Current Chemistry Letters 8 (2019) 97–116

Contents lists available at GrowingScience

Current Chemistry Letters

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A survey on application of MOFs in chemistry

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C H R O N I C L E

Article history:
Received January, 2019
Received in revised form
February 25, 2019
Accepted March 15, 2019
Available online
March 15, 2019

Keywords:
Chemistry
Scientometrics
Bibliography
Metal–organic frameworks (MOFs)

A B S T R A C T

Metal–organic frameworks (MOFs) are combinations of metal ions or clusters accommodated to organic ligands to shape different dimensional structures. MOFs are considered as a subclass of coordination polymers, with the possible characteristics that they are normally porous. The metals are considered to offer flexible, co-ordination environment under virtually various topologies. Besides, because of the usual liability of metal complexes, the shape of the coordination bonds between the metal ions and the organic linkers can be reversible and this helps the rearrangement of metal ions and organic linkers through the process of polymerization to give highly ordered framework structures. The study has indicated that MOFs has maintained extensive applications in Biological imaging and sensing, Drug delivery systems, Methane storage, Semiconductors, Bio-mimetic mineralization, Carbon capture, Desalination/ion separation, Water vapor capture and Ferroelectrics and Multiferroics. This paper presents a scientometrics study on 1273 papers published articles, books, patents, *etc.* indexed in Web of Science database over the period 2001-2019. The study presents the most popular keywords used in the literature, determines the network of scientific scholars and discusses the clusters of keywords used for different surveys. The results indicate that metal-organic frameworks and zeoitic imidazolate frameworks are two keywords considered as motor keywords in MOFs studies.

1. Introduction

Metal–organic frameworks (MOFs) have attracted much attention in different aspects of chemistry, since their first report in 1995 by Yaghi *et al.*[1]. They are combinations of metal ions or clusters accommodated to organic ligands to shape different dimensional structures. MOFs are considered as a subclass of coordination polymers, with the possible characteristics that they are normally porous[1-10]. The metals are considered to offer flexible, co-ordination environment under virtually various topologies[11-20]. Besides, because of the usual liability of metal complexes, the shape of coordination bonds between the metal ions and the organic linkers can be reversible and this helps the rearrangement of metal ions and organic linkers through the process of formation to give a highly ordered framework structure. MOFs are normally formed under solvothermal or hydrothermal conditions in pure *N*,*N*-diethylformamide or *N*,*N*-dimethylformamide implemented as solvents. The organic linker molecules react with metal salts and generates 3D metal-organic networks[21-50]. **Fig. 1** demonstrates a typical MOFs structure.

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doi: 10.5267/j.ccl.2019.003.001

Fig. 1. The structure of a typical MOFs structure

Metal ions include void orbital's which describes their coordination number with size and shape of pores by prescribing how many ligands have to bind to the metal and plays as the secondary building units to build open crystalline frameworks with enduring porosity[51-70]. The organic units are di or tri organic amines, carboxylates, amylates, *etc.* and once connected to metal-containing units, result architecturally robust crystalline MOF structures with some special porosity, generally bigger than 50% of the MOF crystal volume. They have been determined as a class of "porous polymeric materials" including metal ions combined with organic bridging ligands, and they have become a new development on the interface between materials science and molecular coordination chemistry[71-120]. MOFs have been extensively used in different industries such as drug industries, medical devises, gas storage, sensors, *etc.* MOFs applications in the area of biomedical science have also been extensively explored. The preliminary investigations of MOFs in this field demonstrate a promising role for biomedical tools. Stability and the toxicology of the material are considered as the main challenges which ought to be investigated when MOFs are applied in this area. Since a significant number of MOFs have been synthesized to date, it is hard to make a conclusion on the stability of the MOFs. For example, MIL (Fe-MOFs) family has been considered as a unique one for the purpose of storage of biologically essential molecules. The imaging and drug components have to be directly included into the MOFs either as metal-connecting points or as bridging ligands when we perform the MOF synthesis[120-199]. Medical imaging such as MRI depends entirely on big doses of contrast agents to substantiate between normal and diseased tissues. MOFs are biodegradable and their high porosity makes them suitable for targeted delivery of entrapped agents. MOFs have also the capability to resolve different challenges of selectivity that plague other sensor instances and form the basis of strongly-sensitive and compact sensing devices. MOFs maintain some special characteristics, for instance, mesoporous MOFs MIL-100 and MIL-101 adsorb significant amounts of CO_2 and CH_4 [8-11].

