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Research Trends in Ionic Liquids as Green and Low-Cost Solvents: Bibliometric Analysis from 2013 to 2023

Andika Purnama Shidiq^a, Risti Ragadhita^a, Meli Fiandini^a, Asep Bayu Dani Nandiyanto ^{a*}

^a Departemen Pendidikan Kimia, Universitas Pendidikan Indonesia, Bandung, Indonesia * Corresponding author: nandiyanto@upi.edu

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Abstract

Challenged by global awareness of environmental problems and demands of the industrial revolution, the Ionic Liquids (ILs) approach as a cheap and environmentally friendly solvent has been explored in various chemical reactions in many industrial fields. This article aims to perform bibliometric analysis of ILs by combining mapping analysis using Publish or Pheris and VOSviewer software and to pinpoint trends in scientific research. For this purpose, an exploratory study with a quantitative approach was developed based on secondary data from journals in google scholar, between 2013 and 2023. The keywords used in the data search are Ionic Liquid, low cost, and green solvent. The bibliometric indicator refers to 1,000 different relevant articles from 52 publishers with the largest number of articles produced by Elsevier, totaling 423 articles. The results showed that the peak of production occurred in 2014 and then decreased every year. The main reason for this increase is the need for cheap and environmentally friendly solvents to replace expensive and environmentally damaging conventional solvents, while the decline is due to the shift of interest into the field of applied chemistry. From the results of the Bibliometric analysis, there are 9 clusters with 126 related terms including low-cost, green solvent, application, study, and extraction. The most popular terms in 2022-2023 are related to fuel terms. This study shows the importance of bibliometric analysis in providing analytical data about what phenomena occur. This research is expected to help and become a reference for researchers in conducting and determining research themes to be taken.

Keywords: Bibliometric, VOSViewer, ILs, Green Chemistry, Low-Cost.

1. Introduction

Since its inception, Ionic Liquids (ILs), have repeatedly been regarded as green solvents due to their low volatility and non-flammability (Bystrzanowska et al., 2019). The ability of ILs to be non-volatile and flammable has made ILs widely sought after and developed as a substitute solvent that is more environmentally friendly than conventional volatile organic solvents (VOC) (Rogers & Seddon, 2003). Even so, in several reports some ILs were found to still have a negative impact on the environment. For industrial purposes, ILs are also not good because they require high production costs compared to conventional solvents. Therefore, research on ILs that are more efficient, low cost, and non-toxic is the subject of research that is actively being developed to date (Asim et al., 2019).

Generally, ILs are defined as compounds consisting entirely of ions with a melting point below 100 °C (Lei et al., 2017). Typically, they contain an organic cation (ammonium, imidazolium, pyridinium, piperidinium or pyrrolidinium), and halogen,

fluorinated or organic anions (Flieger & Flieger, 2020). These structures formed between cations and anions provide unique physical and chemical properties and can be engineered to increase the efficiency of a wide range of electrochemical, analytical, synthetic and engineering processes (Wasserscheid, 2006). Therefore, as of the end of 2018, more than 80,000 scientific papers have been published, and 17,000 patent applications have been filed for ILs (Kalb, 2020).

Several of the research developments for ILs have resulted in various types and subclasses. The developed ILs can be categorized to subclasses such as Chiral ILs, Supported ILs, Protic ILs, Task specific ILs, Bio ILs, Amphiphilic ILs, Polarizable ILs, Metal slats ILs and Swichable polarity solvents (Sadjadi, 2021). Deep Eutectic Solvents (DESs) were also introduced as an alternative to overcome the weakness of ILs solvents or some researchers call it a subclass of ILs. Compared to its predecessors, the advantages of DESs relate to their easy synthesis and low production costs due to the low price of synthetic raw materials (Płotka-Wasylka et al., 2020).

As a new solvent with potential for the production of low cost and environmentally friendly ILs and their variations have been reported in various articles such as by Jin, et al., wherein phosphonium-based ionic liquids were reported to have become popular green solvents for metal separation (Jin et al., 2021). Research by Ziaei-Rad, et al. (2021) demonstrated a successful and cost-effective pretreatment method by applying low-cost triethylammonium hydrogen sulfate [TEA][HSO4]-based ILs to wheat straw fractionation (Ziaei-Rad et al., 2021). Xu et al. (2019) also reported a low cost AlCl3/Et3NHCl room temperature ILs electrolyte for preparing a practical but high performance Al-graphene battery (Xu et al., 2019). However, research on bibliometric analysis is still lacking in the field of Ionic Liquids as low-cost and environmentally friendly solvents, especially by using Publish or Perish and VOSviewer software as mapping analysis tools.

Publish or Perish" is a software program that retrieves and analyzes academic citations (Baneyx, 2008). VOSviewer is a program developed for creating and viewing bibliometric maps. This program is freely available to the bibliometric research community (van Eck & Waltman, 2010). Bibliometric data analysis that is displayed visually through mapping tools is needed in the era of technological growth that is developing so rapidly as it is today (Al Husaeni & Nandiyanto, 2021).

This research is expected to help and become a reference for researchers in conducting and determining research topics to be taken, especially those related to the field of ILs as green and low cost solvent

2. Methods

The global literatures about ILs as Green and Low-Cost Solvents published between 2013 to 2023 range. The search terms applied to identify the closest matching publication included "Ionic Liquids" or "ILs", "Green Solvent" and "Low-Cost" which was used as the keyword in the title. The reference managers application used in this research is Publish or Perish (PoP). Thus, 1000 articles were obtained which were assessed according to the chosen topic. The collected articles were then saved in *.ris format. Next, we used the VOSviewer application to visualize and analyse trends in the form of bibliometric maps. We did data mapping articles from database sources that have been prepared. Data mapping consists of three types, namely network, density, and overlay visualization. In addition, we also filtered the terms that would be included in the VOSviewer network mapping visualization (Shidiq, 2022).

3. Results and Discussion

3.1. Research Developments in the Field of Ionic Liquids As Green and Low-Cost Solvents

Figure 1 shows an overview of the growth or development of research in the field of ILs as a low cost and environmentally friendly solvent every year. Obtained Meta-data for 1000 Publication from PoP data processing application. With details of the number of articles in 2013 there were 156 articles, in 2014 there were 184 articles, in 2015 there were 145 articles, in 2016 there were 119 articles, in 2017 there were 112 articles, in 2018 there were 107 articles, in 2019 there were 74, in 2020 there were 50 articles, in 2021 there were 27, in 2022 there were 19 articles, and in 2023 there were 7. The peak of publication occurred in 2014 with 184 articles.



Figure 1. Level of development in ionic liquids as green and low-cost solvent keyword research

The trend in the number of publications for ILs as a low cost and environmentally friendly solvent was observed to decrease from 2015 to 2023. The sharp decline occurred in 2018-2020, one of the factors due to the covid pandemic. While the decline in general was also caused by various factors, one of which was because research had entered the era of applied chemistry. Nevertheless, research on ILs as a cheap and environmentally friendly solvent is still important and needs to be increased.



Figure 2. Total publications in ionic liquids as green and low-cost solvent keyword from various publishers.

The articles were published by 47 publishers, although only 8 contributed more than 70%. Figure 2 shows a list of the most contributing publishers and the number of articles they published. This shows that most of the published articles are related to journals that are heavily indexed by Scopus. Where articles were published in 217 journals, such as ACS Sustainable Chemistry & Engineering totaling 66 articles, Green chemistry totaling 23 articles, Green Energy & Environment 20 articles, Chemical Society Reviews 15 articles, and many more. Seeing how approaches to research topics have evolved can help us make sense of these numbers. As in other disciplines, the oldest and most topic-focused journals top the list. The h-index indicator strengthens the ranking position of these journals. Because they constitutively subscribe to diverse viewpoints, other journals provide different perspectives on the arena.

3.2. Cluster Resulting from the VOSviewer Mapping with the Keywors of Ionic Liquids As Green and Low-Cost Solvents

The minimum number of relationships between terms in VOSviewer is governed by 2 terms (Al Husaeni & Nandiyanto, 2021). Research related Ionic Liquids as Green and Low-Cost Solvents based on analysis mapping visualization is divided into 9 clusters with 131 items, namely:

(i) Cluster 1 which is marked dark red has 23 items (see Figure 3), the 23 items are agent, alternative solvent, anion, battery, cation, development, electrolyte, environment, high ionic conductivity, high polarity, hydrophobic ionic liquids, imidazolium ionic liquids,

ionic liquid, low melting point, low temperature, low vapor pressure, low volatility, membrane, metal organic frameworks, non-volatility, organic salt, and thermal stability.



Figure 3. Network visualization of electrolyte term in cluster 1.

(ii) Cluster 2 which is marked light red has 22 items (see Figure 4), the 22 items are alternative, choline chloride, energy, ethyl acetate, ethylene glycol, high purity, imidazolium, low viscosity, mixture, new class, new green solvent, novel green solvent, organic solvent, pil, polymeric ionic liquid, promising green solvent, protic ionic liquids, recovery, researcher, solubility, solution, urea.



Figure 4. Network visualization of alternative term in cluster 2.

(iii) Cluster 3 which is marked dark blue has 18 items (see Figure 5), the 18 items are alternative green solvent, biocatalysis, biodegradability, discovery, enzyme, eutectic solvent, fabrication, high yield, low cost, lower cost, microalgae, new type, overview, processing, production, protein, stability, sustainability.



Figure 5. Network visualization of low cost term in cluster 3.

(iv) Cluster 4 which is marked yellow has 17 items (see Figure 6), the 17 items are acid, acidic ionic liquid, biodiesel production, conversion, effective solvent, extraction, lignocellulosic biomass, lower price, metal, phophonium ionic liquid, potential application, preparation, recyclability, removal, room temperature ionic, separation, solvent extraction.



Figure 6. Network visualization of extraction term in cluster 4.

(v) Cluster 5 which is marked purple has 17 items (see Figure 7), the 17 items are carbon dioxide, catalysis, catalyst, efficient catalyst, friendly solvent, functionalized ionic liquids, high efficiency, interest, low flammability, reaction, reagent, recyclable catalyst, synthesis, taks specific ioniq liquid, unique property.



Figure 7. Network visualization of catalyst term in cluster 5.

(vi) Cluster 6 which is marked bright pink has 10 items (see Figure 8), the 10 items are addition, application, benign solvent, extractive desulfurization, fuel, green chemistry, molecular solvent, research, selection, solvent system.



Figure 8. Network visualization of fuel term in cluster 6.

(vii) Cluster 7 which is marked green has 10 items (see Figure 9), the 10 items are biomass, chemistry, designer solvent hydrolysis, investigation, ion, lignin, lignocellulose, potential green solvent, reduced cost.



Figure 9. Network visualization of biomass term in cluster 7.

(viii) Cluster 8 which is marked brown has 8 items (see Figure 10), the 8 items are absorption, deep eutectic solvent, easy preparation, low price, low toxicity, new generation, oxidative desulfurization, ultrasound.



Figure 10. Network visualization of deep eutectic solvent term in cluster 8.

(ix) Cluster 9 which is marked dark pink has 8 items (see Figure 11), the 2 items are extraction solvent, green alternative, green analytical chemistry, green solvent, green technology, low vapor, new solvent, supercritical fluid.



Figure 11. Network visualization of green solvent term in cluster 9.

3.3. Network Visualization of Ionic Liquids As Green and Low-Cost Solvents Science Keyword

Figure 12 shows the relationship between the terms described in the network. Relationships in network visualization are depicted with lines that come from one term to another (Al Husaeni & Nandiyanto, 2021). Figure 12 shows the cluster of each term studied and related to the topic of ILs as a cheap and environmentally friendly solvent. Based on Figure 12, the term ionic liquid is in cluster 9 with a total strength of 8419. The ideal ILs connected to cluster 3 and 9 are the terms green chemistry and low cost solvent.



Figure 12. Network visualization of Ionic Liquids As Low-Cost and Green Solvent keywords.

Figure 13 shows an overlay visualization in research on ILs as a cheap and environmentally friendly solvent. The visualization overlay shows each term's updates. Figure 3 shows that of the 1000 articles spanning 2013-2023, terms that fall into the appropriate category and are related to the Ionik Liquids keyword are in the range June 2017 to April 2018. The color of each term indicates the novelty of the terms. The brighter the yellow color, the higher the renewal rate. Based on Figure 13, it is shown that the latest research trend recently, namely related to Ionic Liquids as a cheap and environmentally friendly solvent is its application in the fuel sector. Therefore, this can be a reference for future researchers to determine themes related to the field of ILs.



Figure 13. Overlay Visualization of Ionic Liquids As Low-Cost and Green Solvent keywords.

Figure 14 shows a visualization of density, meaning that the darker or lighter the yellow color and the larger the circle diameter of a term, the more often that term appears. That is, research on related terms is increasing in number. If the color of the term fades closer to the background color, the amount of research on the term will decrease (van Eck & Waltman, 2010). Based on Figure 14, it can be seen that research related to application, production, extraction, price reduction, catalyst, and performance has a high number of studies.



Figure 14. Density visualization of Ionic Liquids As Green and Low Cost Solvent keywords.

Based on the results of the mapping of the collected article data, it appears that the keywords that appear the most are low-cost, green solvent, application, study, and extraction. From this data, we can look for research trends on application engineering and deployment. As in the field of fuel, which is the most trending term, but there are still very few people who research it. It could also be research in the field of transportation related to environmental health where there is still little research scope that has ever been carried out.

3.4 Search Result Data Using Publish or Pherish with The Keyword Ionic Liquids As Low Cost and Green Solvent

In the Publish or perish application, we can find out some data information as shown in Table 1 below.

 Table 1. Citation of Ionic Liquids As Green and Low Cost Solvent.

Description	Data
Publication years	2013-2023
Citation years	10 (2013-2023)
Papers	1000.00

Citations	162046,00	
Cites/year	16204,60	
Cites/paper	162.05	
Authors/paper	4.12	
h-index	199	
g-index	324	
hl,norm	101	
Hl, annual	10.10	
hA, index	61	

In addition to the data above, the results of searching the databases of several journals obtained several data articles that meet the research criteria. Of the 1000 articles obtained, there are 20 articles with the highest number of citations as shown in Table 2 below.

Table 2. Examples of articles with the most citations based on the keyword ionic liquids as low cost, and green solvent.

cost, and green solvent.					
No.	Cites	Authors	Title	Year	Source
1	4411	(L. Smith et al., 2014)	Deep eutectic solvents (DESs) and their applications	2014	Chemical reviews
2	1894	(Hayes et al., 2015)	Structure and nanostructure in ionic liquids	2015	Chemical reviews
3	1822	(Dai et al., 2013)	Natural deep eutectic solvents as new	2013	Analytica
4	1623	(Paiva et al., 2014)	potential media for green technology Natural deep eutectic solvents-solvents for the 21st century	2014	Chimica Acta ACS Sustainable Chemistry & Engineering
5	1586	(Macfarlane et al., 2014)	Energy applications of ionic liquids	2014	Energy Environ
6	1513	(Brandt et al., 2013)	Deconstruction of lignocellulosic biomass with ionic liquids	2013	Green Chemistry
7	1261	(Gałuszka et al., 2013)	The 12 principles of green analytical chemistry and the SIGNIFICANCE mnemonic of green analytical practices	2013	TrAC Trends in Analytical Chemistry
8	1193	(Yuan et al., 2013)	Poly (ionic liquid) s: An update	2013	Progress in Polymer Science
9	1192	(Watanabe et al., 2017)	Application of ionic liquids to energy storage and conversion materials and devices	2017	Chemical Reviews
10	1184	(Egorova et al., 2017)	Biological activity of ionic liquids and their application in pharmaceutics and medicine	2017	Chemical Reviews
11	1179	(V. Fedorov & A. Kornyshev, 2014)	Ionic liquids at electrified interfaces	2014	Chemical Reviews
12	1158	(J. Clarke et al., 2018)	Green and sustainable solvents in chemical	2018	Chemical Reviews
13	1133	(Francisco et al., 2013)	processes Low-transition-temperature mixtures (LTTMs): A new generation of designer solvents	2013	Angewandte Chemie International Edition
14	1004	(Alonso et al., 2013)	Gamma-valerolactone, a sustainable platform molecule derived from lignocellulosic biomass	2013	Green Chemistry
15	964	(Sheldon, 2017)	The E factor 25 years on: the rise of green chemistry and sustainability	2017	Green Chemistry
16	900	(Byrne et al., 2016)	Tools and techniques for solvent selection: green solvent selection guides	2016	Sustainable Chemical
17	889	(Duan et al., 2015)	Green chemistry for nanoparticle synthesis	2015	Chemical Society Reviews
18	885	(Lei et al., 2014)	Gas solubility in ionic liquids	2014	Chemical Reviews
19	827	(Vekariya, 2017)	A review of ionic liquids: Applications towards catalytic organic transformations	2017	Journal of Molecular Liquids
20	793	(Song et al., 2017)	Efficient, selective and sustainable catalysis of carbon dioxide	2017	Green Chemistry

4. Conclusion

The bibliometric indicator refers to 1,000 different relevant articles from 52 publishers with the largest number of articles produced by Elsevier, totaling 423 articles. The results showed that the peak of production occurred in 2014 and then decreased every year. The main reason for this increase is the need for cheap and environmentally friendly solvents to replace expensive and environmentally damaging conventional solvents, while the decline is due to the shift of interest into the field of applied chemistry. From the results of the Bibliometric analysis, there are 9 clusters with 126 related terms including low-cost, green solvent, application, study, and extraction. The most popular terms in 2022-2023 are related to fuel terms. This study shows the importance of bibliometric analysis in providing analytical data about what phenomena occur.

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