Review Article

Polyethylene Terephthalate-Based Materials for Lithium-Ion Battery Separator Applications: A Review Based on Knowledge Domain Analysis

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As the key material of lithium battery, separator plays an important role in isolating electrons, preventing direct contact between anode and cathode, and allowing free passage of lithium ions in the electrolyte. Polyethylene terephthalate (PET) has excellent mechanical, thermodynamic, and electrical insulation properties. This review aims to identify the research progress and development trends of PET-based material for separator application. We retrieved published papers (2004–2019) from the Scientific Citation Index Expanded (SCIE) database of the WoS with a topic search related to PET-based material for separator application. The research progress and development trends were analyzed based on the CiteSpace software of text mining and visualization.

1. Introduction

With the increasing global energy crisis and environmental problems, it is becoming a trend for renewable energy to replace fossil fuels. The storage and use of renewable energy are inseparable from the development of the chemical power. In various types of chemical power supply system, lithium-ion battery has become the most popular secondary battery because of its high voltage, high specific energy, long life, and other advantages [1–6]. Lithium-ion battery is mainly composed of anode, cathode, separator, and electrolyte. When charging, Li+ is separated from the anode, passes through the separator in the electrolyte, and inserts into the cathode lattice. At this time, the anode is in the lithium poor state, and the cathode is in the lithium-rich state. During the discharge process, Li+ is released from the lithium-rich anode again, passes through the separator in the electrolyte to reach the lithium poor cathode, and inserts into the anode lattice [7–11]. At this time, the anode is in the lithium-rich state, and the cathode is in the lithium poor state. In order to keep the charge balance, Li+ migrates between the anode and cathode during the charge and discharge process. At the same time, the same number of electrons moves back and forth in the external circuit to form a current. As the key material of lithium batteries, separator plays an important role in isolating electrons, preventing direct contact between anode and cathode, and allowing free passage of lithium ions in the electrolyte. At the same time, the separator plays an important role in ensuring the safe operation of the battery.

In special cases, the separator will be partially damaged, resulting in direct contact between the anode and cathode, which will lead to severe battery reaction and lead to battery fire and explosion [12–15]. Therefore, to improve the safety of lithium-ion battery and ensure the safe and stable operation of the battery, the separator must meet the following conditions: (1) chemical stability: no reaction with electrolyte and electrode materials; (2) wettability: easy to soak with electrolyte and no elongation or shrinkage; (3) thermal
stability: high-temperature resistance, high fusing isolation; (4) mechanical properties: good tensile strength to ensure that the strength and width of automatic winding remain unchanged; (5) porosity: high porosity to meet the requirements of ionic conductivity.

At present, the commercial lithium battery separators in the market are mainly polyethylene- (PE-) and polypropylene- (PP-) based microporous polyolefin separators [16]. This kind of separator is widely used in lithium battery separator because of its low cost, good mechanical properties, excellent chemical stability, and electrochemical stability. PE composite separator mainly includes PP/PE composite separator and PP/PE/PP composite separator. PE separator has good flexibility but low melting point. PP separator has good mechanical properties and high melting point (165°C). The combination of the two makes the composite separator have the advantages of low closed cell temperature and high fusing temperature. Moreover, the outer PP membrane has an antioxidant effect, so the cycle performance and safety performance of this kind of separator have been improved to a certain extent, which makes it widely used in the field of power battery.

In recent years, on the one hand, the strong demand of 3C industry and new energy automobile industry for high-performance secondary batteries has promoted the rapid development of separator production technology. On the other hand, to further improve the specific energy and safety of lithium-ion batteries, researchers have developed many new lithium battery separators. Due to the hydrophobic surface and low surface of polyolefin material, the poor wettability of the separator with the electrolyte is poor, which affects the cycle life of battery. In addition, due to the low thermal deformation temperature of PE and PP (80–85°C for PE and 100°C for PP), the separator will suffer severe thermal shrinkage when the temperature is too high, so this kind of separator is not suitable for use in high-temperature environment. The traditional polyolefin separator cannot meet the requirements of 3C products and power batteries. With the development of lithium-ion battery technology, researchers have developed a variety of new lithium battery separator materials based on the traditional polyolefin separator.

