowing the reaction medium to heat up, helping to reduce the crystallization time compared to other conventional heating methods. In this initial period, USA, with 22 published documents, was the country leading the publications in the field. One important reason accounting for this leadership was the development of the idea of "green chemistry" in response to the Pollution Prevention Act (1990), which fostered the Environmental Protection Agency (EPA) to design actions aimed at the reduction of environmental pollution. In the early 1990s, EPA and the National Science Foundation (NSF) in the USA started to fund research projects on the green chemistry of nanomaterials synthesis ("History of Green Chemistry," 2017). In addition, green chemistry has been initially discussed in Chinese journals in 1994. In 1996 "Green Chemistry and Technology Symposium" was organized by the Academy and Science Department of China (Cui et al., 2011). Among the publications in the same period, the work published by Kumar et al (2000) stands out, due to the greater number of citations (more than 460). These authors synthesized metallic oxide nanomaterials from metal acetates precursors using UI and discussed the mechanisms involved in their synthesis, which probably explains such a high citation number. Among the more active authors, Gedanken. A. et al from Israel with 12 and 28 publications during the periods 1991-2000 and 2001-2005, respectively, owned the highest number of publications in this field (Prozorov et al., 1997). Their work was mainly focused on the synthesis of nanomaterials such as iron nitride using UI. In 2000, Gedanken. A. et al authored 7 papers on UI synthesis of nanomaterials which were highly cited (> 1300 citations), where they discussed the different mechanisms involved in the UI process, supported by the effects of different experimental conditions on the properties of the synthesized NMs (e.g., morphology, size, surface area, band gap, magnetic and optical). They also combined the UI based synthesis with other synthesis methods such as those based on heat treatment (Gedanken et al., 1999; Kumar et al., 2000; Li et al., 2003; Pol et al., 2002; Qiu et al., 2005, 2003; Vijayakumar et al., 2000). Among the keywords with higher frequency over the period 1991-2000 period, the following are here highlighted: nanoparticle, sonochemical synthesis, size, and powder. These keywords evidence that particle size reduction to the nanometer scale was, at the time, an effective concern among the scientific community (Mintova et al., 1998; Vijayakumar et al., 2000). Sonochemical based methods offer several advantages, such as cost-effective, the possibility of being combined with other synthesis methods, being eco-friendly and having the ability to grant the NMs particles with the desired properties (i.e., size, crystallinity, surface area, morphology, etc.) (Kamali et al., 2019b). In conclusion, during the last decade of the twentieth century, prompted by the nascent idea of "green chemistry" or by UI- and Microwave-assisted methodologies, research on the synthesis and control of NMs properties was significantly advanced. However, it is worth mentioning that the usefulness of these achievements for real applications of NMs has not been addressed so far.

The period of 2001-2005 allowed to consolidate the concept of “green synthesis”. This term was used for the first time by Peng. X and Peng. Z. A. (2001) in their one-pot synthesis of cadmium chalcogenides using less toxic and cheaper reagents (i.e. cadmium oxide instead of dimethylcadmium as cadmium source) at room temperature. Later on, in 2003 this term was utilized by other researchers when reporting the synthesis of nanomaterials (Fu and Raveendran, 2003; Mekis et al., 2003). This is the case of Raveendran. P. et al (2003) who reported the green synthesis of nanometals (like silver), with environmental friendly and renewable compounds such as water (solvent), sugar (β-D-glucose, reducing agent), and starch (protecting agent), reaching over 1413 citations to date. EPA introduced “Green chemistry” as being the design of chemical products and processes that reduce or eliminate the use or generation of hazardous substances. Green chemistry can be applied throughout the life cycle of a product, including its design, manufacture, use, and final disposal. Green chemistry is also known as “sustainable chemistry”(EPA, n.d.). The release of around 300 scientific publications highlights the progress of the green synthesis of NMs in that period of time (2001 -2005). Gedanken. A. from Israel, with 28 documents, had the highest number of publications in this field among the authors and the contributing countries. This achievement encouraged the increase of Chinese research activities in the field of green chemistry at the service of NMs synthesis, which pushed the Chinese government to fund a number of projects. The resulting research momentum also led to the establishment of various Chinese scientific institutions and associated laboratories in this field (Cui et al., 2011). In line with this surge of research activities during the first half-decade of the 21$^{st}$ century, it can be observed that keywords like nanoparticle, growth, nanowire, irradiation, nanotube, powder, nanocrystal, size, film, and nanostructure reached their highest citation number during this period (Fig. 7). There are other keywords like shape and morphology, nanomaterials properties and methods of synthesis that have also deserved some attention during that period (Cao et al., 2005; Geng et al., 2005; Sun and Luo, 2005; Yin et al., 2002). More recently, Matus. K.A.J et al, (2012) investigated the existing policies and improvement opportunities for green chemistry and engineering in China, in order to identify the challenges that China has to overcome for sustainable development. The activities mentioned in this section are probably the main reasons accounting for China’s leading position among countries with the highest numbers of publications in this field (Fig. 4), mainly articles (Fig. 3). Briefly, the research carried out during the first years of the current century can be viewed as a prelude to the sustainable development of NMs, essentially focused on the transition to less toxic and environmentally friendly precursors, without taking into account other sustainability pillars such as the economic criterion.