Furukawa et al.[2] is believed as one of the best known studies on the applications of MOFs in different industries. They performed a review on the structures devised and explained the design strategies which help groups of materials be synthesized and modified with almost the same framework topology but different in pore type and size of functional families present on the linkers[2]. Ockwig et al.[2,30] analyzed the structures of all 1127 three-periodic extended MOFs existed in the Cambridge Structure Database and determined their underlying topology. Tranchemontagne et al.[4,22,31,126] provided an essential review of transition-metal carboxylate clusters which could serve as secondary building units (SBUs) towards construction and synthesis of MOFs. Rosi et al.[5,100] presented the benefits of the idea of rod secondary building units for the design and synthesis of MOFs. Henninger et al. [43,96] discussed the applications of MOFs as adsorbents for low temperature heating and cooling tools. According to Fromm et al.[44], "Alkali and alkaline earth metal cations are recognized for their ionic chemistry in aqueous medium, and a varying coordination number, based on the size of the binding partners as well as on electrostatic interactions between the ligands and the metal ions. This makes the strategic synthesis of coordination polymer networks with these metal ions a challenge and explains why few systematic results in the generation of metal–organic frameworks (MOFs) are found in the literature". They presented a comprehensive review on some results in the field, bringing together the systematic approaches with results obtained by serendipity, to provide an overview on current and future works which could be accomplished. Papaefstathiou and MacGillivray[44] shed light on the design and synthesis of cavity-containing and porous MOFs with emphasis on techniques, which helps the functionalization of interior void spaces with organic groups. They also discussed a class of MOFs, recognized as inverted IMOFs, which enables organic functionalization using principles of supramolecular chemistry. According to Keskin and Kızılel[26,46], we see a growth on studies associated with MOFs in a numerous applications in chemical engineering, chemistry, and materials science, including gas storage, gas separation, catalysis and also biomedical applications. There has been a substantial progress of implementing MOFs as a platform in biomedical applications because of their high drug loading capacity, biodegradability, and versatile functionality. Keskin and Kızılel [26,46] explained substantial potentials of MOFs for development and implications in biomedical applications by explaining issues including stability, toxicology, and biocompatibility. Wang and Cohen[7,32,77] investigated the modification of MOFs in a postsynthetic scheme, where it is modified with chemical reagents with conservation of the lattice structure. Farha and Hupp[8] showed the rapid separation of desired MOFs from crystalline and amorphous contaminants cogenerated during synthesis according to their various densities. They also described the mild and effective activation of initially solvent-filled pores with supercritical carbon dioxide, resulting usable channels and high internal surface areas.

The study has indicated that MOFs has maintained extensive applications in Biological imaging and sensing, Drug delivery systems, Methane storage, Semiconductors, Bio-mimetic mineralization, Carbon capture, Desalination/ion separation, Water vapor capture and Ferroelectrics and Multiferroics. This paper presents a bibliographical survey on development of MOFs applications in different industries. The study has extracted 1273 records of information indexed in Web of Science and analyzed them using a scientometrics tools named Biblioshiny in R-software package. The study also reviews some the highly cited articles and discuss future trends based on the information collected from the software.

2. The bibliographic study

2.1. The themes in reviewed articles

The search of articles on the Web of Science database has been accomplished with a keyword “MOFs in chemistry” and there were 1273 articles, patents, books, proceeding, etc. associated with the keyword. The purpose of this study was to do search on highly cited references in this area. **Table 1** demonstrates some of the most cited references associated with the application of MOFs applications in chemistry. As we can observe from the results of Table 1, chemistry, design, MOFs, coordination polymers and adsorption are some of the well-recognized keywords used in the literature. **Fig. 2** presents the factorial analysis of the survey and as we can observe there are two groups of words used in this survey among researchers.

Table 1

The most popular keywords used in studies associated with mesoporous materials

Words	Occurrences	Words	Occurrences
chemistry	601	pore-size	39
design	340	clusters	37
metal-organic frameworks	264	nets	37
coordination polymers	251	units	34
adsorption	173	growth	33
complexes	157	frameworks	32
hydrogen storage	145	methane storage	32
MOFs	131	construction	31
separation	108	single-crystal	31
crystal-structures	99	thin-films	31
zeolitic imidazolate frameworks	90	hydrogen	30
Storage	89	catalysts	29
crystal-structure	83	exchange	29
networks	78	MOF	29

Words	Occurrences	Words	Occurrences
catalysis	77	porosity	29
carbon-dioxide	76	solid-state	29
secondary building units	74	temperature	29
building-blocks	67	CO_2	28
magnetic-properties	67	crystal	28
porous coordination polymers	65	molecular-dynamics simulations	28
topology	62	oxidation	28
molecules	61	coordination	27
solids	61	functional-groups	27
sorption	61	porous materials	27
metal-organic framework	60	surface	27
network	60	gas-adsorption	26
ligands	59	heterogeneous catalysts	26
polymers	57	porous solids	26
coordination polymer	56	organic frameworks	25
reticular chemistry	56	porous coordination polymer	25
stability	56	postsynthetic modification	25
acid	53	adsorption properties	24
ligand	52	architectures	23
functionalization	50	performance	23
surface-area	48	capture	22
water	48	complex	22
carbon-dioxide capture	47	room-temperature	22
hydrothermal synthesis	47	asymmetric catalysis	20
sorption properties	47	functionality	20
crystals	46	self-assembled monolayers	20
nanoparticles	46	efficient	19
sites	43	hydrogen adsorption	19
building units	40	MOF-5	19
drug-delivery	40		

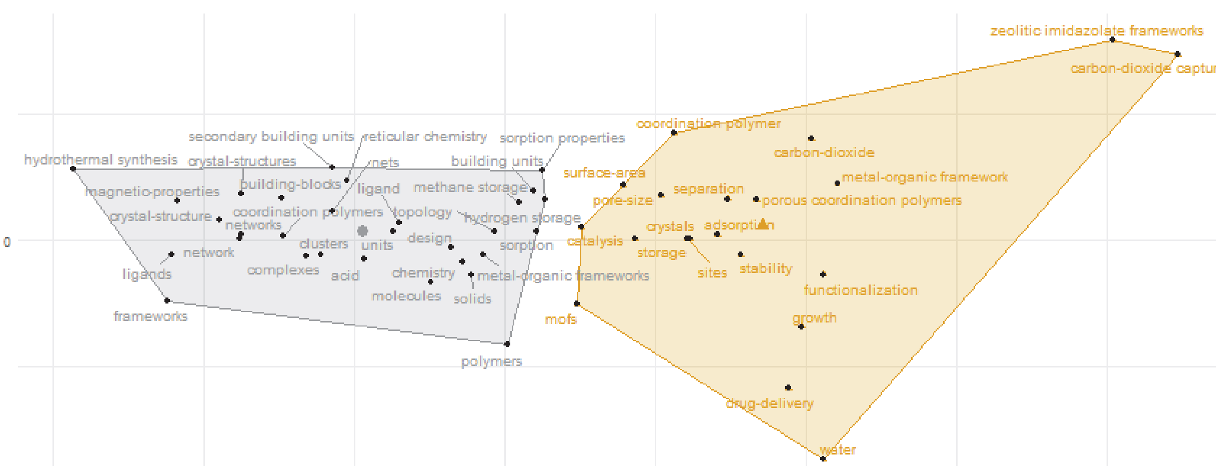

Fig. 2. Factorial analysis

2.2. Country Scientific Production

Fig. 3 presents the distribution of scientific production by various countries and as we can observe, the largest scientific productions are associated with United States and China. In other words, 1143 works which represent nearly 90% of the published scientific works have been accomplished in United States and China.

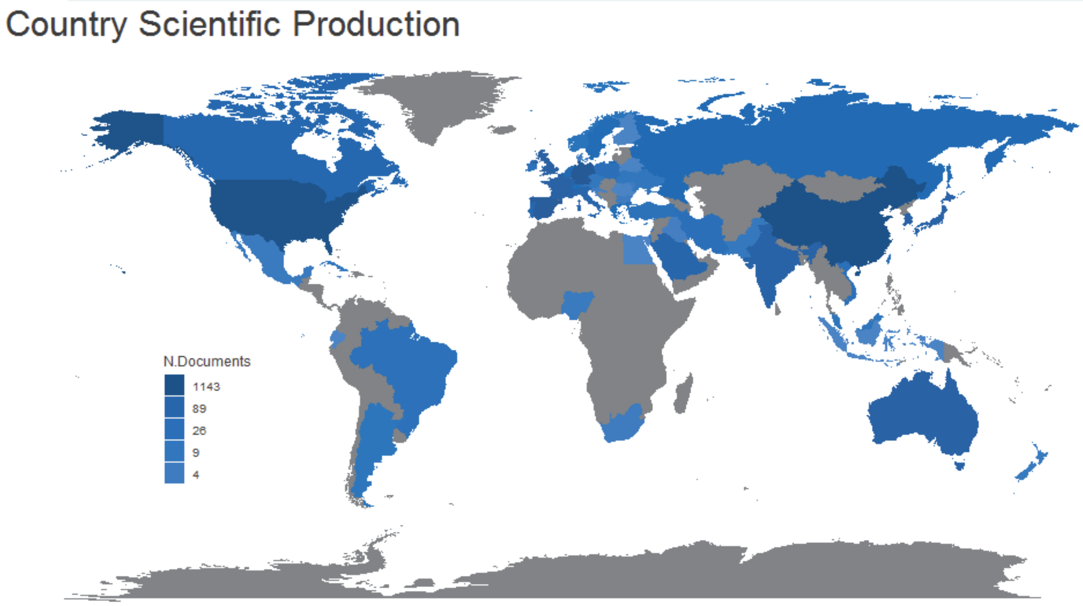

Fig. 3. Country Scientific Production

2.3 Corresponding author's country

Our survey demonstrates that researchers from the United Stated and China have maintained the most contribution in this field followed by the researchers from Germany, India and France. **Fig. 4** shows the details of our survey. Moreover, we see a good collaboration between most countries with other countries.

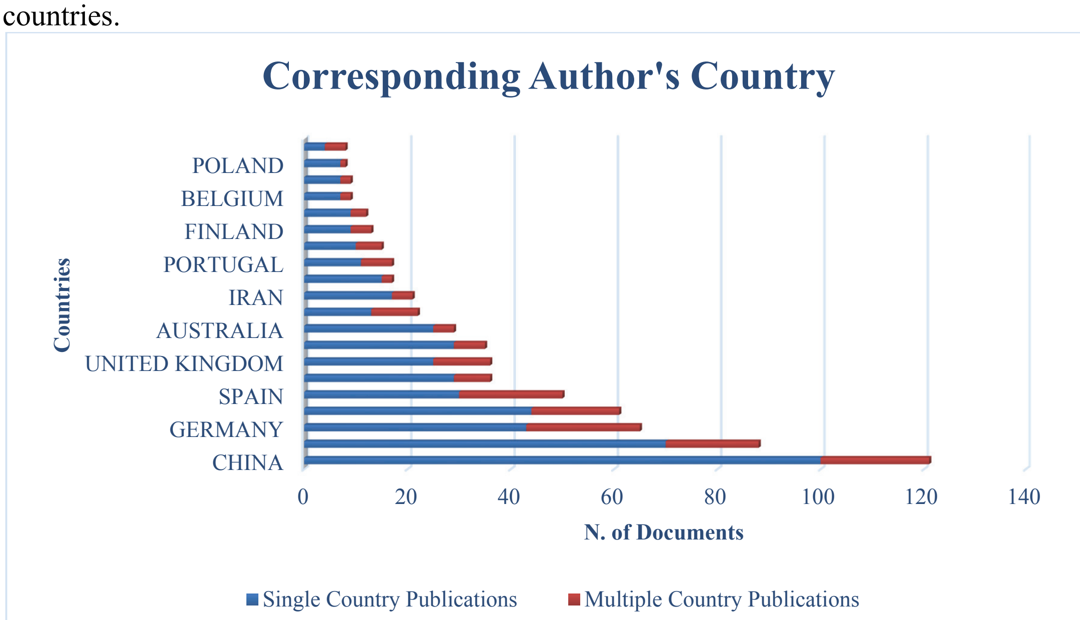

Fig. 4. Corresponding author's country

2.4. The frequency distribution of sources

In this research, most articles from the sources shown in **Fig. 5** are CrystEngComm with 106 articles followed by J. Am. Chem. Soc. with 105 articles.

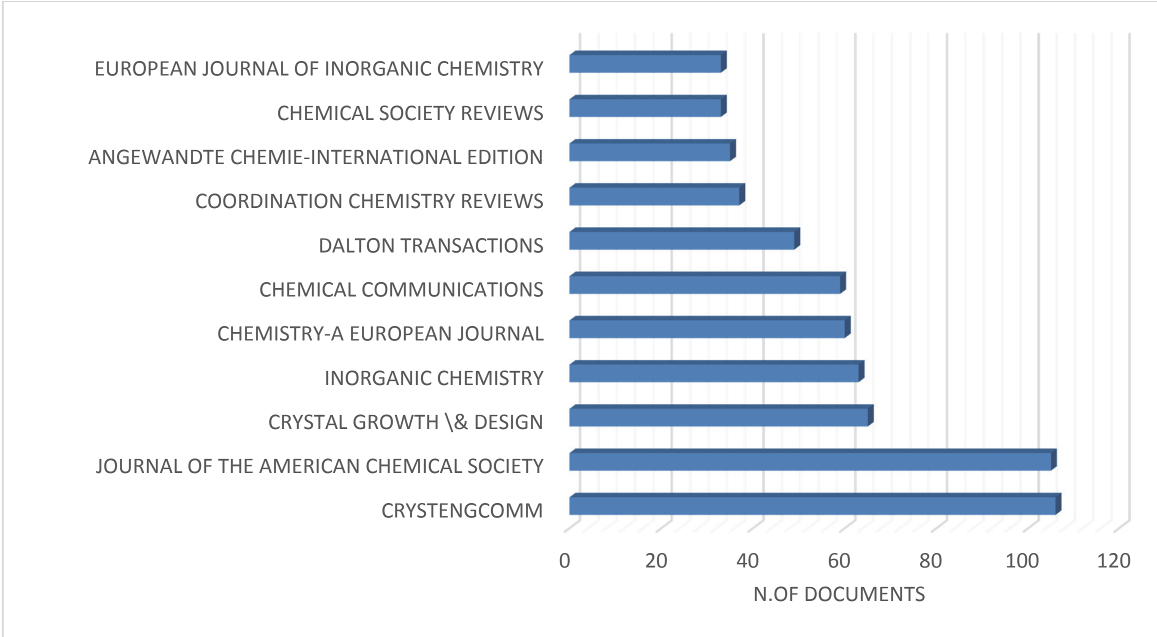

Fig. 5. Most Relevant Source

2.5. Collaboration network

Fig. 6 shows the Author's Collaboration Network and we can observe that different groups of four or five people have executed extensive works in the area of MOFs.

Fig. 6. Author's Collaboration Network

2.5. Cluster classification

As we can observe from the results of **Fig. 7**, there are two clusters associated with application of MOFs in chemistry. The network in **Fig. 7** shows how two groups of people have performed various works over time.

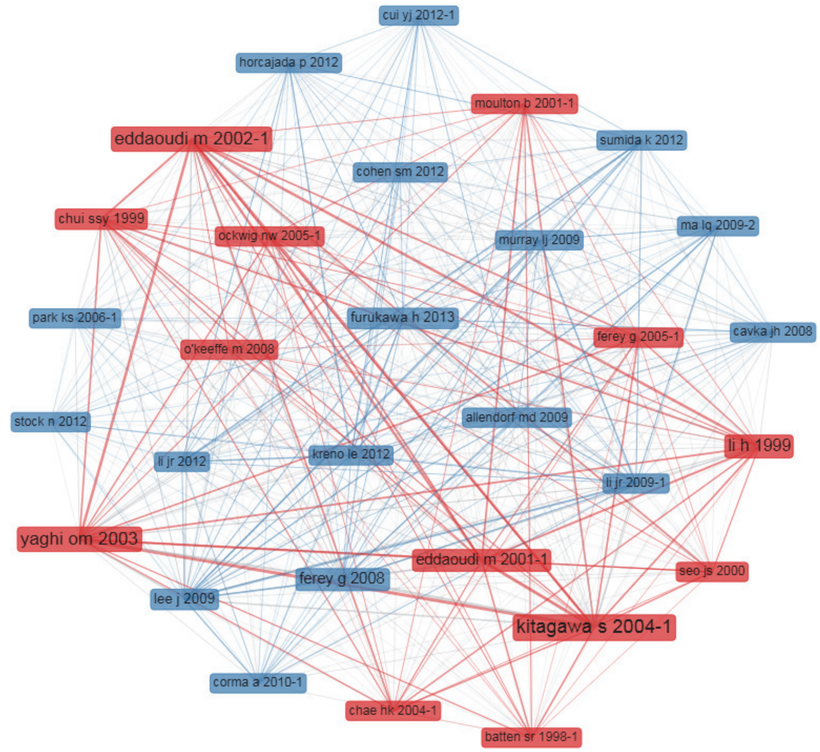

Fig. 7. Demographic of the clusters group of authors

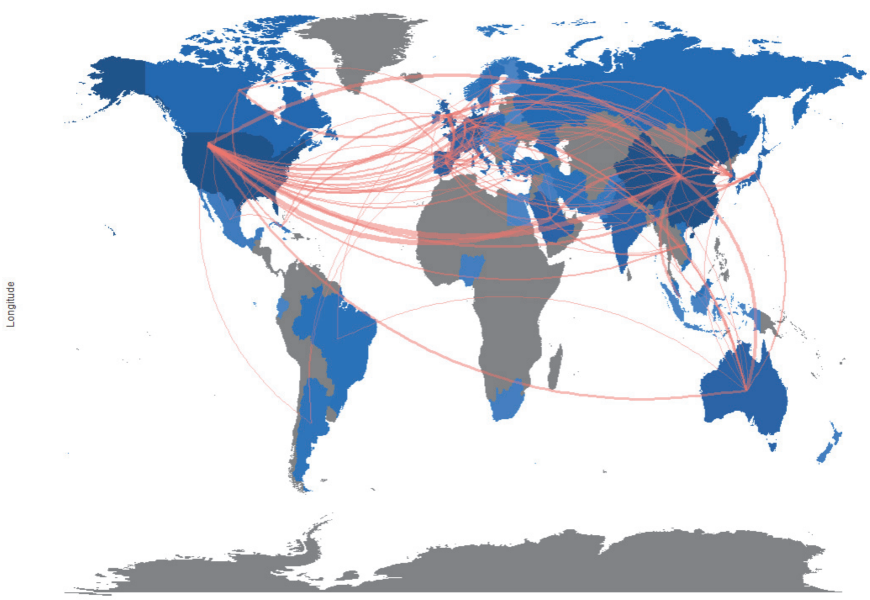

Fig. 8. Country Collaboration map

In terms of the average citation, papers published by researchers in Canada, Australia, and France have received the highest citations. **Fig. 8** shows the results of the collaborations among various countries.

As we can observe from the results of **Fig. 8**, there were some strong collaborations between the researchers in the United States from one side and other countries such as Australia and China. **Fig. 9** demonstrates how many articles have been written by the authors with the highest number of articles during the time, and how many citations each one received. The size of each circle shows the number of articles and the amount of boldness of the circles represents the number of citations in that year. Also, **Fig. 10** shows that metal-organic framework has become the most important keyword used in the literature.

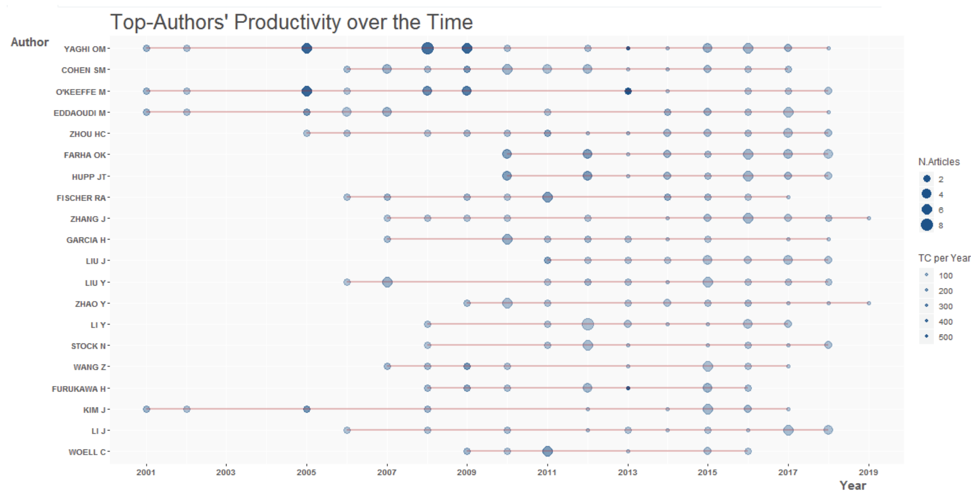

Fig. 9. Top-Authors' productivity over the time

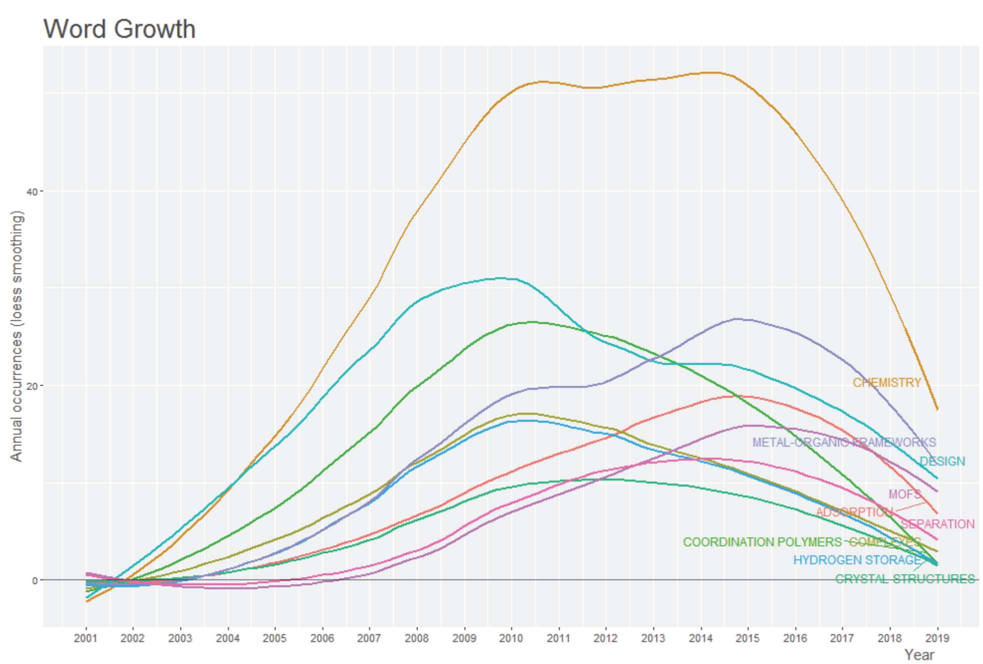

Fig. 10. The trend on Word growth

2.6. Thematic map

Co-word analysis draws clusters of keywords considered as themes. In the strategic diagram presented in **Fig. 11** the vertical axis measures the density – i.e., the strength of the internal links within a cluster represented by a theme –, and the horizontal vertical axis the centrality – i.e. the strength of the links between the theme and other themes in the map.

Thematic map is a very intuitive plot and we can analyze themes according to the quadrant in which they are placed[201-204]:

(Q1) upper-right quadrant: motor-themes;

(Q2) lower-right quadrant: basic themes;

(Q3) lower-left quadrant: emerging or disappearing themes;

(Q4) upper-left quadrant: very specialized/ niche themes.

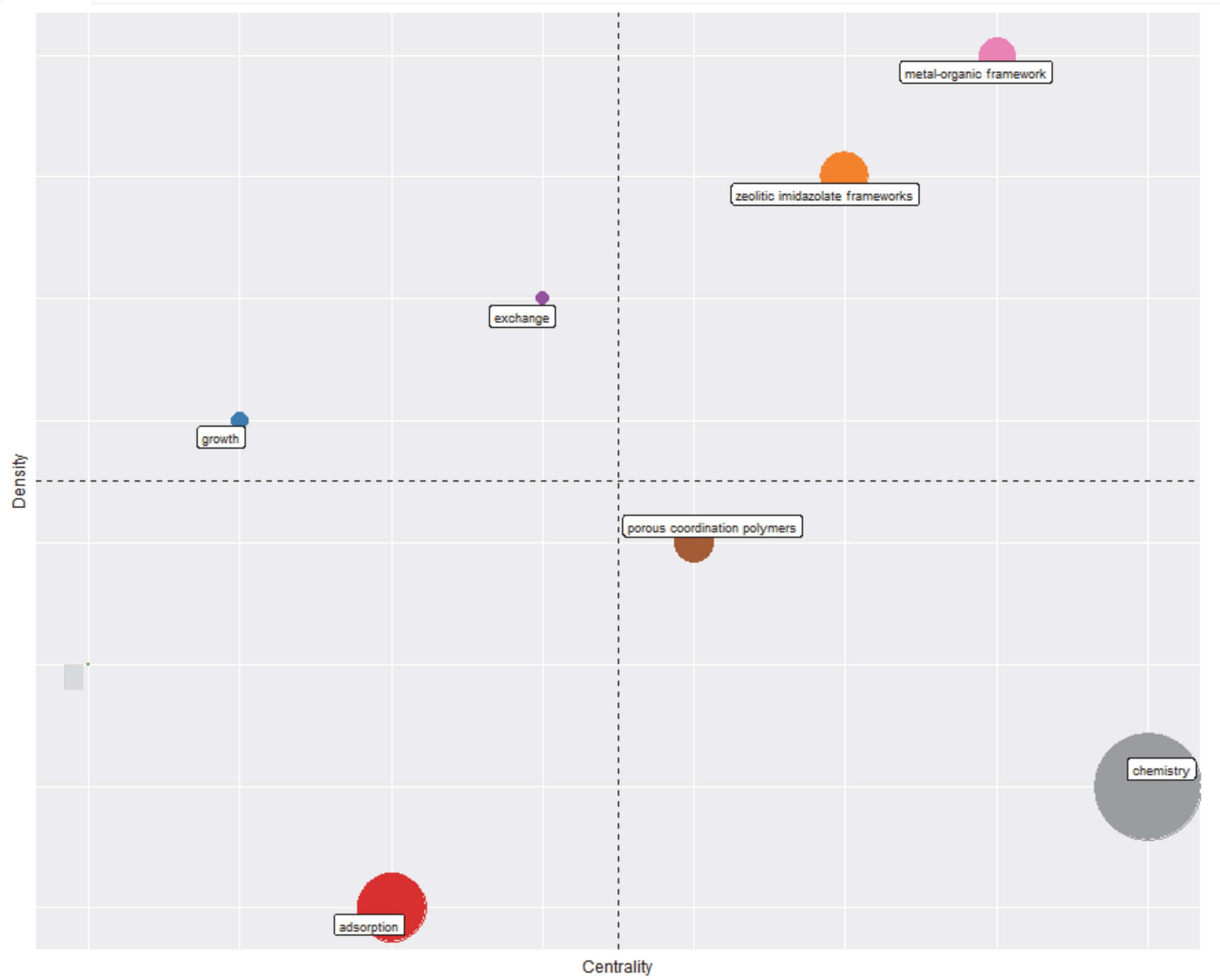

Fig. 11. Thematic Map

Hence, the themes with the highest internal coherence and closest relationship to other themes appear in the first quadrant (the upper right part of the graph) which includes metal-organic framework and zeoitic imidazolate frameworks and these two keywords are considered as motor keywords in MOFs related studies. In the second quarter, porous coordination polymers and chemistry play the basic role for scientific development. Themes in this quadrant are important for a research field but are not developed and they are considered as emerging areas of research.

2.7. Intellectual Structure, Historiographic

The historiographic map is a graph proposed by E. Garfield to represent a chronological network map of the most relevant direct citations resulting from a bibliographic collection. The citation network technique does provide the scholar with a new modus operandi which may significantly affect future historiography. The results of citation cooperation is given in Fig. 12.

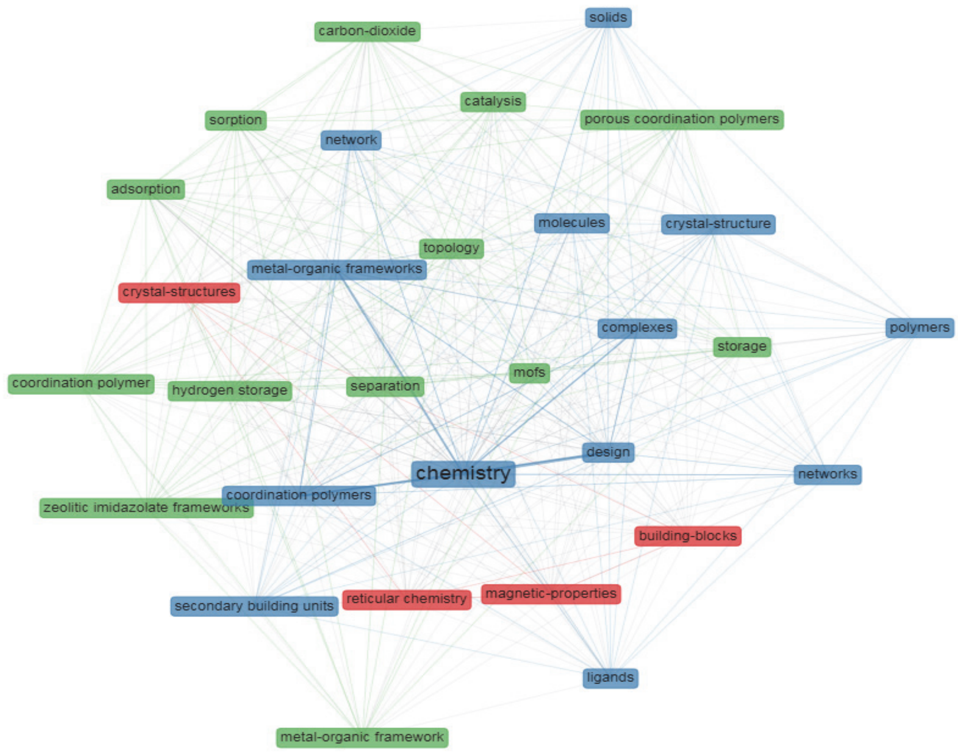

Fig. 12. Historical direct citation network

3. Conclusion

This study has attempted to provide a preliminary review of the scientific studies between 1980 to the second month of 2019 on MOFs in chemistry. The study has indicated that MOFs has maintained extensive applications in Biological imaging and sensing, Drug delivery systems, Methane storage, Semiconductors, Bio-mimetic mineralization, Carbon capture, Desalination/ion separation, Water vapor capture and Ferroelectrics and Multiferroics. The study shed light on some of the most popular keywords implemented in the literature, determined the network of scientific scholars and discussed the clusters of keywords used for various surveys. The results have indicated that metal-organic framework and zeoitic imidazolate frameworks were two keywords considered as motor keywords in MOFs studies.

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