Polyethylene terephthalate (PET) has excellent mechanical, thermodynamic, and electrical insulation properties. The most representative product of PET separator is the composite film with ceramic particles coated on PET membrane substrate developed by Degussa company of Germany. It shows excellent heat resistance, and the closed cell temperature is as high as 220°C. Xiao et al. [17] prepared PET nanofiber separator by electrospinning method. The product has three-dimensional porous network structure. The product has a smooth surface with an average diameter of the fiber of 300 nm. The melting point of electrospun PET separator is much higher than that of PE separator with a maximum tensile strength of 12 MPa and a porosity of 89%, which is much higher than the Celgard separator on the market. The ionic conductivity reached $2.27 \times 10^{-3} \text{s/cm}$ with an excellent cycle performance. The porous fiber structure of PET separator remained stable after 50 cycles of battery cycle.

Thus far, scientific research on PET-based materials for separator application has mainly focused on composite preparation, characterization, and performance analysis. Some of these areas have been studied for a long time; on the other hand, some areas of research have just begun. In this study, we aim to identify the research progress and development trends of PET-based materials for separator application. We retrieved published papers (2004–2019) from Scientific Citation Index Expanded (SCIE) database of the WoS with a topic search related to PET-based materials for separator applications. However, in the face of the substantial amount of literature, there are certain limitations, subjectivity, and one-sidedness in the analysis by reading and induction. The application of modern scientometrics and information metrology technology can carry out multivariate and diachronic dynamic analysis of massive literature data. Therefore, in this work, the research progress and development trends were analyzed based on the CiteSpace software of text mining and visualization.

2. Materials and Methods

CiteSpace is a document data mining and visualization software developed by Chen’s team [18, 19]. It combines cluster analysis and social network analysis. It can analyze the basic knowledge and research frontiers, research characteristics, and evolution trends of a certain field through the cocitation and coupling of documents, scientific research, cooperation networks, and theme contributions [20–35]. The CiteSpace used in this analysis is 5.7R2. The time span is 2004–2019 (Slice Length = 1). This work selects the academic articles which contain the search of “PET” and “battery separator”, and a total of 78 articles are obtained after screening [36–100].

3. Results and Discussion

3.1. Characteristics of Publication Output. According to the annual distribution statistics analysis (Figure 1), a preliminary understanding of the history of research progress for PET-based materials for separator application was formed. The relevant research progress can be grouped into three phases. The first phase was from 2004 to 2010. The number of papers published has not fluctuated much over the years, accounting for about 18% of the total output. The second phase was from 2011 to 2017. The research on PET-based materials for separator application began to prosper, especially in 2015 with more than 8 papers. In the final stage, the research on PET-based materials for separator experienced a period of descent.

Figure 2 shows the dual map of published papers with the references cited in the content. It can be seen that the citing articles are mainly concentrated in the fields of physics, materials, and chemistry. The cited articles are mainly concentrated in the fields of physics, materials, chemistry, systems, and computing. Based on the overall profile of the published papers, it can be seen that although the research of PET-based materials for separator application has more than 15 years, it is still in a very active state. At the same time, the
research of PET-based materials for separator application does not involve many interdisciplinary subjects.

3.2. Author and Institution Analysis. Figure 3 shows the time-zone diagrams of different countries participating in the PET-based material for separator application. This figure not only shows which countries have participated in the PET-based material for separator application but also the degree of their participation (the larger the radius, the more times they participated) and the time of their first participation. Belgium first participated in the research of PET-based materials for separator applications in 2004. Japan, France, and Romania also reported papers on PET-based materials for separator applications in 2005. Then, South Korea, India, Italy, Portugal, and Sweden began publishing papers on PET-based material for separator applications from 2007 to 2010. After 2011, many countries have also joined this area, such as the USA, China, and Poland. It is worth noting that the time mentioned here for each country participating in PET-based materials for separator application investigation only represents the time when scholars from these countries published in the journals indexed in WoS and does not completely represent the time when they started research on PET-based materials for separator application.

It can be seen from Figure 2 that China, South Korea, France, and Japan have larger circle radii, which means that they have contributed the most to the papers of PET-based materials for separator application. Table 1 lists the number and centrality of published papers in different countries. China has published the largest number of papers and has the highest centrality, which means that China has the
greatest influence in the research of PET-based materials for separator application. Although South Korea was the started research on PET-based material for separator application at a very early stage and published 15 papers, its centrality is significantly lower than that of Germany, which has only published 2 papers. This means that although South Korea is a pioneer, these works did not have a very deep impact on follow-up research.

CiteSpace author cooccurrence analysis can identify the cooperation and mutual citation relationship between core researchers in a research field. Figure 3 shows the author’s collaboration network map. As can be seen from the figure, there are 4 main networks and some scattered research among the people engaged in PET-based materials for separator application. As can be seen from Table 2, Sangyoung Lee has been working on PET-based material for separator application since 2010. He published a total of 9 papers. It has a very close research collaboration with Jong Hun Kim, Eunsun Choi, and Hyunseok Jeong. These authors have published quite a number of papers. Dong Wang, Baojia Xiaohe, and Lucian Dascalescu each lead the research network. Among them, Lucian Dascalescu participated in the work of two small groups and became a bridge between the two groups.

Figure 4 shows the author collaboration and topic clustering analysis. In the timeline, we can see clearly how different authors influence each other’s research. We can also get a rough idea of what different authors focus on at different times. We can see that the work of some authors, such as Jung Hun Kim, Eunsun Choi, Wei Xiao [18, 95], and Jinglei Hao [17], has influenced the recent research.

From the above results, we can see that the number of publications of an author is not necessarily proportional to his value. To better measure the academic value of an author, author cocitation is a meaningful index. Figure 5 is the network of author co-citation. We can see that Hoik Lee is an essential author. He connects the two largest clusters in the network. Only one paper in the literature we selected included this author [117]. This work reported a novel membrane for the separator in a lithium-ion battery via a mechanically pressed process with a poly(vinylidene fluoride) (PVDF) nanofiber subject and PET microfiber support. This article plays an important role as a bridge because it refers to the important literature before it, and at the same time, the literature published after it also cited it. Authors who have played a similar role are Sangyoung Lee and Eunsun Choi, which we mentioned earlier in Table 2. In addition, some authors are not directly involved in this field, but their work plays an important role in the development of this field, such as Higashiyama Y [118, 119] and Augustin S [120].

We further conducted statistics and analysis with the author’s institution. Figure 6 shows the institution collaboration network map. Although it can be seen from Table 1 that different countries are conducting research on PET-based materials for separator application, according to Figure 7, most of these institutions are related.

We can see that there are links between Chinese institutions and those of Japan and the USA, which means that there is a cooperative relationship between these institutions. At the same time, there are also links among Romania, France, Germany, and Switzerland, indicating that there is a wide range of cooperation among European countries. There
may be some links between the institutions in South Korea and India. Poland, Sweden, and Italy are the exceptions. There is no international exchange in their research.

3.3. Keyword Analysis. In the CiteSpace keyword cooccurrence analysis, analyzing the changes in the number of cooccurring keywords in each year can not only judge the richness

| Name            | Paper no. | Started time | Reference                                      |
|-----------------|-----------|--------------|------------------------------------------------|
| Sangyoung Lee   | 9         | 2010         | [38, 101–108]                                  |
| Jong Hun Kim    | 6         | 2010         | [101, 103, 105, 106, 109, 110]                 |
| Eunsun Choi     | 5         | 2011         | [38, 102, 103, 105, 107]                       |
| Hyunseok Jeong  | 4         | 2007         | [102, 105, 107, 108]                           |
| Lucian Dascalescu| 3         | 2016         | [37, 39, 111]                                  |
| Baojia Xia      | 3         | 2014         | [36, 112, 113]                                 |
| Dong Wang       | 3         | 2014         | [114–116]                                      |
| Ke Liu          | 3         | 2014         | [114–116]                                      |

Figure 4: Timeline of author collaboration and topic clustering analysis.

Figure 5: Author cocitation network map.
of the research field but also judge the update speed of the content in the field and the vitality of the subject. By extracting the keywords of the PET-based material for separator application from 2004 to 2019, a total of 154 keywords were obtained. As shown in Figure 8, in the first two years of PET material for separator application, there were a lot of keywords. However, the number of keywords dropped suddenly in the third year, indicating that there were not many new studies published in this year, which is consistent with our previous analysis of the number of articles. Since 2007, the research on PET material for separator application showed a slowly increasing trend and reached the maximum value.

Figure 6: Institution collaboration network map.

Figure 7: Author’s collaboration network map.
in 2012. After that, the emergence of keywords began to decline. By 2017, only five new keywords appeared. Interestingly, although there are not many papers published in 2018-2019, the keywords show a new growth trend. This phenomenon may be due to the emergence of a new research direction, so a small number of papers contributed more keywords.

Table 3: Top 20 keyword frequency distribution of published papers.

| Keywords                  | Count | Centrality | Keywords                  | Count | Centrality |
|---------------------------|-------|------------|---------------------------|-------|------------|
| Membrane                  | 14    | 0.09       | Thermal stability         | 8     | 0.04       |
| Separator                 | 6     | 0.08       | Electrochemical performance| 6     | 0.04       |
| Performance               | 6     | 0.02       | Electrostatic separation  | 6     | 0.01       |
| Lithium-ion battery       | 5     | 0.05       | PET                       | 5     | 0.01       |
| Electrospun               | 4     | 0.03       | Polyethylene separator    | 4     | 0.03       |
| Liquid                    | 3     | 0.02       | Electrical property       | 3     | 0.02       |
| Composite separator       | 3     | 0.02       | Coating layer             | 3     | 0.01       |
| Electrolyte               | 3     | 0.01       | Cell                      | 3     | 0.00       |
| Particle                  | 3     | 0.04       | Composite                 | 2     | 0.03       |
| Cycling performance       | 2     | 0.02       | Cathode                   | 2     | 0.02       |

Figure 8: Annual distribution statistics visualization map of extracted keywords.

Figure 9: Top 8 keywords with the strongest citation bursts.

Top 8 keywords with the strongest citation bursts

| Keywords                   | Year  | Strength | Begin  | End    | 2004-2019 |
|----------------------------|-------|----------|--------|--------|-----------|
| Electrostatic separation   | 2004  | 2.65     | 2005   | 2009   |           |
| Electrospun                | 2004  | 1.52     | 2010   | 2019   |           |
| Lithium-ion battery        | 2004  | 2.02     | 2012   | 2019   |           |
| Pet                        | 2004  | 1.87     | 2013   | 2019   |           |
| Thermal stability          | 2004  | 2.39     | 2016   | 2019   |           |
| Polyethylene separator     | 2004  | 1.88     | 2016   | 2019   |           |
| Performance                | 2004  | 2.85     | 2016   | 2019   |           |
| Separator                  | 2004  | 2.31     | 2017   | 2019   |           |
Table 3 shows the top 20 keyword frequency distribution of published papers. We can see that some properties of PET separator, such as thermal stability, electrochemical properties, and electrical properties, are particularly concerned. This is because these properties directly affect the application of PET separator in batteries. At the same time, we found that PET composites also received the attention of researchers. This is because the combination of PET and other materials can improve the performance of the separator. Figure 9 shows the top 8 keywords with the strongest bursts. Their order is as follows: electrostatic separation, electrospray, lithium-ion battery, PET, thermal stability, polyethylene separator, performance, and separator. In chronological order, the burst words before 2010 are electrostatic separation and electrospray. These keywords represent that the research in this stage mainly focuses on the basic properties and preparation methods of PET. After 2010, the burst keywords have become more diverse.

Researchers began to use PET in lithium batteries and paid special attention to its thermal stability.

4. Conclusions

In this work, we used the bibliometrics software CiteSpace to analyze and analyze the literature published on PET-based materials for separator applications. The peak of this research area was 2011-2017. The works of this period mainly focused on the use of PET in lithium batteries and paid special attention to its thermal stability. Japan, France, and Romania are pioneers in this field. However, China, South Korea, and the United States have also contributed a lot of papers in this field. Sangyoung Lee, Jong Hun Kim, Eunsun Choi, Hyunseok Jeong, Dong Wang, Baojia Xiaohe, and Lucian Dascalu are the most representative authors in this field, and their work has an important impact on the whole field. On the whole, pet-based material for separator application has attracted researchers from different countries and generated extensive international cooperation.

Conflicts of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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