The period 2006 to 2010 can be considered a birth period for new synthesis processes of NMs such as biosynthesis methods. In addition, the manipulation of the synthesis conditions aiming to improve the properties of NMs was also particularly pursued in this period. Hence more than 1000 documents were published, and the rate of publication increased notably in this field. China maintained its leading position, as a country, while among the authors Varma et al. from the USA owned the highest number of publications in this period, thanks to their investment on the control of nanomaterials morphology, especially during 2006 and 2007 (Mallikarjuna and Varma, 2007; Nadagouda et al., 2007; Nadagouda and Varma, 2006). They explored the preparation of NMs using plants as precursors (Braydich-Stolle et al., 2010; Nadagouda et al., 2009). For instance, they synthesized Ag and Pd nanoparticles (20-60 nm) from powders of dehydrated coffee and tea leafs at room temperature without the need of surfactants or capping agents often reported (Nadagouda and Varma, 2008). The authors found that polyphenol existing in the used natural precursors can act as a suitable reducing and capping agent for the sake of controlling the size of synthesized NMs in the range of 20-60 nm (Nadagouda and Varma, 2008). Among the documents published in this period, the paper by Sharma, Yngard, and Lin (2009) received the largest number of citations, over 1996. In this study, the authors reviewed the green synthesis process of Ag nanoparticles and the mechanisms underlying their antibacterial activity. Another highly cited paper in the same period belongs to Guo et al. (2009) who introduced a green method for the synthesis of graphene nanosheets via electrochemical reduction of exfoliated graphite oxide. This article has been cited more than 1614 times until 2018. The analysis of keywords in this period (Figures 6 and 7) reveals the words nanoparticle, growth, nanocrystal, nanostructure, nanowire and nanorod as the most frequently cited. This results indicates that, in this period, scientists were focused on the manipulation of the synthesis conditions in order to tailor the properties of the synthesized nanomaterials (Bilecka and Niederberger, 2010; Hassan et al., 2009; Jin et al., 2010; Muraliganth et al., 2010; Murugan et al., 2009; Tian et al., 2006; Tompsett et al., 2006; Yan et al., 2010; Zhu et al., 2009). Despite the significant progresses achieved in this period regarding the control and improvement of NMs properties, no comprehensive study encompassing all green synthesis principles was published.

The following years, from 2011 to 2014, brought rapid developments to the production and application of NMs within the framework of green synthesis principles. Some oxides like ZnO or TiO2 are here cited as examples of antibacterial, wound healing and antioxidant (Agarwal et al., 2017) or photocatalyst and antiparasitic applications (Nadeem et al., 2018). A particular focus was also paid to the understanding of the mechanisms involved in green synthesis processes. More than 2200 documents were published during this period, thus demonstrating the great interest raised by this subject among the scientific community. China kept its leadership in the number of publications and, among the authors, M. Salavati-Niasari from Iran, authored the largest number of publications in this field (Figures 5, 8 and 9). UI and microwave approaches were deeply explored by several authors, especially by Salavati-Niasari, regarding the preparation of various types of nanoparticles with specific properties to be used for different purposes such as environmental clean-up and energy applications (Hosseinpour-Mashkani et al., 2012; Soofivand et al., 2013) (see Figures 9 and 10). Among the published documents, the review paper from Iravani, (2011) was the most cited work (citations >1000) (Fig. 11). This paper reviewed the synthesis methodologies of metallic nanoparticles using plants as precursors (such as Aloe Vera, Black tea, Eucalyptus, Parthenium hysterophorus, etc.). Another highly cited work in this period, with more than 600 citations belongs to Lu et al. (2012). This paper reports the synthesis by a hydrothermal method of fluorescent carbon nanoparticles to be applied in sensing systems targeted to detect $Hg^{2+}$ in aqueous media. The following keywords can be highlighted in this period: nanoparticle, nanocrystal, nanostructure, silver nanoparticle, gold nanoparticle, nanocomposite, green synthesis, growth, particle, optical property, nanorod, film, reduction, microwave, composite, size, nanowire, catalyst, biosynthesis, metal nanoparticle, oxide, fabrication, and thin film. This finding demonstrates clearly that the number of scientific terms used in publications on green synthesis of NMs kept increasing. However, key aspects regarding economic considerations have not been discussed in those papers, hence indicating that one of the principles of green chemistry (Table 9) has not yet been appropriately addressed, even in this more recent period (Badawy, 2014; Coman et al., 2014; Huang et al., 2014; Kim et al., 2013; Mtimet et al., 2012; Yan et al., 2013). Concluding, despite being a significant milestone, the application of NMs engineered according to the green synthesis principles remained confined to the lab-scale in many important fields, not spilling over to real applications. In fact, although the rate of knowledge transfer from the lab to real applications underwent a significant increase in some areas like biomedical engineering, no evidence of commercialization has been observed in other important applications such as the treatment of polluted waters and wastewaters.

