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Insights into Ionic Liquids for Flame Retardant: A Study Based on Bibliometric Mapping

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Abstract: Fire is a typical disaster in the processing industry. Ionic liquids, as a type of green flame retardant, play an important role in process safety. In order to grasp the current research status, hotspots, and frontiers in the field of ionic liquids in flame retardancy, the bibliometric mapping method is applied to study the relevant literature in Web of Science datasets from 2000–2022 in this paper. The results show that the research on ionic liquids in flame retardancy is multidisciplinary and involves some disciplines such as energy science, material science, and environmental protection. Journal of Power Sources, Polymer Degradation and Stability, ACS Applied Materials and Interfaces, and Chemical Engineering Journal are the core journals in the field. The results of keyword co-occurrence indicate that the hotspots of research can be divided into five components: the improvement and application of pure ionic liquids electrolytes, the research of gel polymer electrolytes, applying ionic liquids to enhance the polymer materials' flame retardancy properties, utilizing ionic liquids and inorganic materials to synergize flame retardant polymers, and using ionic liquids flame retardant to improve material's multiple properties. The burst terms and time zone diagram's results point out the combination of computational quantum chemistry to study the flame retardancy mechanism of ionic liquids, the study of fluorinated electrolytes, ionic liquids for smoke suppression, phosphorus-containing ionic liquids for flame retardant, and machine learning-assisted design of ILs flame retardants are the research frontiers and future research trends.

Keywords: ionic liquids; flame retardant; lithium-ion battery; polymers; bibliometric mapping

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1. Introduction

Ionic liquids (ILs) usually refer to liquid salts at room or near room temperature, generally composed of organic cations and inorganic or organic anions [1]. As early as 1914, the first room temperature IL ([$EtNH_3$][NO_3]) was accidentally synthesized by Welton [2] but did not attract widespread attention due to its poor stability and certain dangers. Subsequently, the first generation of ILs, chloroaluminate ILs [3], was synthesized by Wiler and Hurley, which was very unstable to water and air and easily decomposed. So, in the next 40 years, the application of ILs was greatly limited until Wilkes [4] synthesized the first anti-hydrolysis IL ([Emim][BF_4]) in 1992, and ILs came into the development stage of the second generation of anti-hydrolytic ILs. Then, into the 21st century, with the rise of the green chemistry concept, ILs have developed rapidly, and many functionalized ILs have been synthesized and developed. This kind of IL is the third generation ILs [5], which exhibits not only properties such as nonvolatility, good thermal stability, high chemical stability, high thermal conductivity, and wide electrochemical window [6] but also has the advantage of high designability. Therefore, the third generation of functionalized ILs has been gradually applied in organocatalysis [7], metal corrosion inhibition [8], and material flame retardancy [9].

Among the many applications of ILs mentioned above, flame retardancy research can be considered one of the emerging hot spots in recent years. Compared with traditional flame retardants, IL flame retardants tend to be non-toxic, green [10], and highly designable [11]. Moreover, they also tend to improve multiple properties of materials, such as thermal conductivity, mechanical properties, and electrical conductivity, when used as flame retardants for materials [12,13]. Kuo et al. [14]obtained a high ionic conductivity and non-flammable gel polymer electrolyte prepared using a mixture of oligomeric IL, PVDF-co-HFP, and organic liquid electrolyte. Aghmih et al. [15] used four ILs ([C_6MIM][Ac], [C_6MIM][PF_6], [N-2226][Ac] and [N-2226][PF_6]) as additives for the production of flexible biocomposite films. The final product, flexible biocomposite film, is characterized by thermal insulation, fire resistance, smooth surface, high transparency, good ductility, and toughness. Although ILs have shown many advantages in flame retardancy, more research in this field may still be at a preliminary stage, and the review literature is also very scarce. Therefore, a systematic overview of the research on ILs in flame retardancy and a frontier outlook will be of great significance for constructing the whole field, the bibliometric mapping method can exactly be applied to perform this systematic overview [16].

The bibliometric mapping method integrates traditional bibliometrics with modern text mining, complex networks, mathematics, statistics, computer science methods, and visualization technologies to study the distribution characteristics, change patterns, and quantitative relationships of the literature and to further explore the hot spots and frontiers of research within a specific research field [17–20]. In the traditional sense, this method requires a specialized knowledge base in bibliometrics and informetrics. Therefore, the bibliometric method is unsuitable for non-specialized bibliometric researchers to analyze the literature in their research fields, which makes applying this method more limited. In recent years, with the development of computer technology, a group of highly reliable and widely used bibliometric software has emerged, such as VOSviewer, CiteSpace, etc. This kind of software can help non-specialist bibliometric researchers to conduct bibliometric analysis and systematically obtain the current status, hotspots, and frontiers in their research fields. For example, Lawal et al. [18] conducted a bibliometric analysis of the application of ILs during 1930–2017 using VOSviewer software. The paper described the possibility of using ILs to modify adsorbents and adsorb organic pollutants. On the other hand, Lang et al. [19] conducted a bibliometric analysis of fire safety-related papers between 2000–2019, applying VOSviewer and CiteSpace software. It can be observed that the quantity of such studies in a particular field with the help of bibliometric software is expanding, which forms an emerging research boom. Thus, in this paper, we will systematically review the research on ILs in the flame retardancy field with the assistance of bibliometric software to capture the current status, research flow, research hotspots, and frontiers. We expect this paper can provide a reference for future in-depth research on ILs in flame retardancy, thus contributing to developing more environmentally friendly and efficient IL flame retardant materials.

2. Data and Methods

2.1. Data Collection

Web of Science (WOS) database is one of the most authoritative, widely used, and comprehensive academic databases in the world today. It covers various scientific literature under different disciplines and is an effective tool for the literature search and bibliometric analysis [20]. Therefore, in this paper, the SCI-E and SSCI datasets from the WOS core database will be utilized to collect information on the literature related to ILs in the field of flame retardancy [21]. In order to search as much literature as possible and to make the searched literature highly relevant to the research content of ILs flame retardant, different search formulas were applied in this paper, and a group of three researchers judged the relevance of the searched literature, and the results are shown in

Table 1. Based on the combination of relevance and the literature quantity, we finally determined the search formula for this paper as: (TI = "ionic liquid" AND (TI = "flame retardant" OR TI = "fire retardant")) OR (AB = "ionic liquid" AND (AB = "flame retardant" OR AB = "fire retardant")) OR (AK = "ionic liquid" AND (AK = "flame retardant" OR AK = "fire retardant")). Finally, according to this search formula, 74 highly relevant pieces of literature were obtained (similar to the number of literature obtained using the Scopus database and EI database search), and the information was then stored in "Data Set 1". Due to the small amount of literature in Data Set 1, there are limitations in using the literature in this database for bibliometric analysis. Thus, we expanded the number of literature in Data Set 1 to obtain more comprehensive literature content.

Table 1. Search results obtained using different search formulas.

NO.	SF	TP	RR
F1	TI = "ionic liquid" AND (TI = "flame retardant" OR TI = "fire retardant")	27	H
F2	AB = "ionic liquid" AND (AB = "flame retardant" OR AB = "fire retardant")	56	H
F3	AK = "ionic liquid" AND (AK = "flame retardant" OR AK = "fire retardant")	25	H
F4	TS = "ionic liquid" AND (TS = "flame retardant" OR TS = "fire retardant")	116	M
F5	(F1) or (F2) or (F3)	74	H

SF: Search Formula; TP: Total Publications; RR: Relevance to the Research; H: High Relevance; M: Medium Relevance; TI: Tittle; AB: Abstract; AK: Author Keywords; TS: Topic.

The extension method: If the literature has cited one of these 74 pieces of literature, it will be included in the extended data set [22]. By this method, we acquired 1273 pieces of literature and stored the information of this literature in "Data Set 2". After the above operation, we merged the data in "Data Set 1" and "Data Set 2" into "Data Set 3" and finally obtain 1308 pieces of literature. All literature types in "Data Set 3" are shown in Table 2. As we can see from Table 2, the most numerous type of literature is the article, whose literature total reached 1120 with an average citation of 15.34. The second highest-ranking literature type is the review article, which contains 182 pieces. It is worth noting that the ratio of review articles to articles in the table is high, reaching about 1/6. A large amount of review articles in this field helps scholars efficiently and accurately grasp the current status of research on ILs flame retardant, promptly identify the shortcomings and unsolved problems in existing research, and clarify future research problems and directions for consideration. In addition, the average citation of review articles is at a high level, reaching 75.79. This phenomenon may be caused by researchers reading and citing many review articles replacing the original literature to save time during the study. In contrast, a large gap between the total amount of literature in the proceedings paper type and the first two, with 18 papers in this category and an average citation of 27.01. Other types of literature include early access, book chapters, and edition materials, all of which have low average citations and H-indexes.

Table 2. Types of literature related to ILs in the field of flame retardancy.

NO.	TD	TP	SOTC	ACI	H-Index
1	Article	1120	17,180	15.34	60
2	Review Article	182	13,794	75.79	51
3	Proceedings Paper	18	486	27.01	9
4	Others	45	33	0.73	3

TD: Type of Document; TP: Total Publications; SOTC: Sum of Times Cited; ACI: Average Citations per Item.

2.2. Analysis Tools

In this paper, VOSviewer, Citespace, and Histcite bibliometric software were mainly performed to analyze and visualize the bibliographic data in "Data Set 3". Among them, VOSviewer is a bibliometric software developed by Van Eck and Waltman from Leiden University in the Netherlands, which has been widely used in bibliometric research due to its user-friendly interface and reliable analysis results [23,24]. This software allows us to perform Co-authorship analysis (country, authorship, institutional partnership), Keywords Co-occurrence analysis, and Co-citation analysis (literature co-citation, journal co-citation analysis) on the collected literature [18] to obtain the research's current status of ILs in flame retardancy. The developer of Citespace software is Professor Chaomei Chen of Drexel University, USA [25]. Based on the literature co-citation network and specific algorithms in this software, we can achieve information measurement of research features related to ILs flame retardancy, thus revealing the research hotspots and frontiers in the field of ILs flame retardancy [19,26]. The Histcite software is a visualization system introduced by SCI founder Eugene Garfield and his colleagues. This software allows analysis of the literature related to ILs in the field of flame retardancy based on the time series of citations and citation relationships. It also can be applied to visualize the original authors under the ILs flame retardant research topic and the ins and outs of the research topic's development with a citation analysis chart. Then ultimately reflects the development history and research patterns in the field of ILs flame retardant [27].

Although all three bibliometric softwares' have certain limitations, they can effectively compensate for their shortcomings when combined, while the respective advantages of these three software will be organically combined [28]. Therefore, VOSviewer, Citespace, and Histcite bibliometric software were carried out in this paper to enable an in-depth exploration of the current status, development history, frontiers, and hotspots of research on ILs in flame retardancy [29].

2.3. Analysis Method

This paper presented a systematic literature study related to ILs in flame retardancy with bibliometric mapping methods. The study was divided into two main parts; (1) characterization of the current research status; (2) analysis of research flow and frontier hotspots. The overall analysis method and process are shown in Figure 1, in which the results of research status characterization will be the basis for detecting the research flow and frontier hotspots. After combining the current research status, flow, and frontier hotspots of ILs in the flame retardancy field, we will finally form an objective overview of this research field. Then, the results will provide other scholars with a comprehensive reference for ILs in flame retardancy research and point out possible future research and application directions.

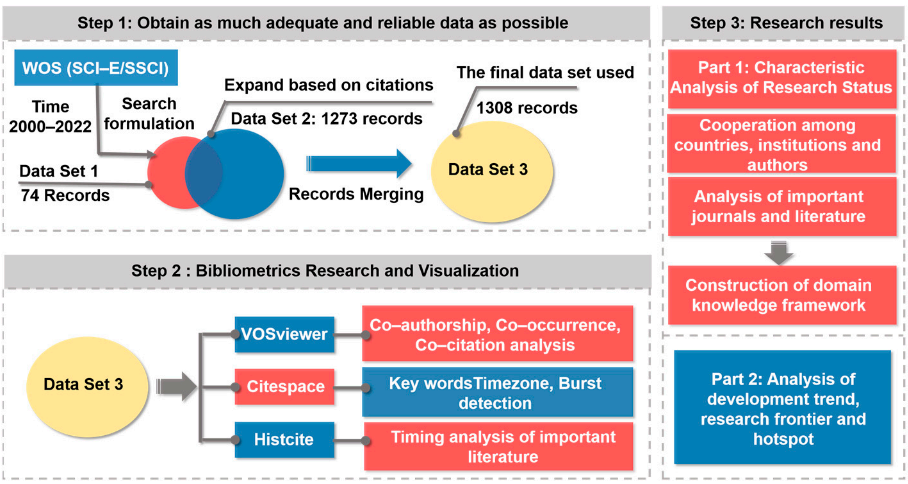


Figure 1. The main research steps and research methods of this paper.

In the current research status characterization, the VOSviewer software was applied to study the cooperative relationships between countries, institutions, and authors [30,31]. Then, the related social networks were mapped thus to grasp the essential development of ILs in flame retardancy. In addition, this paper also reviewed the critical literature (highly cited literature, highly co-cited literature) on ILs in the flame retardancy field and deduced the knowledge base in this field. In the analysis of research flow and frontier hotspots, the Histcite software was performed to visualize the network relationship of important literature citation time series, thereby identifying the flow of significant research on ILs in the flame retardancy field. The Citespace software was also used to obtain the time zone diagram and perform burst term analysis of keywords to derive the possible frontier directions of ILs in flame retardancy research.

3. Results

3.1. The Literature Publication Trends Analysis

Based on the "Data Set 3" established in Section 2.1 above, this paper will first analyze the literature's publication trends pattern to derive the general trend of ILs in flame retardancy research. The literature's publication trends are shown in Figure 2, and this figure also contains the publication trends by active countries (the top three countries in the total number of publications). SOTC in this figure is the literature's total citations, and the H-index represents the published literature's overall impact level. It can be seen from Figure 2 that the number of publications on ILs in flame retardancy has generally shown an increasing trend worldwide. Of all countries, China, with the most significant number of publications, also has the most prominent growth rate in recent years, and its growth rate is much larger than that of the United States and Australia. In addition, the SOTC value and H-index are generally increasing (the SOTC data for 2022 is incomplete, and the H-index for 2021 and 2022 is lower due to delayed citation), indicating that the research enthusiasm of ILs in the field of flame retardancy has been increasing.

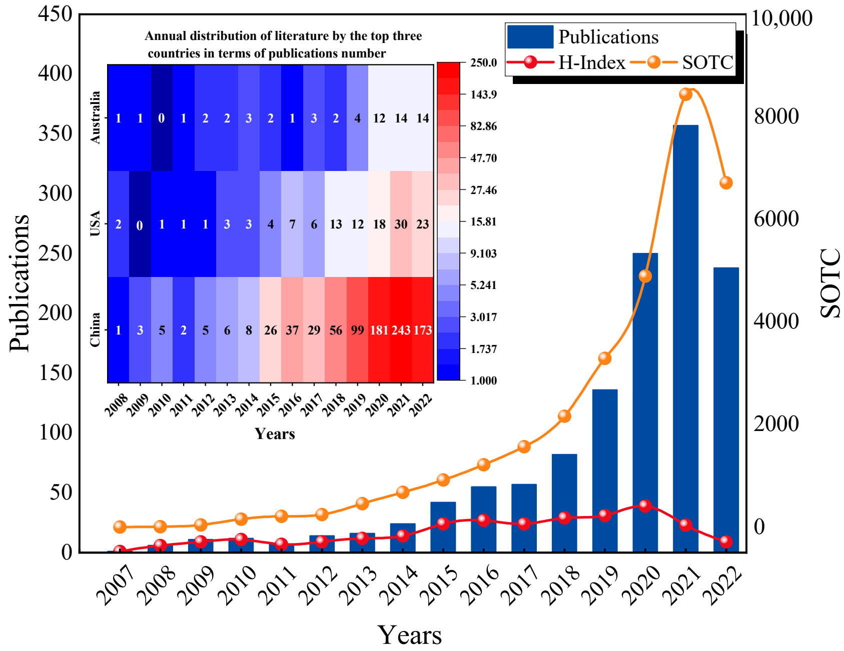
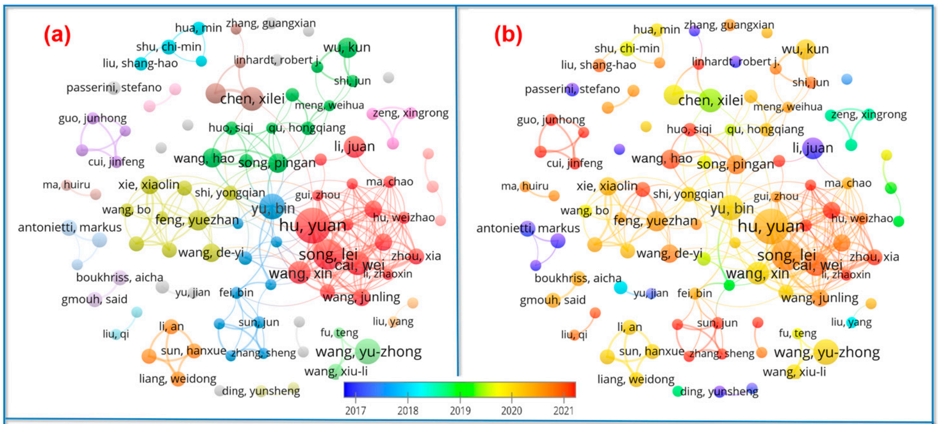


Figure 2. The literature publication trends related to ILs in the flame retardancy field.

3.2. Principal Researchers and Cooperation Relationships Analysis

Authors of publications are the scientific research's main body [32]. Analyzing the structural characteristics of the posting authors and their collaborative networks can reflect the core group of authors and their collaborative relationships in ILs in flame retardancy research. In this paper, the VOSviewer software was carried out to choose authors with more than six pieces of articles for analysis, and the 99 nodes and a total of 28 clusters were formed, as shown in Figure 3a. Among these 28 clusters, 12 have more than three authors, while 8 contain only one author. The top three clusters in terms of author number are the red cluster (17 authors), the green cluster (11 authors), and the blue cluster (10 authors). It can be seen from Figure 3a that the red cluster is not only the largest in overall size but also contains nodes of equally large size compared to other clusters.



NO.	Author	Quantitiy	Institution	Country	Links	ACI
1	Hu,Yuan	30	University of Science and Technology of China	China	122	27.8
2	Song, Lei	24	University of Science and Technology of China	China	111	24.5
3	Wang, Yuzhong	19	Sichuan University	China	16	32.0
4	Yu, Bin	19	University of Science and Technology of China	China	42	62.1
5	Cai, Wei	17	University of Science and Technology of China	China	85	21.4
6	Chen, Xilei	17	Qingdao University of Science and Technology	China	21	9.6
7	Wang, Xin	16	University of Science and Technology of China	China	56	29.6
8	Li, Juan	15	Chinese Academy of Sciences	China	3	21.3
9	Feng, Yuezhan	14	Zhengzhou University	China	42	35.1
10	Jiao, Chuanmei	14	Qingdao University of Science and Technology	China	19	7

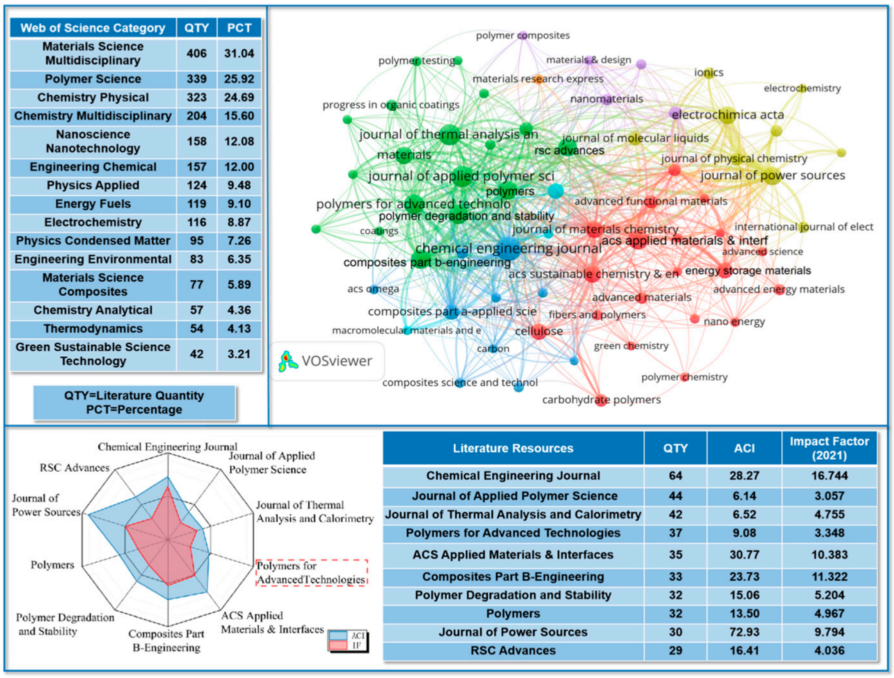
Figure 3. (**a**) Key researchers and collaborations in the field of ILs in flame retardancy, (**b**) the time superimposed figure of key researchers and collaborations.

Meanwhile, we can also find that the red cluster has been more active in recent years from the time superimposed in Figure 3b. This situation suggests that the collaborative group represented by the red cluster is leading in terms of the collaboration scale and publication number. It may be a leading research group about ILs in the flame retardancy field. When analyzing the authors' affiliations in the red clusters, we concluded that the authors from the University of Science and Technology of China formed a strong cooperation relationship. We also noticed that the green and blue clusters in Figure 3a are interspersed and dense with connected lines, indicating a close collaboration between the two research groups. The author affiliations' analysis result of these two clusters revealed complex academic collaborations between authors from multiple institutions, such as the University of Science and Technology of China, the Chinese Academy of Sciences, and the University of Southern Queensland. Therefore, we conclude that the author collaborations in the red clusters are relatively homogeneous and tend to be limited to the same institution, while authors in the green and blue clusters appear to have more extensive and good academic collaborations. In addition, some smaller clusters also exist in Figure 3a, such as orange and purple clusters. The authors in these clusters tend to post fewer articles, while the intensity of collaboration among authors is low.

When combining the top 10 authors in Figure 3, we can find that two of the top three authors are from the University of Science and Technology of China, namely Hu, Yuan, and Song, Lei. Their article numbers reached 30 and 24 items, respectively, but their ACI (27.8, 24.5) was only at an average level (average ACI = 27.04) among the top ten authors. From the published literature, the two scholars likely belong to the same research team, and their related studies have focused more on the flame retardancy of polymers [33,34]. The third author, Wang Yuzhong, is from Sichuan University, with 19 publications and an ACI of 32.0, which is at a higher level. This author's research team has synthesized two new phosphorus-containing ILs ([Pmim]CH_3SO_3 and [Pmim]Ts) with different sizes of sulfonate anions and comparatively studied their flame retardant effects on nylon 6 [35]. Interestingly, author Yu, Bin connects the red, green, and yellow clusters among the blue clusters, while the scholar also has the highest ACI ranking in the table. This phenomenon may imply that Yu's team has contributed more to developing the ILs flame retardant field, and this team's research has focused more on developing nanocomposites' flame retardancy functions [36]. In addition, Feng Yuezhan from Zhengzhou University and Wang Xin from the University of Science and Technology of China also have high ACI and good external collaboration situations.

3.3. Subject Areas and Journal Distribution Analysis

In this section, the disciplinary distribution pattern of literature related to ILs in flame retardancy was studied, the top 15 disciplines of literature volume were extracted for analysis, and the results are shown in Figure 4. The table in Figure 4 shows that the top three disciplines in the number of articles are materials science multidisciplinary, polymer science, and chemistry physical, which account for the vast majority of all literature. Due to the widespread application of flame retardants in polymers and batteries, when ILs flame retardants are used as flame retardants, most of their applications revolve around materials science, polymers, electrolytes, etc. In addition, from these 15 disciplines, we can also find that the literature has strong relevance to environmental protection and sustainable development, such as engineering environmental and green sustainable science technology, which contain about 10% of the literature. This phenomenon laterally reflects ILs' environmental friendliness and sustainability in flame retardant applications. When combining the results of the above disciplinary analysis, it can be regarded that there is a crossover of several disciplines in the research of ILs for flame retardancy.



Web of Science Category	QTY	PCT
Materials Science Multidisciplinary	406	31.04
Polymer Science	339	25.92
Chemistry Physical	323	24.69
Chemistry Multidisciplinary	204	15.60
Nanoscience Nanotechnology	158	12.08
Engineering Chemical	157	12.00
Physics Applied	124	9.48
Energy Fuels	119	9.10
Electrochemistry	116	8.87
Physics Condensed Matter	95	7.26
Engineering Environmental	83	6.35
Materials Science Composites	77	5.89
Chemistry Analytical	57	4.36
Thermodynamics	54	4.13
Green Sustainable Science Technology	42	3.21

QTY=Literature Quantity
PCT=Percentage

Literature Resources	QTY	ACI	Impact Factor (2021)
Chemical Engineering Journal	64	28.27	16.744
Journal of Applied Polymer Science	44	6.14	3.057
Journal of Thermal Analysis and Calorimetry	42	6.52	4.755
Polymers for Advanced Technologies	37	9.08	3.348
ACS Applied Materials & Interfaces	35	30.77	10.383
Composites Part B-Engineering	33	23.73	11.322
Polymer Degradation and Stability	32	15.06	5.204
Polymers	32	13.50	4.967
Journal of Power Sources	30	72.93	9.794
RSC Advances	29	16.41	4.036

Figure 4. Discipline and journal distribution analysis of literature on ILs in the flame retardancy field.

After grasping the disciplinary distribution pattern in the ILs flame retardancy field, we analyzed the research's distribution pattern related to ILs flame retardancy in journals. In this section, we selected journals with more than five publications for cluster analysis, resulting in 66 nodes forming seven clusters. The information about each cluster and the top 10 journals ranked by the number of publications is shown in Figure 4. We can see that the top three clusters in the figure in scale are the red cluster, green cluster, and blue cluster, which respectively contain 20, 18, and 10 nodes. The journals in the green and blue clusters are mainly associated with materials chemistry and polymers, while the journals in the red cluster are mainly related to environmental protection, sustainable development, and energy. In addition, it is worth noting that the Chemical Engineering Journal, represented by the largest node in the figure, is closely associated with the red and green clusters. This phenomenon suggests that the Chemical Engineering Journal may be fundamental and critical in the field of ILs in flame retardancy. The top three journals in terms of publication numbers are the Chemical Engineering Journal (64), the Journal of Applied Polymer Science (44), and the Journal of Thermal Analysis and Calorimetry (42). The ACI and the impact factors of the remaining two journals are low except for Chemical Engineering Journal. This situation indicates that, except for the top-ranked journal, the remaining two journals are of average influence in research of ILs in flame retardancy.

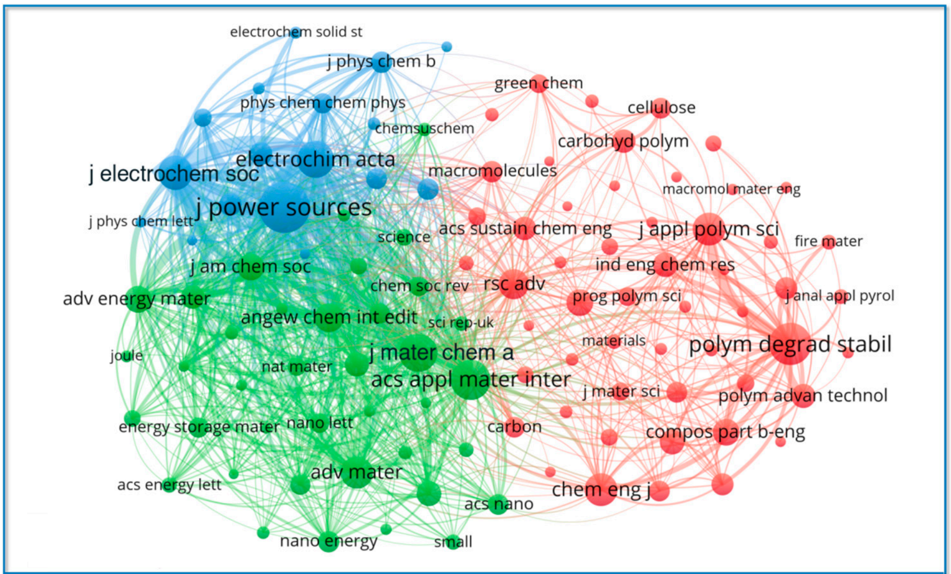
For the average citation aspect, the top three journals are the Journal of Power Sources, ACS Applied Materials & Interfaces, and Chemical Engineering Journal, with ACI of 72.93, 30.77, and 28.27, respectively. Meanwhile, the impact factors of these journals are also at high levels, at 9.794, 10.383, and 16.744, respectively. After combining the contents of the previous subject areas analysis, the three journals are mainly concerned with two topics: polymers and batteries. The Journal of Power Sources mainly focuses on batteries, so this journal may strongly influence ILs for batteries' flame retardant applications. The ACS Applied Materials & Interfaces and Chemical Engineering Journal have both batteries and polymers research topics, so both journals may strongly influence ILs for batteries and polymers' flame retardant applications. In addition, we can also observe from the radar plot in Figure 4 that, except for Polymers for Advanced Technologies, the ACI of the journals show a positive correlation with the impact factor of the journals in general. It means that the journal's impact factor in the field of ILs flame retardant batteries and polymers has a specific relationship with the journal's ACI but not with the publication number.

3.4. The Research Current Status and Basic Knowledge Analysis

Co-citation analysis originated from the concept of literature co-citation proposed by H. Minor in 1973 [37], which means that two articles appear together in a third article, these two articles will form a co-citation relationship [38]. Now the research object of co-citation analysis has been extended from literature to authors and journals. Through co-citation analysis, we can have a more systematic grasp of the evolutionary relationship of knowledge structure, key journals and literature, and hot research topics in a particular field [39].

3.4.1. Highly Co-Cited Journals Analysis

Journal co-citation is a method to explore the association between journals through the external perception of journals. The relationship between journals and core journals can be determined using journal co-citation analysis [40]. In Figure 5, the node's size represents the journal's total co-citation number; the larger the node, the higher the total number of journal co-citations. Meanwhile, the connection's thickness and length between the two nodes also reflect the closeness of the co-citation relationship between journals. The thicker and shorter the connecting line, the more frequently the two journals are cited simultaneously, the closer the relationship, and the higher the similarity of the journals. So, from Figure 5, we can see that the red cluster has an enormous scale and contains 53 nodes, while the green group and blue collection have the following largest scale and contain 38 and 17 nodes, respectively. After analyzing and summarizing the journals included in the three clusters, we can find that most of the topics in the red cluster are related to polymers. Such as "Polymer Degradation and Stability", "Journal of Applied Polymer Science", "Polymers for Advanced Technologies", etc. At the same time, most of the journals in the blue cluster are related to electrochemistry and power sources, such as the "Journal of Power Sources", "Electrochimica Acta", "Journal of the Electrochemical Society", etc. While the journals in the green cluster are often in materials chemistry, such as "ACS Applied Materials & Interfaces", "Journal of Materials Chemistry A". This cluster also contains more energy-related journals, such as "Advanced Energy Materials", "Energy Storage Materials", and "Nano Energy".



NO.	Highly co-cited journals	Citation	Links	Cluster
1	Journal of Power Sources	4713	86	🔵
2	Polymer Degradation and Stability	3208	86	🔴
3	ACS Applied Materials & Interfaces	2845	86	🟢
4	Journal of Materials Chemistry A	2551	86	🟢
5	Electrochimica Acta	2383	86	🔵
6	Journal of the Electrochemical Society	2027	85	🔵
7	Chemical Engineering Journal	1860	86	🔴
8	Journal of Applied Polymer Science	1844	86	🔴
9	Advanced Materials	1837	86	🟢
10	RSC Advances	1556	86	🔴

Figure 5. High co-citation journals' distribution analysis of ILs in the flame retardancy field.

The table in Figure 5 shows the top 10 journals regarding co-citation frequency and their various parameters. Among them, the "Journal of Power Sources", "Polymer Degradation and Stability", and "ACS Applied Materials & Interfaces" were the top three journals in terms of co-citation frequency, their total citations are 4713, 3208, and 2845 times, respectively. Therefore, after a comprehensive analysis of the clustering as well as the high co-citation journals, we extracted "Chemical Engineering Journal", "Journal of Power Sources", "Polymer Degradation and Stability", and "ACS Applied Materials and Interfaces" as the core journals of ILs in the field of Chemical Engineering, power sources, polymer materials, and materials chemistry related to flame retardancy research.

3.4.2. Core Literature Analysis

In the initial bibliometric analysis, we used the extended dataset containing the most significant amount of literature (Data Set 3) as the literature data source. The reason for employing this dataset is to rely on the broader literature to provide a comprehensive picture of the overall research on ILs in the flame retardancy field. However, when combing the core literature in the IL flame retardancy field to extract the most accurate core literature with the best relevance, Data Set 1 was selected as the source for core literature analysis. Because citation frequency is a standard measure to judge the influence of literature, in this section, the top 15 articles ranked by total citations were selected as the core literature [41], and the results are shown in Table 3. In this table, it can be found that inter-institutional cooperation exists in 10 documents, but only three papers have inter-country cooperation relationships. This phenomenon means there is already sizeable cooperation in IL flame retardancy research, but this cooperation is still limited to institutions in the same country and has not yet formed a more extensive worldwide academic cooperative research relationship. Furthermore, from the perspective of the publication year and the citation number of core literature, among all core literature, the average ACY value of literature published within the last five years is significantly higher, at 13.91, while the average ACY value of all core literature is 10.14. From the viewpoint of application fields, the core literature can be divided into two categories: flame retardant in polymers and flame retardant in batteries. After further analysis, it can be found that among the literature with higher-than-average ACY values, the literature about polymers' flame retardants occupies most of the literature, and the literature about batteries' flame retardants occupies a relatively small percentage. Therefore, combining the above results, we speculate that flame retardants in polymers may be hotter than flame retardants in batteries in the recent research of ILs flame retardant applications.

It can be seen that the top cited paper in Table 3 is "Application of nonflammable electrolyte with room temperature ILs (RTILs) for lithium-ion cells" by Nakagawa et al. [9] published in "Journal of Power Sources" in 2007. This article has been cited 134 times, but its average annual citation frequency was low compared to the rest of the literature in Table 3, at 8.38 times. In this paper, the authors presented a new hybrid electrolyte composed of a flammable organic solvent and a nonflammable room-temperature IL (PP13-TFSI) to improve lithium batteries' safety. Experimental results showed that lithium-ion batteries using the hybrid electrolyte exhibit good discharge performance, and the mixed electrolyte is nonflammable at or above 40% IL content. Therefore, this nonflammable room-temperature IL has the potential to become a flame retardancy additive for organic electrolytes used in lithium batteries. Shi et al. [42], whose paper ranked second in total citations, designed a new phosphorus-containing halogen-free IL flame retardant ([Dmim]Tos) for epoxy resins. With the addition of 4 wt% [Dmim]Tos to the epoxy resin, the epoxy resin achieved a fire rating of UL-94 V-0. The value of the limiting oxygen index was increased to 32.5%, while the peak heat release rate was reduced by 37%. At the same time, adding this flame retardant can also solve traditional flame retardants' adverse effects on the epoxy resin's curing process, mechanical properties, and transparency. The literature with the third highest total citations was the paper by Kuo et al. [14]. Their team utilized the oligomeric IL mixed with PVdF-co-HFP and organic liquid electrolyte to obtain a gel polymer electrolyte with high ionic conductivity, good interfacial compatibility with battery electrodes, and non-flammability. This gel polymer electrolyte has an ultimate oxygen index of 29%, and its flame retardancy ability under air conditions can help improve the thermal safety of lithium-ion batteries.

Eight of the remaining 12 articles are related to the research of ILs in polymer flame retardation (e.g., epoxy resins). Chen et al. [43] constructed an efficient intumescent flame retardancy system for polypropylene by adding polyoxyvinyl IL (PIL) to the composite of conventional polypropylene and intumescent flame retardant (IFR). The experimental results showed that the composite could achieve UL-94 V-0 fire rating when 14.5 wt% of IFR and 0.5 wt% of PIL are added. When only a single IFR is added to the polypropylene material, a 25 wt% IFR addition is required to achieve this fire rating. Li et al. [13] prepared epoxy resin-based nanocomposites with excellent thermal conductivity and flame retardancy using non-covalent IL flame retardant-functionalized boron nitride nanosheets as additives. To address the interfacial incompatibility of the emerging black phosphorus nanosheets (BP) in fabricating high-performance polymer composites, Cai et al. [44] applied polymeric IL to link the BP nanosheets and thermoplastic polyurethane (TPU). Then the final TPU/IL-BP material showed a significant decrease in both the heat release rate and the peak of total heat release was obtained. Sonnier et al. [45] investigated the flame retardancy effect of ILs with different levels of quaternary phosphates on epoxy resins using thermogravimetric analysis, pyrolytic combustion flow calorimetry, and cone calorimetry. The results showed that the quaternary phosphate salt IL changed the epoxy resin's pyrolysis path and promoted the epoxy resin's carbon formation. Thus, flame suppression was achieved. Except for the literature related to polymer flame retardant, there are also three papers related to the study of ILs in the thermal safety of lithium-ion batteries. Guo et al. [46] prepared and studied a flexible composite IL gel polymer electrolyte with $Li_{1.5}A_{l0.5}Ge_{1.5}(PO_4)_3$ as a carrier, effectively combining good electrochemical properties and thermal safety. Kim et al. [47] designed a new binary electrolyte consisting of pyrroline-based IL and carbonate to improve the poor mobility of lithium-ions and battery safety during the lithium battery's charging/discharging process. Then the experiment showed that the lithium-ion conductivity is better in this binary electrolyte than in 100 wt%

IL or 100 wt% carbonate-based electrolytes, and the flammability of this electrolyte is significantly suppressed.

Table 3. Top 15 core articles in Data Set 1 in terms of total citation.

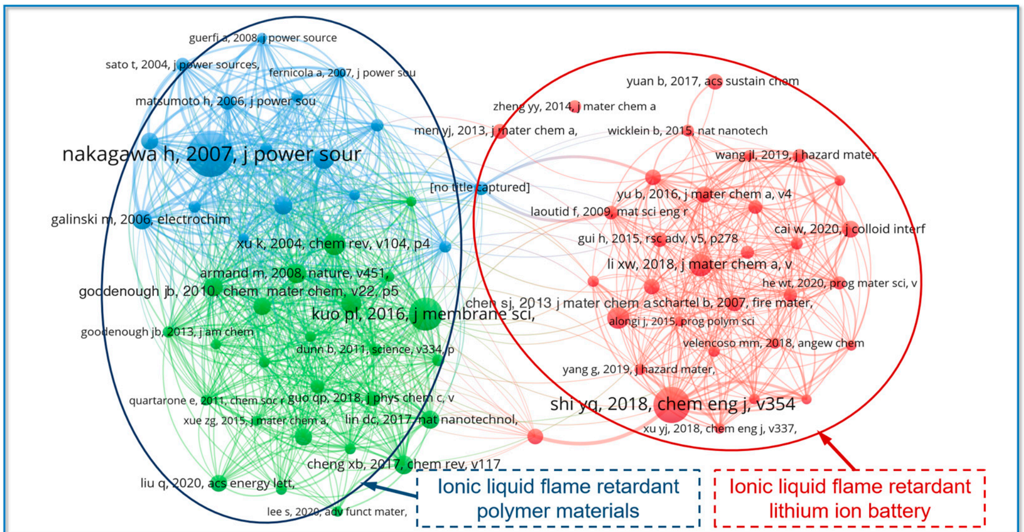
NO.	Title	Journal	Author	Year	IN	CN	ACY
1	Application of nonflammable electrolyte with room temperature ionic liquids (RTILs) for lithium-ion cells	Journal of Power Sources	Nakagawa et al. [9]	2007	2	1	8.38
2	Novel phosphorus-containing halogen-free ionic liquid toward fire safety epoxy resin with well-balanced comprehensive performance	Chemical Engineering Journal	Shi et al. [42]	2018	1	1	21.2
3	A new strategy for preparing oligomeric ionic liquid gel polymer electrolytes for high-performance and nonflammable lithium ion batteries	Journal of Membrane Science	Kuo et al. [14]	2016	1	1	13.71
4	Highly thermally conductive flame retardant epoxy nanocomposites with multifunctional ionic liquid flame retardant-functionalized boron nitride nanosheets	Journal of Materials Chemistry A	Li et al. [13]	2018	4	1	13.2
5	Increasing the efficiency of intumescent flame retardant polypropylene catalyzed by polyoxometalate-based ionic liquid	Journal of Materials Chemistry A	Chen et al. [43]	2013	2	1	6.4
6	Self-healing Janus interfaces for high-performance LAGP-based lithium metal batteries	ACS Energy Letters	Liu et al. [48]	2020	4	3	16.67
7	Self-assembly followed by radical polymerization of ionic liquid for interfacial engineering of black phosphorus nanosheets: Enhancing flame retardancy, toxic gas suppression and mechanical performance of polyurethane	Journal of Colloid and Interface Science	Cai et al. [44]	2020	4	3	16.67
8	Transparent cellulose-silica composite aerogels with excellent flame retardancy via an in situ sol-gel process	ACS Sustainable Chemistry & Engineering	Yuan et al. [49]	2017	3	1	7.83
9	Flame retardancy of phosphorus-containing ionic liquid-based epoxy networks	Polymer Degradation and Stability	Sonnier et al. [45]	2016	5	1	6.71
10	Low fractions of ionic liquid or poly(ionic liquid) can activate polysaccharide biomass into shaped, flexible and fire-retardant porous carbons	Journal of Materials Chemistry A	Men et al. [50]	2013	3	1	4.6
11	Synergistic effect of graphene and an ionic liquid containing phosphonium on the thermal stability and flame retardancy of polylactide	RSC Advances	Gui et al. [51]	2015	1	1	5.63
12	Flame retardant and stable Li1.5Al0.5Ge1.5(PO4)(3)-Supported ionic liquid gel polymer electrolytes for high safety rechargeable solid-state lithium metal batteries	Journal of Physical Chemistry C	Guo et al. [46]	2018	1	1	8.8
13	Synergy effect between quaternary phosphonium ionic liquid and ammonium polyphosphate toward flame retardant PLA with improved toughness	Composites Part B-Engineering	Jia et al. [52]	2020	1	1	13
14	Uniform nanoparticle coating of cellulose fibers during wet electrospinning	Journal of Materials Chemistry A	Zheng et al. [53]	2014	2	2	4.22
15	Pyrrolinium-based ionic liquid as a flame retardant for binary electrolytes of lithium ion batteries	ACS Sustainable Chemistry & Engineering	Kim et al. [47]	2016	3	1	5.14

IN: number of institutions; CN: Number of countries; ACY: Average Citations per Year.

3.4.3. Knowledge Base Analysis

Different scholars studying the flame retardant function of ILs cite highly relevant literature depending on the knowledge base involved. When the article is co-cited by two or more articles simultaneously, these three articles constitute a literature co-citation relationship. So, the literature co-citation relationship can reflect a research field's knowledge base and structure [54]. In this section, the literature with a citation frequency of fewer than 30 times was selected for the literature co-citation analysis, and then 74 nodes were obtained, forming three clusters in total, as shown in Figure 6. The node's size in the figure represents the citation frequency of the literature, and the larger the node, the more frequently it is cited. Moreover, the co-citation relationship between the literature was represented by the lines between the nodes.

From Figure 6, we can find thick lines between the green and blue clusters, while the lines between the red, green, and blue clusters are sparser and farther apart from the green and blue groups. This situation suggests that the literature in the green and blue clusters are similar and may have the same or similar knowledge base. On the other hand, the red group possesses a different knowledge base than the green and blue clusters. After analyzing the representative literature in these three clusters, we found that most of the literature in the green and blue clusters is related to ILs in the lithium-ion battery's flame retardancy. As in the paper by Nakagawa et al. [9], "Application of nonflammable electrolyte with room temperature ionic liquids (RTILs) for lithium-ion cells". In contrast, the literature in the red cluster may be related to ILs in polymer's flame retardancy, such as the paper by Shi et al. [42], "Novel phosphorus-containing halogen-free ionic liquid toward fire safety epoxy resin with well-balanced comprehensive performance". Therefore, based on the results of the literature co-citation analysis, we can divide the knowledge base into two parts: ILs in the lithium-ion battery's flame retardancy and ILs in the polymer's flame retardancy.



NO.	Highly co-cited journals
1	Application of nonflammable electrolyte with room temperature ionic liquids (RTILs) for lithium-ion cells
2	Novel phosphorus-containing halogen-free ionic liquid toward fire safety epoxy resin with well-balanced comprehensive performance
3	A new strategy for preparing oligomeric ionic liquid gel polymer electrolytes for high-performance and nonflammable lithium ion batteries
4	Highly thermally conductive flame retardant epoxy nanocomposites with multifunctional ionic liquid flame retardant-functionalized boron nitride nanosheets
5	Increasing the efficiency of intumescent flame retardant polypropylene catalyzed by polyoxometalate based ionic liquid
6	Nonaqueous liquid electrolytes for lithium-based rechargeable batteries
7	Ionic-liquid materials for the electrochemical challenges of the future
8	Issues and challenges facing rechargeable lithium batteries
9	Building better batteries
10	Ionic liquids as electrolytes

Figure 6. Total citations of the literature related to ILs in the flame retardancy field [9,13,14,36,42–44,46,48–51,53,55–77].

(1) ILs in the lithium-ion battery's flame retardancy: In recent years, high-capacity commercial lithium-ion batteries have begun to be widely used in ample power supplies. For this commercial lithium-ion battery, the carbonate-based electrolyte consisting of a lithium salt with a low flash point, highly flammable, and poorly electrochemically stable carbonate solvent is typically used [78]. So, when this kind of lithium-ion battery is misused, it is prone to thermal runaway, fire, and even explosion accidents. At the same time, ILs are composed of both anions and cations and appear as liquids at room temperature, and they are always characterized by very low volatility, non-flammability, wide electrochemical stability window, high solubility, and high thermal stability [79]. Therefore, ILs are suitable for preparing flame-retardant electrolytes in the lithium-ion battery. Wu et al. [80] synthesized a novel IL consisting of the symmetric tetra-butyl-phosphonium cation and the (non-fluorobutanesulfonyl)(trifluoromethanesulfonyl) imide anion. This new IL combines excellent flame retardancy, thermal stability, electrochemical stability, and good electrical conductivity at room temperature when used as an electrolyte in lithium-ion batteries. Kale et al. [81] obtained flexible solid polymer electrolytes with good thermal stability and flame retardancy using the environmentally friendly materials cellulose triacetate, PEGMA, and IL (Pyr14TFSI).

(2) ILs in the polymer's flame retardancy: The polymer's microscopic combustion mechanism can be generally considered a chain reaction mechanism. During the combustion process, free radicals such as H· and HO· are the key to maintaining the chain reaction of polymer combustion, and these free radicals' content determines the occurrence of polymer combustion and the speed of discharge [82,83]. Furthermore, when from the macroscopic point of view, the thermal decomposition of the polymer will produce a volatile combustible substance. These flammable substances will produce flame and combustion under proper oxygen concentration, temperature, and an ignition source. Therefore, as a substance that traps free radicals generated during polymer combustion, ILs can cause chain reactions to be terminated and absorb large amounts of heat and release non-flammable gases. In this way, ILs can reduce the oxygen concentration in the combustion system and achieve the flame retardancy effect. In addition, the structural designability of ILs can be used to introduce flame-retardant elements such as phosphorus and boron to the molecules, giving the material good flame retardancy through the condensed phase. This kind of ILs can make the material form a protective layer (such as a carbon layer) on its surface during combustion to prevent heat conduction and thus play a role in flame retardation [13,42,45]. Zhou et al. [84] synthesized a room-temperature IL tetrabutylphosphonium thiocyanate containing phosphorus and sulfur elements as a flame retardant in flexible polyurethane foams. This flame retardant was realized to enhance the flame retardancy of flexible polyurethane foam at low doses. Zhu et al. [85]adopted polymerization of vinyl tetrafluoroborate IL with polydivinylbenzene to synthesize a porous material with good thermal insulation and flame retardancy. This material has a low peak heat release rate during combustion and exhibits self-extinguishing behavior in the torch burn test without molten dripping.

3.5. Research Flow, Hot Spots and Frontiers Analysis

3.5.1. Key Literature's Research Flow Analysis

HistCite software provides a method to sort out the citation lineage of classical literature and identify the flow of research through the key literature citation sequential chart [86]. After importing all the literature data from Data Set 3 into this software, we selected LCS as the reference index and set the number of key literature analyzed to 50. Then we obtained the key literature citation sequential figure, as shown in Figure 7, and Table 4 summarized the information of the top 20 LCS values in the key literature citation sequential figure. The value of LCS refers to the frequency of literature cited in the current Data Set 3 database. It can reflect the influence and authority of the literature in a field [21], and the value is represented by the circle size in Figure 7, while the connecting lines with arrows in the figure indicate the citation relationship between the literature. According to the citation network and the literature content in Figure 7, we can classify the research flow of key literature on ILs in the flame retardancy field into two parts. They are the flow of research related to the flame retardancy subfield of lithium-ion batteries and the research related to the flame retardancy subfield of polymer materials.

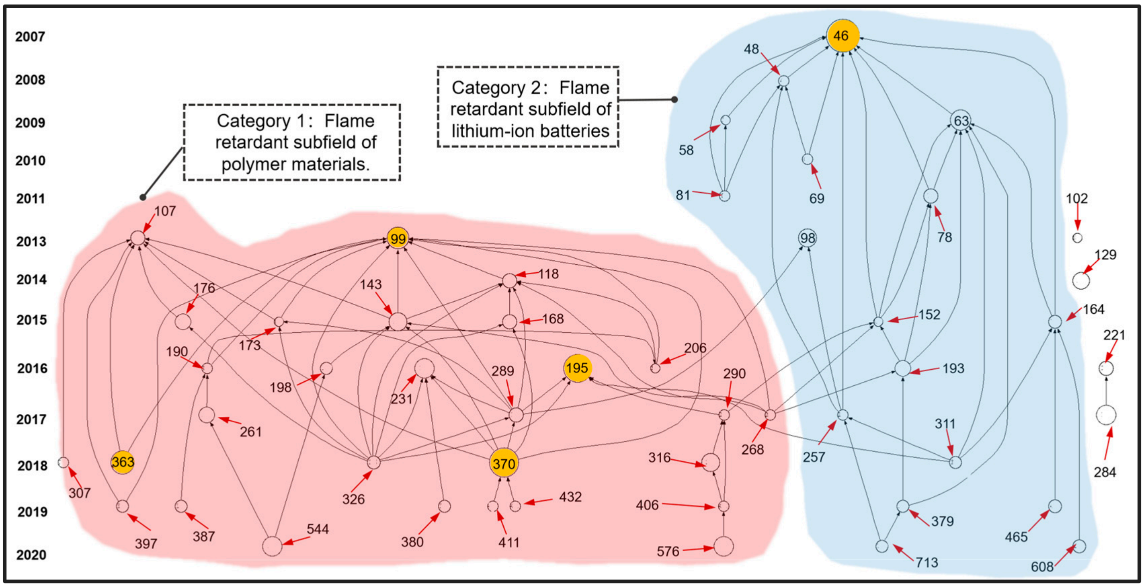


Figure 7. The key literature citation sequential figure for ILs in the flame retardancy field.

(1) The flow of research related to the flame retardancy subfield of polymer materials: In this subfield, the literature No. 99, 195, 363, and 370 have high LCS values and are essential literature in the development of polymer materials flame retardancy research. Among them, No. 99 was published in the earliest year and can be regarded as the pioneering literature on ILs in polymer materials' subfield of flame retardancy. In that paper, Chen et al. [43] constructed an efficient intumescent flame retardancy system for polypropylene by adding polyoxyethylene-based IL to a conventional PP/IFR composite. Literature Nos. 363, 370, and 99 have a strong research succession relationship. Among them, literature No. 363 produced epoxy resin-based nanocomposites with excellent thermal conductivity and flame retardancy using non-covalent IL flame retardant-functionalized boron nitride nanosheets as additives [13]. Literature No. 370, on the other hand, designed a novel phosphorus-containing halogen-free IL flame retardant [42]. The linkage of literature No.195 bridges the subfield of flame retardancy for lithium-ion batteries and flame retardancy for polymeric materials. The results of this literature have given a solid impetus for related research in both subfields around 2018. In that literature, the research team prepared a high ionic conductivity, nonflammable gel polymer electrolyte using oligomeric ILs synthesized from phenolic epoxy resins to mix with PVDF-co-HFP and organic liquid electrolytes [14]. Notably, that literature No. 326 cites the vast majority of the literature in the subfield of polymeric materials' flame retardancy in Figure 7, so we believe this literature is an excellent integration of the previous research base. In that literature, Xiao et al. synthesized a new multifunctional IL-based metal-organic hybrid for efficient flame retardation of epoxy resins [87]. In addition, a series of general-sized circles emerged under this subfield during 2018–2020, such as those representing literature Nos. 380, 397, 411, and 544. The study in literature No. 397 showed that HGM@ [EOOEMIm][BF_4]-based thermoplastic polyurethane composites prepared by IL ([EOOEMIm][BF_4]) modified hollow glass beads (HGM) have good flame retardancy and thermal stability [88].

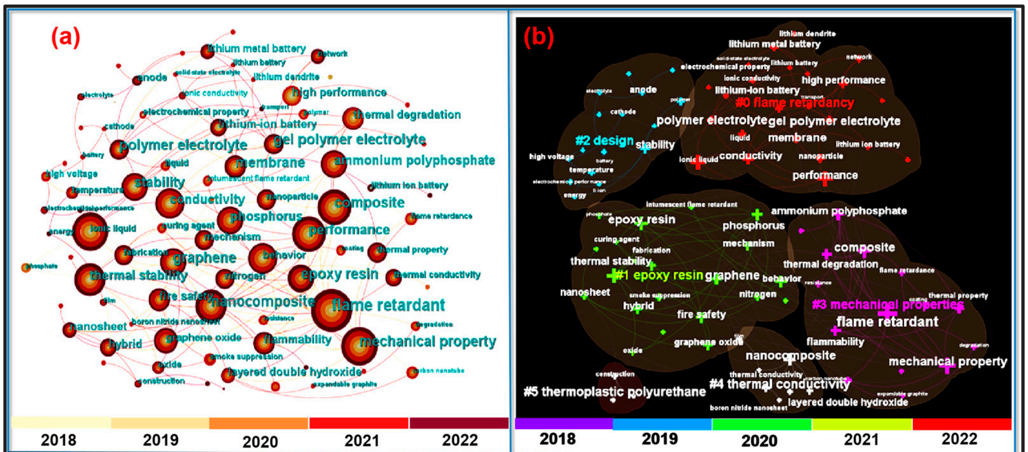
(2) The flow of research related to the flame retardancy subfield of lithium-ion batteries: In the research related to this subfield, the paper published by Nakagawa et al. is at the first position in the time series, with the LCS value of this paper reaching 132, which is also at the first position among all the literature. Their paper proposed a new hybrid electrolyte consisting of a flammable organic solvent and a nonflammable room-temperature IL to improve lithium batteries' safety [9]. Based on the paper's publication year and LCS value, we regard this paper as one of the most important and influential papers in the history of ILs in the flame retardancy subfield of lithium-ion batteries. The LCS value of the paper No. 63 by Lewandowski et al. is also at a high level, and there is a citation relationship between this paper and the paper by Nakagawa et al. Therefore, the paper by Lewandowski et al. can be considered another vital literature appearing in this subfield after Nakagawa et al. In this paper, Lewandowski et al. reviewed the properties of room-temperature ILs as electrolytes for lithium-ion batteries [89]. Literature No. 193 connects another subfield with a more complex citation relationship with the rest of the literature. In that paper, Kim et al. [47] designed new binary electrolytes consisting of pyrroline-based IL and carbonates to improve poor lithium-ion mobility and lithium-ion battery thermal safety during the charge/discharge. Furthermore, from 2018 to 2020, there are more general-sized circles in this subfield, such as literature Nos. 379, 608, and 713, which Chinese scholars mostly publish. Combined with China's recent positive research dynamics in this subfield, these papers may become crucial nodes in the future lithium-ion batteries flame retardant subfield. Among them, literature Nos. 379 and 713 are both review papers. In literature 379, Wang et al. reviewed commonly adopted Ils additives and composite flame retardancy additives in optimizing electrolytes to improve the thermal safety of lithium-ion batteries and their flame retardant mechanisms [90]. Meanwhile, in literature No. 713, Tian et al. reviewed recent advances in the design of safe electrolytes for lithium-ion batteries. These advances include the addition of flame retardancy additives, overcharge additives, solid-state electrolytes, IL electrolytes, and thermosensitive electrolytes. As for literature No. 608, the authors introduced the IL Pyr13FSI into the hybrid network to obtain a series of gel polymer electrolytes with excellent flame retardancy, thermal stability, and electrochemical stability [91].

Table 4. Information on the top 20 LCS values in the key literature citation sequential figure.

No.	Author	Year	LCS	No.	Author	Year	LCS
46	Nakagawa et al. [9]	2007	132	231	Sonnier et al. [45]	2016	46
48	Seki et al. [92]	2008	17	257	Watanabe et al. [93]	2017	17
58	Profatilova et al. [94]	2009	12	261	Shi et al. [95]	2017	30
63	Lewandowski et al. [89]	2009	52	268	Chen et al. [96]	2017	17
69	Xiang et al. [97]	2010	16	284	Yuan et al. [49]	2017	46
78	Arbizzani et al. [98]	2011	29	289	Xiao et al. [99]	2017	29
81	Nadherna et al. [100]	2011	14	290	Guo et al. [101]	2017	14
98	Men et al. [50]	2013	44	307	Jiang et al. [102]	2018	16
99	Chen et al. [43]	2013	63	311	Yang et al. [6]	2018	19
102	Kivotidi et al. [103]	2013	13	316	Guo et al. [46]	2018	44
107	Yang et al. [104]	2013	28	326	Xiao et al. [87]	2018	24
118	Chen et al. [105]	2014	26	363	Li et al. [13]	2018	66
129	Zheng et al. [53]	2014	36	370	Shi et al. [42]	2018	105
143	Gui et al. [51]	2015	44	379	Wang et al. [90]	2019	20
152	Wilken et al. [106]	2015	13	380	Huo et al. [107]	2019	18
164	Kalhoff et al. [108]	2015	24	387	Bentis et al. [109]	2019	20
168	Tan et al. [110]	2015	27	397	Jiao et al. [88]	2019	18
173	Hu et al. [111]	2015	12	406	Yu et al. [112]	2019	15
176	Alongi et al. [74]	2015	30	411	Wang et al. [113]	2019	15
190	Chen et al. [114]	2016	17	432	Jian et al. [115]	2019	16
193	Kim et al. [47]	2016	34	465	Bose et al. [116]	2019	22
195	Kuo et al. [14]	95	2016	544	Cai et al. [44]	2020	50
198	Huang et al. [117]	19	2016	576	Liu et al. [48]	2020	49
206	Chen et al. [118]	12	2016	608	Li et al. [91]	2020	22
221	Yuan et al. [119]	28	2016	713	Tian et al. [120]	2020	20

3.5.2. Research Hotspots Analysis

To inspect the current research hotspots in the field of Ils in flame retardancy, the Citespace software was used to perform a keyword co-occurrence analysis of the literature published in the last five years [121]. In this software, one year as 1-time slicing was set, then the hot study interval was divided into five partitions, and the minimum spanning tree for network pruning was selected. Then, after filtering out the keywords with co-occurrence frequency in the top 4%, we finally obtained a keyword co-occurrence map with 87 nodes, 185 links, and a network density of 0.0495, as shown in Figure 8a. Meanwhile, in Figure 8a, the node's size represents the frequency of keyword appearances, and the larger the node, the more frequently the keyword appears. The node color represents the co-occurrence time, and the darker the color indicates the later the co-occurrence time. The centrality, on the other hand, can be used to measure whether the node assumes the role of a hub within the research domain (Centrality >0.1 is considered to be more central), thus helping to infer research hotspots [122,123]. So, by combining the tables in Figure 8, we can find that keywords such as flame retardant, mechanical property, performance, composite, epoxy resin, nanocomposite, and conductivity have a high frequency and centrality. While Figure 8b shows the six clusters formed by these 87 nodes, the Q-value of this cluster map is 0.05336, and S-value is 0.8361. Based on this Q-value and S-value, we can assume that the cluster structure is significant and its clustering information is valid [124]. The four larger clusters among these six clusters are #0 clusters (flame retardancy), #1 clusters (epoxy resin), #2 clusters (design), and #3 clusters (mechanical properties). Finally, based on the clustering level, frequency of keyword occurrence, and centrality, we divided the research hotspots of Ils in flame retardancy in recent years into three major categories. Namely, they are the research of Ils in lithium-ion battery electrolytes' flame retardancy, Ils in the polymer materials' flame retardancy, and Ils in improving materials' multiple properties during flame retardancy.



NO.	Keyword	Count	Centrality	Year
1	flame retardant	305	0.40	2018
2	mechanical property	195	0.13	2018
3	performance	184	0.26	2018
4	ionic liquid	160	0.07	2018
5	thermal stability	148	0.08	2018
6	composite	132	0.27	2018
7	epoxy resin	120	0.12	2019
8	nanocomposite	113	0.21	2018
9	behavior	100	0.03	2018
10	conductivity	89	0.18	2018

Network: N=87, E=185, (Density=0.0495)

Modularity Q=0.5336

Weighted Mean Silhouette S=0.8361

Harmonic Mean (Q, S)=0.6514

Figure 8. (**a**) High-frequency keywords for Ils in the flame retardancy field in the past 5 years, (**b**) high-frequency keywords' clustering for Ils in the flame retardancy field in the past 5 years.

(1) The research of Ils in lithium-ion battery electrolytes' flame retardancy: In Figure 8b, the larger nodes included in the #0 cluster are performance, conductivity, polymer electrolyte, gel polymer electrolyte, high performance, lithium-ion battery, etc. At the same time, the larger nodes included in the #2 cluster are stability, anode, cathode, etc. Therefore, we believe that the research of Ils in lithium-ion battery electrolytes' flame retardancy is one of the current research hotspots in the field of Ils in flame retardancy. According to the larger scale nodes: gel polymer electrolyte, polymer electrolyte, conductivity, and other words, we can delineate two popular research directions of Ils in studying the electrolyte's flame retardancy for lithium-ion batteries. These two directions are improving and applying pure IL electrolytes and researching gel polymer electrolytes. In the improvement and application of pure IL, common electrolytes include imidazole-based IL electrolytes, quaternary ammonium salt-based liquid electrolytes, and pyrrole-based and piperidine-based. However, these pure IL electrolytes often have high viscosity and poor compatibility with negative electrodes [125]. Therefore, scholars have made a series of improvements to address these issues. For example, when Guerfi et al. applied pure imidazolium-based ILs with FSI- anions as electrolytes for lithium-ion batteries, they found that this electrolyte could show better compatibility with graphite cathodes [126]. On the other hand, Lee et al. [68] introduced a locally concentrated IL electrolyte with a non-solvating, fire-retardant hydrofluoroether. It can be used to improve the problems of high viscosity and poor ion transport of IL electrolytes in concentrated lithium salts. Another popular research direction, gel polymer electrolyte, refers to combining IL with a gel polymer matrix. It can avoid the problem of battery leakage caused by liquid ILs. For example, Kuo et al. [14] used oligomeric IL mixed with PVdF-co-HFP and organic liquid electrolytes to prepare a high-performance, non-flammable gel polymer membrane. Li et al. [91] introduced the IL (Pyr13FSI) into the hybrid network and obtained gel polymer electrolytes with excellent flame retardancy, thermal stability, and electrochemical stability. Liu et al. [127] generated polymer electrolytes with flame retardancy and good mechanical properties by doping lithium salts and ILs in an ABA triblock copolymer.

(2) The research of ILs in the polymer materials' flame retardancy: The larger scale nodes included in cluster #1 are thermal stability, epoxy resin, graphene, phosphorus, etc. Therefore, we can assume that the research of ILs in polymer materials' flame retardancy has become a research hotspot for ILs in flame retardancy. Based on the larger nodes, we can divide this research hotspot into two directions: Applying ILs to enhance the polymer materials' flame retardancy properties and utilizing ILs with inorganic materials to synergize flame retardancy polymers. In applying ILs to enhance the polymer materials' flame retardancy properties, more scholars have focused on the effect of ILs on the flame retardancy properties of epoxy resins [13,42,45]. Xiao et al. [99] synthesized a phosphate-based IL and added it to an epoxy resin. When the mass fraction of this IL in the epoxy resin reached 4 wt%, the fire rating of the epoxy resin would have UL-94 V-0 class. The limiting oxygen index value will also rise from 25.9% in the pure epoxy resin to 34.9%. Bi et al. [128] prepared a novel IL-modified flake material, after blending 6% of this material into the epoxy resin, the epoxy resin's limiting oxygen index value increased from 24.4% to 30.3%. At the same time, the peak heat release rate, total heat release rate, and maximum release rate of CO also decreased. In the study of utilizing ILs with inorganic materials to synergize flame retardant polymers, ILs often interact with inorganic materials through chemical bonding, electrostatic adsorption, hydrogen bonding, π-π stacking, etc. For instance, ILs are usually bonded with inorganic materials such as boron nitride, ammonium polyphosphate, graphene, and carbon nanotubes [13]. Feng et al. [129] prepared epoxy matrix nanocomposites using IL-modified hydroxylated boron nitride nanosheets to enhance the fire safety of epoxy resins. Jia et al. [52] added the IL tetrabutylphosphonium tetrafluoroborate into poly(lactic acid) as a synergist for ammonium polyphosphate. Then they found due to the synergistic effect between the IL and ammonium polyphosphate, it can obtain good flame retardant properties when only a low mass fraction of IL and ammonium polyphosphate is added to poly(lactic acid). Gao et al. [130] prepared IL ([BMIM] PF6) functionalized phosphorus-containing graphene oxide (ILGO) from graphene oxide, and introduced it into flexible polyurethane foam. The experiment result suggested that ILGO can effectively inhibit the release of heat, smoke, and toxic gases during the combustion of flexible polyurethane foam.

(3) The research of ILs in improving materials' multiple properties during flame retardancy: Cluster #3 contains nodes of larger size: flame retardant, mechanical property, composite, ammonium polyphosphate, etc. Therefore, we can assume that the research hotspot reflected by this clustering is the research of ILs in improving materials' multiple properties during flame retardancy. According to the literature, ILs also improve material's other properties when applied for flame retardation in high polymers and lithium-ion batteries. For instance, high polymer thermosetting, thermal conductivity, mechanical properties, soot suppression during combustion, and room temperature conductivity of lithium-ion battery electrolytes [13,14]. This improvement in the material's other properties increases the prospect of ILs research and application in flame retardancy. Yang et al. [104] developed a new intumescent flame retardancy system consisting of phosphorus-containing IL ([PCMIM] Cl) and ammonium polyphosphate to enhance the flame retardancy and drip resistance of polypropylene. In their experiment, the polypropylene material also showed sound processing and mechanical properties after using the new intumescent flame retardancy system. Czlonka et al. [131] obtained tough polyurethane materials with better flame retardancy properties and better compression strength by adding different mass fractions of melamine, silica, and IL ([EMIM] Cl) to rigid polyurethane foams. Bose et al. [116] prepared a nanocomposite membrane by combining IL-functionalized ZnS nanoparticles onto PVDF-HFP copolymers. The gel polymer electrolytes made from this nanocomposite membrane have high conductivity and lithium-ion mobility numbers at room temperature.

3.5.3. Exploring the Frontiers of Research

The burst term analysis can detect highly active research topics in the field and indicate emerging research trends and currents. In this section, we will analyze the literature in Data Set 3 by combining the development stages of ILs flame retardant research with the blast word analysis, and the results are shown in Figure 9. Among them, the stages of ILs flame retardant research are divided based on the literature numbers published annually. In Data Set 3, the annual number of published literature on ILs flame retardant was 0 before 2007. Between 2007 and 2011, the annual number of published literature did not exceed 3 at most, while between 2012 and 2017, the annual number of published literature ranged from 10 to 100, and after 2017, the annual number of published literature reached more than 100. Therefore, we divided the ILs flame retardant research into three stages, namely the first stage (2007–2011), the second stage (2012–2017), and the third stage 3 (2018-present).

In Figure 9, we listed the ten burst terms with the most outburst intensity. The arrow's starting position in this figure is the year the keyword began to burst, and the position pointed by the arrow is the year when the keyword ended burst. According to Figure 9, we can observe that the burst terms IL and graphite in the first stage continued to burst for a long time, spanning two stages, while the burst term Li/Lico2 cell was only one year. More burst terms emerge within the second stage, including thermal degradation, system, carbon nanotube, high performance, and challenge. However, the overall duration of these burst terms is not prolonged, and they do not cross two stages. The burst terms in the third stage are phosphate and smoke suppression, and they may represent the research frontiers and future trends of ILs in flame retardancy. After reviewing the relevant literature, we summarized these research frontiers and future trends in studies of ILs for smoke suppression and phosphorus-containing ILs for flame retardants.

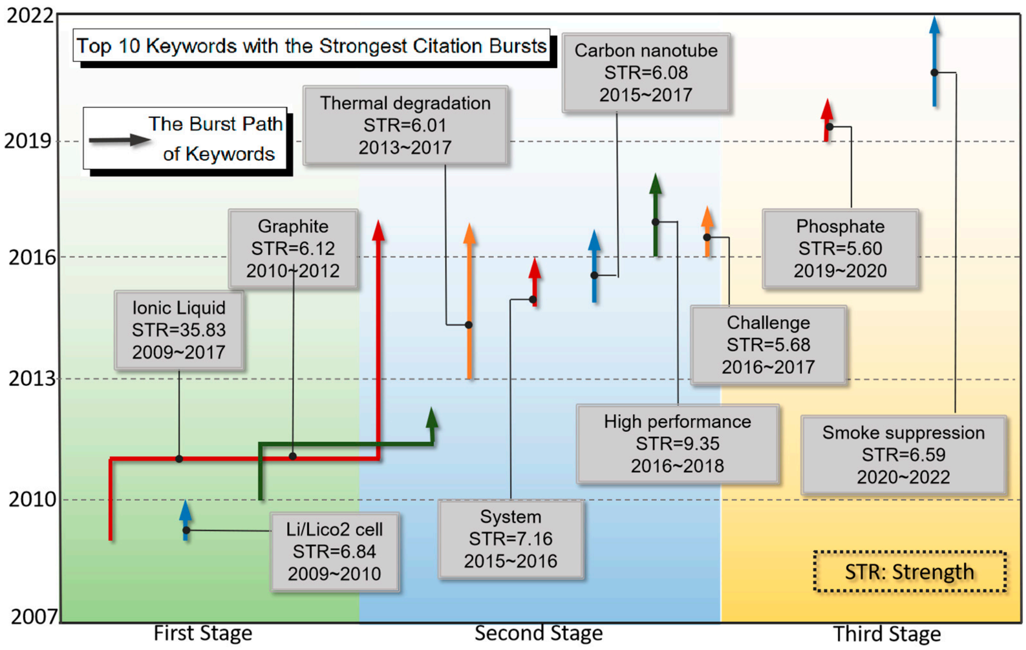


Figure 9. Keyword burst path for ILs in the flame retardant field.

(1) Combination of computational quantum chemistry to study the flame retardancy mechanism of ILs: In the past, the study of the flame retardancy mechanism of ILs in high polymers and lithium-ion battery electrolytes was commonly achieved through macroscopic experiments. For example, combining thermal analysis experiments and kinetic theory to infer the mechanism of flame retardancy through materials' thermal behavior and combustion properties [132]. Liu et al. [133] investigated the effect of cyano ILs on the flame retardant melamine's thermal behavior by applying a simultaneous thermogravimetric analyzer. In their paper, the model-free method and the Coats-Redfern method were performed to derive the decomposition mechanism of melamine mixtures with ILs. However, although macroscopic experiments can explain the flame retardancy mechanism to a certain extent, they are susceptible to errors, instrumentation, and experimental design, and the study of the mechanism is often not accurate and thorough enough. So, to remedy these shortcomings, studies combining computational quantum chemistry and macroscopic experiments to reveal the flame retardancy mechanism have emerged in recent years. This approach will likely become the frontier of research on ILs in flame retardancy. Meng et al. [134] reported on the thermal stability and decomposition kinetics of 1-alkyl-2,3-dimethylimidazole nitrate ILs with different alkyl chains through the help of thermal analysis kinetic theory and density functional theory calculations. Li et al. [135] studied the thermal decomposition products and properties of the IL ([Bmim][DBP]) using gas chromatography-mass spectrometry and Fourier infrared spectroscopy, and they also calculated the main bond energy energies of [Bmim][DBP] through a density functional theory approach. Li et al. [136] presented the inhibition effect of imidazolium-based ionic liquids on pyrophoricity of FeS.

(2) Research on fluorinated electrolytes: Fluorinated electrolytes exhibit good oxidation resistance, high ionic conductivity, and superior flame retardancy properties, and they are the preferred materials for manufacturing high energy density and high safety lithium-ion batteries [137]. Thus, integrating ILs with fluorinated electrolytes may become a research frontier for ILs in lithium-ion battery flame retardancy. Hou et al. [138] demonstrated that fluorinated flame retardancy electrolytes could reduce flammability in lithium-ion batteries and disclosed the different pathways of thermal runaway in lithium-ion batteries with fluorinated flame retardant electrolytes versus lithium-ion batteries with conventional electrolytes. Mei et al. [139] proposed a method for synthesizing fluorine-containing room temperature ILs (FRTILs) and investigated the potential of FRTILs as electrolyte solvents for lithium-sulfur batteries, with the results showing excellent thermal stability of FRTILs.

(3) ILs for smoke suppression during the material combustion process: More polymer materials itself is not only highly combustible, but in the combustion will also emit a large volume of toxic smoke containing CO, NO, and other substances [44], easy to cause environmental pollution and pose a threat to human health. The introduction of ILs in polymers can not only play a role in flame retardation but also inhibit the generation of smoke during combustion [87], which aligns with the concept of environmental protection in polymer applications. So, exploring the smoke suppression function of ILs in the material combustion process will also become a research frontier of ILs in the flame retardancy field. Huang et al. [140] designed a new metal-organic frameworks (MOF) composite for flame retardancy through the synergistic interaction of MOFs and phosphorus-containing nitrogen ILs. The ILs in this material can be available to trap free radicals during polymer pyrolysis and reduce smoke emissions during combustion. Chen et al. [114] investigated the IL ([Emim]PF_6) and aluminum hypophosphite's synergistic flame retardancy effect and smoke suppression properties on thermoplastic polyurethanes. The smoke density test revealed that the smoke generation during the thermoplastic polyurethane's combustion reached the lowest value at a [Emim]PF6 content of 0.0625 wt% and an aluminum hypophosphite content of 19.9375 wt%.

(4) Research on phosphorus-containing ILs for flame retardants: ILs are highly flexible in design and have a wide range of species, so phosphorus, nitrogen, boron, halogens, and other flame retardancy elements can be introduced into the anion or cation of ILs. These functionalized ILs can exert excellent flame retardancy ability during the combustion of materials. Based on the results of burst term analysis, the study on phosphorus-containing ILs for flame retardants will likely become a research hotspot for ILs in the flame retardancy field. Jiang et al. [132] analyzed the flame retardancy effect of phosphorus-containing ILs ([Bmim] [DBP]) on epoxy resins of flammable materials with the help of limit oxygen index and vertical combustion experiments. Li et al. [11] synthesized an imidazolium-based polyionic liquid (PDVE [DEP]) containing phosphate anion to improve PLA's flame retardancy. The limit oxygen index, vertical combustion, calorimetric, and thermogravimetric tests exhibited that the PDVE [DEP] improved the PLA's flame retardancy by melting-dissociation mode.

Although burst term analysis pointed out the above four research frontiers and future research trends, it was also noted during our review of other literature that machine learning may become an important direction in future ILs flame retardancy research [141]. Machine learning is a method in which a computer automatically discovers patterns in the data by analyzing and learning from the training data, and makes predictions and decisions based on these patterns [142]. At present, machine learning has begun to play a great advantage in the fields of flame retardant design [143], fire engineering, and flame combustion [144,145], but in the field of ILs flame retardancy, research on machine learning is still very scarce. Chen et al. [146] investigated the relationship between the organic phosphorus-containing flame retardants' structure and the addition amount with the epoxy resins' flame retardant performance by machine learning. Dhakal and Shah [147], on the other hand, used an artificial neural network model to predict the ionic conductivity of 1102 ionic liquids formed by every possible combination of 29 cations and 38 anions in the NIST ILThermo database and found that the model was able to accurately predict the conductivity of several ionic liquid mixtures. Due to the good designability of ILs, the machine learning-assisted design of ILs for flame retardancy electrolytes or polymers will likely be an important direction for future research. The assisted design will probably be based on a large amount of combustion experimental data and different algorithms to predict the flame retardant performance indexes of ILs, such as ignition time, peak heat release rate, flame retardant index, etc., and then select the optimal ILs flame retardant.

4. Conclusions

This paper applied bibliometric mapping methods to analyze the literature on ILs in the field of flame retardancy in SCIE and SSCI databases from 2000–2022. The main research contents include the analysis of academic collaborations, the current state of research and knowledge base, and the research flow, hot spots, and frontiers. The conclusion details are as follows.

(1) Scholars from these institutions have formed a large-scale academic collaboration. There is also a multidisciplinary intersection in ILs in flame retardancy research. Much of the research is centered on materials science and integrates environmental protection and sustainable development. Chemical Engineering Journal, Journal of Applied Polymer Science, and Journal of Thermal Analysis and Calorimetry are the journals with the highest number of articles on ILs in flame retardancy.

(2) All co-cited journals can be broadly classified into three major categories: polymer materials, power sources, and materials chemistry. Journal of Power Sources, Polymer Degradation and Stability, ACS Applied Materials & Interfaces, and Chemical Engineering Journal are core journals in these major categories. In addition, ILs flame-retardant lithium-ion batteries and polymer materials are the knowledge base of IL in flame retardancy research.

(3) The research hotspots of ILs in flame retardancy can be divided into three major categories. Namely, the research of ILs in lithium-ion battery electrolytes' flame retardancy, ILs in the polymer materials' flame retardancy, and the research of ILs in improving materials' multiple properties during flame retardancy. Among them, the improvement and application of pure IL electrolytes and the research of gel polymer electrolytes are two hot directions in the flame retardancy of ILs in lithium-ion battery electrolytes. Correspondingly, applying ILs to enhance the polymer materials' flame retardancy properties and utilizing ILs with inorganic materials to synergize flame retardant polymers are two hot research directions in polymer materials' flame retardancy of ILs.

(4) The combination of computational quantum chemistry to study the flame retardancy mechanism of ILs, the study of fluorinated electrolytes, ILs for smoke suppression, phosphorus-containing ILs for flame retardants, and machine learning-assisted design of ILs flame retardants are the research frontiers and future research trends.

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