The last stage of scientific progress in this field covers the years 2015 to 2019, where biosynthesis of nanomaterials has drawn increasing attention from the scientific community. It can be inferred that researchers were now more attentive to the 12 main principles of green chemistry defined by EPA (Table 9). In this period about 5114 documents were published and the number of publications still kept increasing continuously. Despite the previous stages, India surpassed China and owned the highest number of publications. This is not surprising as India benefits from a strong material and chemistry industrial basis. India is currently considered a leading country in many industries such as pesticides, pharmaceutical and petrochemical with a number of environmental adverse effects such as the production of hazardous waste and wastewater, and the emission of harmful gases. These emissions are at the origin of different environmental impacts, such as severe environmental resources pollution (groundwaters, for instance) and health problems such as cancer, reproductive damage, breathing problems, allergy, liver and kidney problems (Jain et al., 2016; Sharma, 2018). In this regard, green chemistry principles can guide chemical industries in developing countries like India to achieve sustainable development (Kidwai, 2001). Indian universities have also tried to promote their activities on the green synthesis of NMs. The University of Delhi, for instance, has recently launched the Green Chemistry Institute with a focus on activities supporting green chemistry innovation (Yadav, 2006). In 2014 the first national chemical policy was approved in India (Petrochemicals, 2014), which has been raising an increasing national sensitivity to the benefits of green synthesis. Among the authors, Masoud Salavati-Niasari remained the top author in terms of the number of published documents with 59 papers published. Regarding highly cited documents, the review paper by Ahmed et al. (2016) (Ahmed et al., 2016) on the synthesis of Ag mediated by plants extract for antibacterial activities received more than 597 citations. In this review paper, biosynthesis and conventional methods for the synthesis of Ag nanoparticles are compared while the advantages of this green methodology in terms of economic, environment-friendly, and efficiency aspects are also addressed. Among the keywords introduced in literature during this period, nanoparticle, silver nanoparticle, green synthesis, and biosynthesis were the most cited ones. Although green synthesis terminology has reached a certain maturity in this period (2015-2019), it is here considered that a step forward can be taken by introducing sustainability considerations towards the "sustainable synthesis" of NMs. Sustainability is a concept that integrates social and economic aspects in addition to the environmental conditions required by green synthesis. Sustainability aims to conserve natural resources, while promoting materials production techniques assisted by economic considerations and raising a high degree of social acceptability. Some literature reports have already introduced economic considerations criteria in the selection of a synthesis process, (Kumar et al., 2015; Sayed and Polshettiwar, 2015; Song et al., 2012; Wang et al., 2015). However, highly important aspects such as investment and equipment maintenance costs have not yet been included (Cinelli et al., 2016; Yuan et al., 2018). Furthermore, no report is currently found on the social impacts of methods capable of large scale synthesis of NMs, such as social acceptance and the ability to create new job opportunities (Cinelli et al., 2016; Siegrist et al., 2007). As a final comment, we may refer that some evidences of real applications of NMs were recorded in this last period. For instance, the first commercial nano-reactors were introduced for the treatment of polluted waters and wastewaters (Kamali et al., 2019c). However, despite the wide range of applications for nanotechnology-based products, the number of NMs transferred from lab to real applications is still limited. The main reasons that explain this gap include the lack of information on economic issues as well as the social acceptability of the NMs produced and the lack of knowledge about the potential of methods for scaling-up. To overcome these still existing barriers, future studies on the synthesis of NMs are highly encouraged to address the sustainability criteria defined very recently in the scientific literature including their technical, environmental, economic and social pillars (Kamali et al., 2019a). It is envisaged that this strategy will help the parties involved to select the most sustainable methods for NMs production in order to further boost the commercialization of the methodologies already developed while attracting future investments in the search for novel methodologies, more compliant with principles of sustainability.

**Table 9.** The 12 main principles of green chemistry, adopted from EPA (EPA, n.d.).

| Row | Green chemistry principles |
|---|---|
| 1 | Prevent waste |
| 2 | Maximize atom economy |
| 3 | Design less hazardous chemical syntheses |
| 4 | Design safer chemicals and products |
| 5 | Use safer solvents and reaction conditions |
| 6 | Increase energy efficiency |
| 7 | Use renewable feedstocks |
| 8 | Avoid chemical derivatives |
| 9 | Use catalysts, not stoichiometric reagents |
| 10 | Design chemicals and products to degrade after use |
| 11 | Analyze in real time to prevent pollution |
| 12 | Minimize the potential for accidents |

## 5. Final remarks and future research directions

So far, many efforts have been directed towards the synthesis of various types of NMs for various applications, from biomedical applications to water and wastewater treatment. The present scientometric study has demonstrated that the NMs green synthesis reported in the literature cannot satisfy the objectives of sustainable development that require that a certain product be of high quality (in this case efficient), be environmentally friendly, economical and socially acceptable. The main emphasis was placed on the technical conditions necessary for the green synthesis of NMs and the intended application for developed NMs, being very scarce the reports encompassing the economic issues underlying the NMs production process. Furthermore, the environmental impacts of the synthesis methodologies developed and their social acceptability have not been sufficiently addressed in the literature. These limitations can hinder the widespread utilization of the developed NMs in various applications (especially for state-of-the-art) where there is an urgent need for efficient, cheap, safe, and socially acceptable materials and technologies. Hence, the following research directions can be suggested here for future studies:

a) To identify the main technical, environmental, economic, and social criteria and their relevant sub-criteria that may affect the sustainability of NMs synthesis processes.
b) To adopt efficient synthesis methodologies in order to prioritize the identified sustainability criteria and sub-criteria that ensure the involvement of the scientific community in this process. Multi-criteria decision making processes such as fuzzy-Delphi methodology can be suggested as a useful tool for this purpose (Kamali et al., 2019c).
c) To apply cost-effective methodologies such as Taguchi experimental design which can considerably reduce the investments required for the optimization of the properties of the nanomaterials for the desired applications, especially for the most efficient methodologies identified in (b).
d) To attract investments for pilot and full-scale applications of NMs taking into account all the sustainability criteria discussed in this study, in order to facilitate their commercialization.

In this regard, future studies addressing sustainability criteria in the synthesis of NMs, in order to facilitate the transfer of innovative methods from lab to full-scale application are highly welcome. It is here envisaged that such studies may also trigger a new surge of publications. Furthermore, it is anticipated that merging green chemistry and economic principles may also provide a basis for addressing the synthesis of nanomaterials in line with frugal innovation, with a significant and positive impact on developing countries.

## 6. Conclusion and perspective

A map of the scientific progress in green synthesis of NMs has been presented here. Information on the number of documents published annually, types of document, countries and authors' contributions, trends in the evolution of scientific keywords, the most cited authors and journals, scientific categories appearing in this field over time, and the most cited articles were collected and utilized to critically evaluate the 8761 documents extracted from WoS database using an appropriate set of keywords. The analysis of the results evidences a relative maturity of the scientific progress in the field of green synthesis of NMs. So far, the term "green synthesis" is intended to exclude synthesis processes with adverse effects on the environment, such as environmental pollution by produced wastes. However, there are numerous technical, environmental, economic and social considerations that need to be taken into consideration, in order to meet the current and future needs for efficient, cost-effective and environmentally-friendly methods for the production of NMs. To address this gap, it is suggested in this study to refocus on "sustainable synthesis" by including all the technical, social and economic considerations, in addition to environmental drawbacks.

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# Supplementary Information

**Keywords used for this study:**

## Green Synthesis of Nanomaterials - A Scientometric Assessment

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**Highlights**

- A scientometric assessment on nanomaterials green synthesis methods is provided.
- Science evolution milestones in this field have been identified.
- Nanomaterials green synthesis principles are critically discussed.
- Integration of sustainability criteria may fill an existing gap in this field.

**Declaration of interests**

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: