A State-of-the-art Review of Polyimide Foams Research

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ABSTRACT

Polyimide foams (PIFs), renowned as some of the best materials for excellent performance in harsh temperature environments, have been developed to meet the increasing demands of high-tech industries.. A state-of-the-art review of advanced PIFs research was conducted, covering the general research profiles and trends, institutions and scholars, major journals, research categories, and application fields through a bibliometric analysis of publications.Research on polyimide-based foam has increased approximately 30-fold over the past 40 years. One hundred forty-two research institutions from 20 countries have conducted research related to PIFs. Literature co-citation analysis reveals that the knowledge base of the PIFs research field primarily focuses on chemical synthesis foaming. This spotlight on engineering applications systematically describes the synthesis mechanisms, various typical fabrication methods, and the microstructure of PIFs. The advantages and disadvantages of this methods have been compared. Representative functions and corresponding mechanism models, which include thermal, mechanical, sensing, electromagnetic, flame retardant, oil-water separation, and other fields, have been outlined.. This review offers insights into the technological development prospects and opportunities. Finally, the arduous tasks and challenges of PIFs research are summarized, providing valuable guidance for researchers interested in this field.

***Keywords*:** Polyimide foams; Bibliometric analysis; Foaming method; Engineering application; Challenges.

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Introduction

Polymeric foam is a polymer-based (plastics, rubber, elastomers, or natural polymer materials) microporous material with numerous bubbles - also referred to as cells or cavities - in a polymer melt or a mixture of monomers. It can also be regarded as a composite material with gas as a filler, followed by a phase transition that stabilizes the resulting porous structure [1]. For polymers, most traditional thermoplastic and thermoset polymers can be foamed, *e.g.,* polyurethane (PU) foam [2], polystyrene (PS) foam [3], poly (lactic acid) foam [4], polyethylene terephthalate (PET) foam [5], polyvinyl chloride (PVC) foam [6], and polyimide (PI) foam [7], etc. Generally, polymeric foams are described as closed-cell and open-cell foams. In closed-cell foam, the gas forms discrete pockets, each surrounded by solid material, whilst in open-cell foam, gas pockets are connected [8,9]. The polymer phase guarantees the material's mechanical strength, while the gaseous phase ensures outstanding lightness, thermal insulation, and low conductivity. At an equal strength-to-weight ratio, there is no other material that can compete with the properties of polymeric foams [10,11]. For these reasons, they are now irreplaceable in various applications, including electronics, packaging, transportation, building, furnishing, clothing, aerospace, marine, etc. [12,13] However, conventional polymeric foams with poor thermal stability and mechanical properties cannot maintain the original morphology after been used in high temperatures, which limits their applications in high-temperature and other unique environments.

Polyimide foams (PIFs), one kind of ultra-high performance thermoset polymeric foams with imide rings in the leading chains under harsh temperature environments, have been developed to respond to the growing needs of high-tech domains, including aerospace, weaponry, microelectronics, sensors, and other industries, etc. [14-16] PIFs with specific chemical structures can readily withstand temperatures from as low as -250 °C to as high as 450 °C and maintain in long-term service at temperatures up to 300 °C [17], which are superior to traditional polymeric foams. Therefore, a wide variety of PIFs, including flexible or rigid foams with different cell structures (open, closed, or a combination of open/closed cells) in different micro and macro scales, can be used as heat insulation, sound absorption or natural materials, and also as an advanced composite sandwich in many high-tech fields. The first publication on the synthesis of aromatic polyimide was published in 1908 [18]. Nevertheless, PIFs were first produced and commercialized by Du Pont and Monsanto company until the 1960s [10-21]. Subsequently, the launch of commercial PIFs such as Solimide®, TEEK®, and Rexfoam® has dramatically expanded the global market as thermal insulation materials in high-tech fields [22,23]. As such, they had gained interest from domestic and foreign academic communities and industrial partitioners for their remarkable performance. However, the most

popular processes of PIFs are yet to be understood. This is because numerous reaction conditions, such as chemical additives, solvent, blowing agent, temperature, reaction time, moisture, impurity, etc., have a considerable impact on the polymerization reaction of aromatic dianhydrides with aromatic diamines [24]. Therefore, there is a crucial need for an in-depth analysis of research on PIFs.

A well-defined cell morphology of PIFs would involve their mechanical, dielectric, and thermal properties, etc., as well as their application range in different fields. For example, polyimide closed-cell rigid foams have demonstrated outstanding mechanical strength and thermal properties, which are often used as structural materials and flame resistant materials [25-27]. Polyimide open-cell foams can display conspicuous acoustic and electromagnetic wave performance on account of their excellent adsorption storage ability, which is useful in sectors like aerospace and military industries [28,29]. Moreover, there are other types of PIFs based on the open-cell morphology, such as aerogels [30], sponges [31], and nanoporous films [32], which are primarily employed for thermal insulation, biomedical and photocatalysis materials in various high-tech fields because of their ultralow density, ultrahigh porosity, and ultralow thermal conductivity [33]. The physical and chemical properties of PIFs depend on several factors, such as types of dianhydride and diamines [34], chemical constitutions, cell structure, gas properties, density, environmental temperature, pressure, time, facilities, etc. [26,35] At present, many kinds of PIFs with foaming mechanisms and controllable density have been described and reported [36-38]. In particular, PIFs have remarkable mechanical properties, including tensile, compression resistance, bearing capacity, flexural resistance, elasticity, and fatigue properties [29,39], thermal properties (i.e., thermostability, cold-resistant performance, and thermal insulation) [40,41], electromagnetic interference (EMI) shielding [42,43], superior flame retardancy [44,45], acoustic and wave absorption, oil-water separation performance [46-48], and environmentally friendly functions (i.e., hygroscopic, inhibited the proliferation of bacteria) [49-51], etc. Therefore, PIFs can not only act as structural materials but also play the role of functional materials [16,52]. They belong to advanced available materials, which are increasingly used as critical materials for thermal insulation, flame-retardant, vibration and noise reduction in aerospace, microelectronics, sensor, transportation, military hardware, and other high-tech areas [53,54]. The schematic illustration of fabrication methods, cell morphologies, multifunctional properties characterization, and applications of polyimide-based foams are shown in **Fig. 1**.

In the past decades, global research outputs in PIFs have increased remarkably, and research articles have significantly facilitated the promotion and application of PIFs. However, at present, there is a lack of state-of-the-art review on the systematic categories and quantitative summary of this field, such as general

profile and trends, cell morphology, synthesis mechanism, foaming method, multifunctional properties characterization, and application field, making it difficult to understand the overall structure of the intellectual landscape of PIFs research. A bibliometric analysis is an effective tool for conducting a quantitative analysis of academic literature using mathematical statistics techniques [55,56]. It has been widely used in various disciplines to explore the distribution of articles, countries, institutions, journals, scholars, and methods [57]. More importantly, it can accurately identify the relationships between fundamental and applied research, and explore the direction of technological development to better understand theoretical and technological innovations by summarizing the literature content. However, few studies have conducted research on the knowledge structure of polymer materials using bibliometric analysis. To the best of the authors' knowledge, this is the first study that aims to conduct a state-of-the-art review of PIFs research by using a bibliometric analysis. At the same time, it is the first study that conducts a systematic and in-depth analysis of the foaming method, reaction mechanism, microstructure, multifunctional properties characterization, and applications compared to other polyimide-based foam reviews.

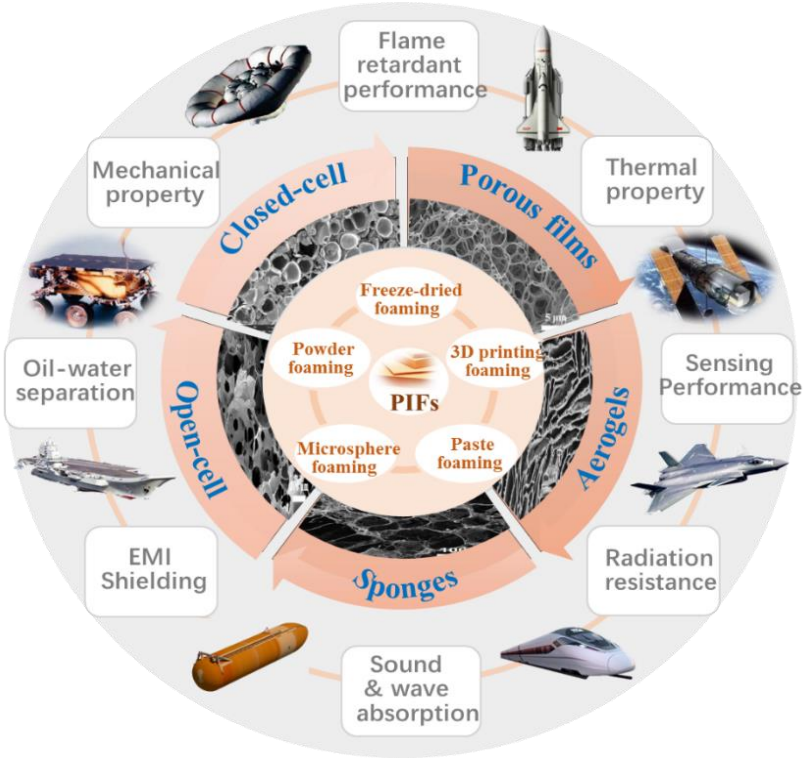

Fig. 1. The schematic illustration of fabrication methods, cell morphology, multifunctions properties characterization, and application of PIFs.

Therefore, this review paper attempts to conduct a state-of-the-art review and an in-depth analysis of PIFs research in the past 40 years based on a bibliometric analysis approach. A systematic literature research

was conducted to retrieve relevant articles from 1981 to 2022 (at the end of August). More specifically, we discussed five significant aspects, including 1) general profiles and trends, 2) countries, institutions, and scholars, 3) journals (impact factors and citations), 4) research categories (synthesis process, foaming mechanism and methods, chemical characterizations and physical morphologies of PIFs, multifunctional properties characterization, and applications of PIFs), 5) conclusions and future prospects. Finally, this review study also provided information on the technological development prospects and opportunities, which will be helpful for other researchers interested in PIFs.

Methods

Bibliometric is widely recognized as a well-established research method for investigating the literature quantitatively, particularly for evaluating the research performance of academics and universities [58,59]. Bibliometric analysis contributes to determining the size of studies in a particular scientific field and provides a complete view of the field. It is also useful for researchers, journal editors, and referees as it transforms qualitative data into quantitative data. Hence, the bibliometric analysis provides more objective and scientific results compared to manual reviews [60,61]. This study conducted a state-of-the-art review of polyimide foams research by using bibliometric analysis techniques based on several studies [62,63]. A combined keyword search string, including "polyimide foam OR PI foam OR PIFs OR porous polyimide OR polyimide aerogel OR polyimide sponge," was searched in the "article title/abstract/keywords" fields in each database from 1981 to 2022 (August). Searched databases include WOS Core Collection, MEDLINE, SciELO Citation Index, KCI-Korean Journal Database, Science Citation Index Expanded (SCIE), and Social Science Citation Index (SSCI). Articles that were not related to polyimide foam, as well as duplicates, were excluded. Books, book chapters, patents, and reports were also excluded. Afterward, PIFs-related articles were then sorted by using Note Express software to label the article titles, main topics, the first and corresponding authors, affiliations, abstracts, keywords, publication year, impact factor, journals, etc. In the end, 439 articles related to polyimide-based foam research were identified and used for further analyses in this study. The research methodology steps followed in this study can be seen in **Fig. 2**.

General profiles and trends

Fig. 3a presents an overview of the growth of studies on PIFs and the number of polyimide-based foam article types. The annual publication trend increased from 2 in 1981 to 64 in 2021 (at the end of August 31, 2022). In general, the development of polyimide-based foam-related studies was increasing yearly. Although few articles were reported every year before 2012 (from 1982 to 1996), the number of published articles has increased significantly since then. It was found that 369 processing and synthesis type articles

(including chemical blowing and physical blowing), 60 application type articles (polyimide-based foam as raw material), six reviews, and four 3D printing articles were included among the 439 articles retrieved. Notably, there were six articles that covered both synthesis and application. In the following analyses, the data for all the types of articles related to the studied topic were combined unless otherwise specified.

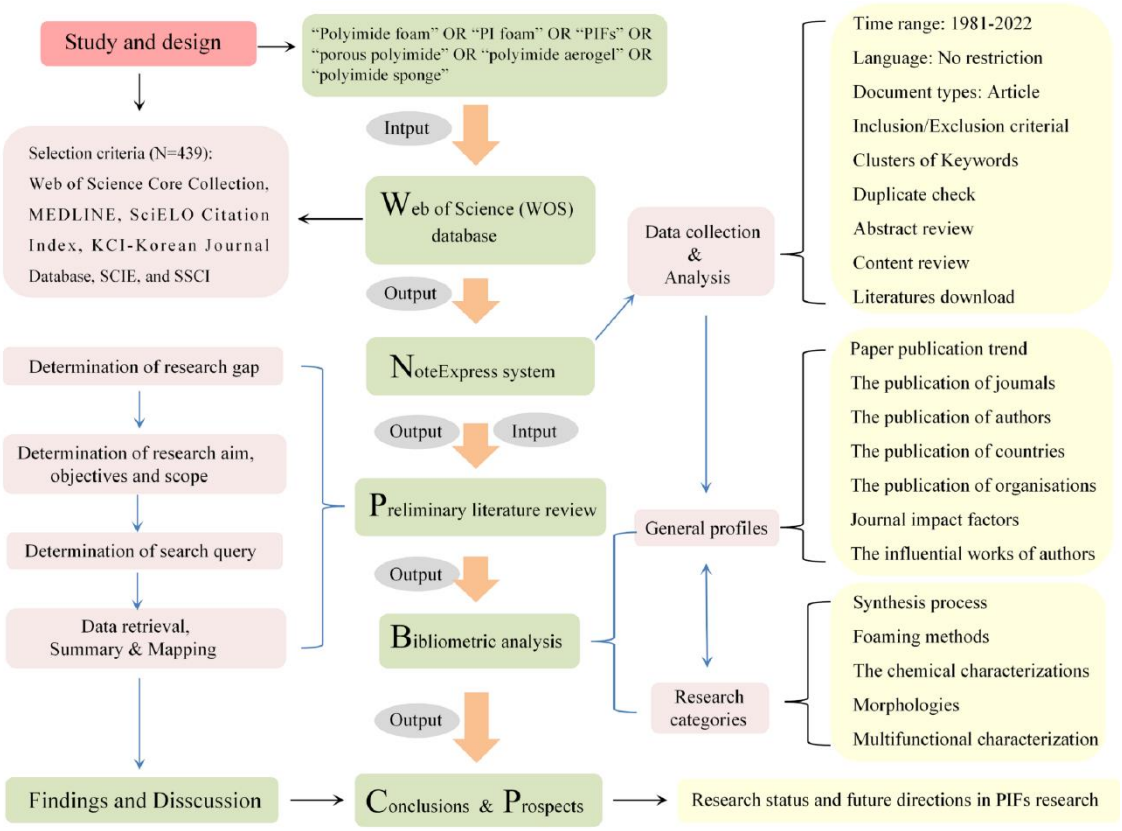

Fig. 2. Research methodology steps.

PIFs is increasingly gaining attention from researchers mainly due to its outstanding high-temperature resistance, excellent mechanical properties, lightweight, and good chemical stability. These characteristics are in growing demand in high-tech sectors such as aerospace, electronics, and automotive, where the performance requirements for materials are extremely high. This has driven investment in and technological innovations of polyimide foam research and development. As technology advances and new applications are developed, the number of related scientific papers has also increased, attracting more scientists to engage in research in this field. Moreover, with growing awareness of environmental protection and the demand for lightweight, durable materials, research into PIFs as a high-performance material has become even more active. Additionally, the development of PIFs has had a positive impact on both industry and academia. In the industrial sector, the application of this material has facilitated enhancements in product performance and energy efficiency optimization, particularly against the backdrop of increasing demand for high-performance insulation and lightweight materials. In academia, research into PIFs not only

expands the theoretical foundation of materials science but also provides a platform for the integration of related fields such as chemical engineering, physics, and environmental science. This accelerates the incubation and application transformation of innovative technologies, driving the advancement of scientific frontiers and the cultivation of talent.

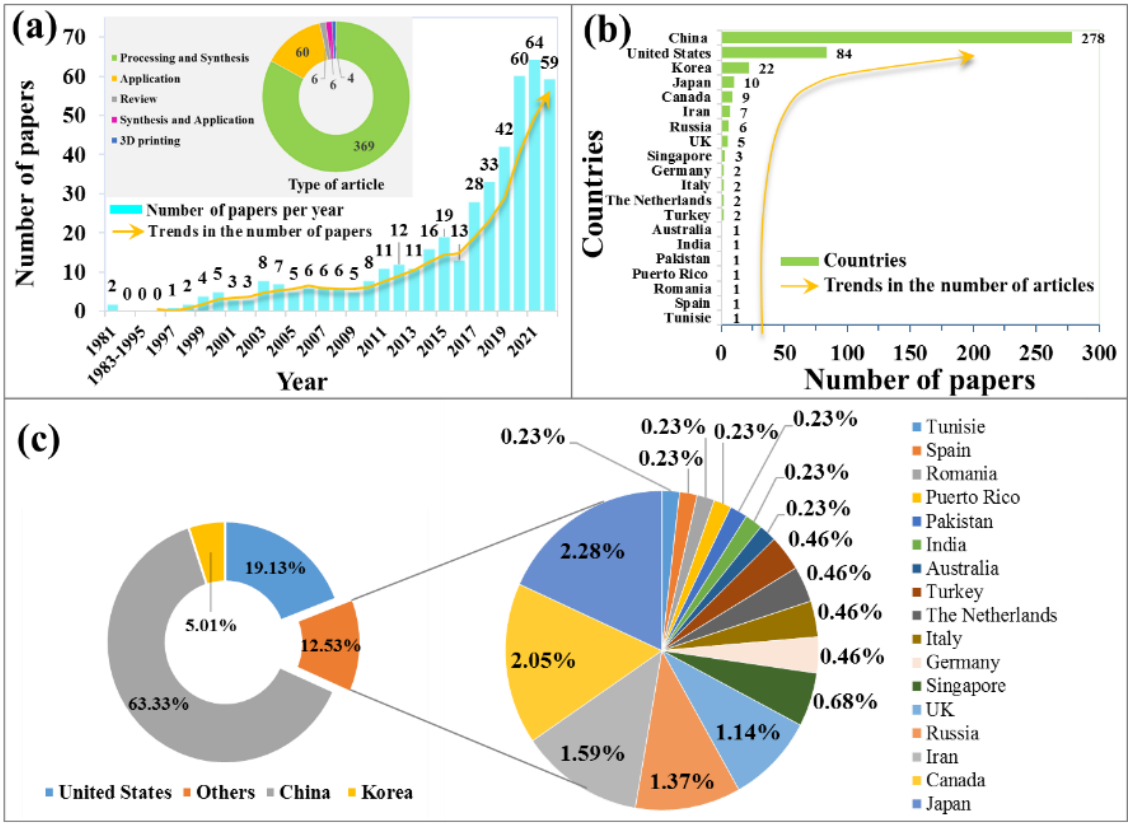

Fig. 3. (a) Annual trend of PIFs article numbers and the type summary. (b) Statistics of polyimide-based foam articles published in all countries. (c) Pie chart showing the proportion of articles published by the country.

Preliminary results revealed that the 439 articles originated from a total of 20 countries, where countries of corresponding authors were only accounted for in the analysis. As shown in **Fig. 3b**, the top ten countries were China, United States, Korea, Japan, Canada, Iran, Russia, United Kingdom, Singapore, and Germany. Amongst them, China contributed to 278 articles, accounting for 63.33 % of the total articles, and the United States contributed to 84 articles (19.13 %). Next, Korea had published 22 articles (5.01 %), the remaining 17 countries had published 55 articles in total (accounting for 12.53 %) (**Fig. 3c**). It can be found that PIFs-related research has experienced rapid development during 2017-2022, reaching a high peak in 2021 with 64 PIFs articles (out of which 54 articles are from China). Again, it should be noted that the search was conducted at the end of 31 August 2022. The high numbers of published articles in China may be explained due to the 10th China International Polyimide Material Technology and Application Exhibition in 2020-2021 with support from the Chinese Ministry of Science and Technology, thus

encouraging domestic polyimide materials (including polyimide foams) research. It can be concluded from **Fig. 3b,c** that the increase in PIFs research publications in recent years was mainly attributed to the contribution of Chinese and United States scholars.

INSTITUIONS AND SCHOLARS

Institutions

One hundred forty-two research institutions all over the world were involved in PIFs research during 1981 to 2022 (August) period, where the research institutes of the first and the corresponding authors were used for this analysis. The sub-institutes within a university or authorized institutes were classified as a single research institution. The results of research institutions on PIFs are presented in **Table 1**. The most productive PIFs institutions had published more than five articles accounting for 24 (16.9 %) of the total institutions. Research institutions with 2 to 4 articles accounted for 29 (20.42 %), while 88 (61.97 %) of the institutions contributed to only one article, finding that these research institutions may be considered as new institutes and could contribute to more PIFs research in the future. Unsurprisingly, Sichuan University was the most prolific institution with 29 articles from 2015 to 2022. Other five well-known research institutions including Donghua University, Beihang University, National Aeronautics and Space Administration (NASA), Harbin Engineering University, and the University of Akron have also published more than 15 articles. Many scientists, professors, researchers, and students have been working on part-time basis as visiting scholars. There were eight institutions with an average impact factor (IF) per article higher than 6.00 (published more than five articles), including Fudan University (IF, 11.56), Beijing University of Chemical Technology (8.36), Donghua University (8.07), Jiangnan University (7.81), Tongji University (7.75), Harbin Institute of Technology (6.41), Qilu University of Technology (Shandong Academy of Sciences) (6.38) and Beijing Jiaotong University (6.34). Probably these institutions were the first teams involved in PIFs field in the early stage producing some fundamental and typical articles.

It should also be noted that some promising new prolific scholar in China since late 2010 have published PIFs research of more than 10 articles including the Chinese Academy of Sciences, Beijing Technology and Business University, Northwestern Polytechnical University, Beijing University of Chemical Technology, etc. Undeniably, research institutions from the United States made more contributions to polyimide-based foam before 2010, such as NASA, Kennedy Space Center, Purdue University and The University of Akron, and so on, which also laid the foundation for the rapid development of subsequent PIFs research.

Table 1

Institutions Published PIFs Articles (1981-2022) and Statistical Analysis of the Scientific Impact of the Most Prolific Institutions.

Rank	Institutions	Country	Number of papers	Average impact factor	Time span
1	Sichuan University	China	29	5.11	2015-2022
2	Donghua University	China	26	8.07	2018-2022
3	Beihang University	China	22	3.78	2010-2021
4	National Aeronautics and Space Administration (NASA)	United States	20	2.48	1998-2020
5	Harbin Engineering University	China	15	4.84	2014-2022
6	The University of Akron	United States	15	5.26	2004-2021
7	Chinese Academy of Sciences	China	14	5.08	2012-2022
8	Beijing Technology and Business University	China	13	2.90	2015-2022
9	Northwestern Polytechnical University	China	12	3.31	2010-2022
10	Beijing University of Chemical Technology	China	10	8.36	2006-2022
11	Kennedy Space Center	United States	9	1.20	2000-2009
12	Yonsei University	Korea	9	4.45	2006-2022
13	Beijing Jiaotong University	China	8	6.34	2016-2022
14	Iowa State University of Science & Technology	United States	8	1.49	1981-2001
15	Jiangnan University	China	8	7.80	2019-2022
16	Qilu University of Technology (Shandong Academy of Sciences)	China	8	6.38	2019-2022
17	Ohio Aerospace Institute	United States	8	5.41	2011-2020
18	Fudan University	China	7	11.56	2016-2022
19	Harbin Institute of Technology	China	7	6.40	2015-2021
20	University of Toronto	Canada	7	4.46	2018-2020
21	Iran Polymer and Petrochemical Institute	Iran	6	2.19	2007-2014
22	Tongji University	China	6	7.75	2019-2022
23	Beijing Institute of Technology	China	5	5.33	2015-2021
24	Zhejiang University	China	5	4.47	2003-2020

25	118 Institutes published less than 5 PIFs articles	/	161	/	1981-2022
	2 Institutes published 4 PIFs articles		8		
	10 Institutes published 3 PIFs articles		30		
	18 Institutes published 2 PIFs articles		36		
	88 Institutes published only 1 PIFs articles		88		

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Authors/scholars and their influential works

Table 2 illustrates the top 15 articles published as the first author and corresponding author, as well as their affiliated research institutions. It was found that Xinfu Zhao from Qilu University of Technology (Shandong Academy of Sciences), as the first author, has published a total of 6 PIFs articles. Tianxi Liu from Jiangnan University, as the corresponding author, has published 18 PIFs-related articlesAugust during the studied period. Similarly, other scholars have published more than 10 PIFs articles as the corresponding authors, such as Maosheng Zhan (13 PIFs papers, Beihang University), Aimin Xiang (10, Beijing Technology and Business University) and Sadhan C. Jana (10, The University of Akron).

Some reviews and empirical research articles on PIFs were found to be highly cited during the studied period. **Table 3** presents the top 20 authors and their influential publications during the studied period August. The paper "Lightweight, Superelastic, and Mechanically Flexible Graphene/Polyimide Nanocomposite Foam for Strain Sensor Application," written by Yuyang Qin in 2015, was on top of the list with a cited count of 528, published in *ACS NANO* with an impact factor of 18.03. This paper aims to demonstrate a novel approach to fabricate a lightweight, super-elastic, and mechanically flexible nanocomposite foam by introducing water-soluble polyimide precursor into GO sheets. The flexible nanocomposite foam was very promising as a strain sensor. Another article titled "Templating nanoporosity in thin-film dielectric insulators," [64] written by professor Hedrick, J. L. in 1998, had cited count of 308 and an impact factor of 32.09, published in *ADVANCED MATERIALS*. This work discussed two routes to prepare nanoporous polyimide thin films. A method has been developed to generate nanoporous PIFs from block copolymers in which the dispersed phase underwent decomposition thermally. Another method describes the use of branched, highly functional macromolecules as molecular templates in the vitrification of silsesquioxane. The two above-mentioned articles are of great reference value to scholars or researchers engaged in the research of polyimide-based porous membranes and aerogels. Other approaches used branched, highly functional macromolecules as molecular templates during the vitrification of a siles-quioxane. These articles were very worthy of reference for some scholars or researchers researching polyimide-based porous films and aerogels.

Table 2

The Number of Top 15 Articles Published as the First Author and Corresponding Author in 1981-2022 (August) along with Their Institutions.

Rank	The first author			The corresponding author		
	Authors	Number of papers	Institutions	Authors	Number of papers	Institutions
1	Xinfu Zhao	6	Qilu University of Technology (Shandong Academy of Sciences)	Tianxi Liu	18	Jiangnan University
2	Haiquan Guo	4	Ohio Aerospace Institute	Maosheng Zhan	13	Beihang University
3	Mary Ann B. Meador	4	National Aeronautics and Space Administration (NASA)	Aimin Xiang	10	Beijing Technology and Business University
4	Camilo I. Cano	3	Purdue University	Sadhan C. Jana	10	The University of Akron
5	Gaohui Sun	3	Harbin Engineering University	Pengbo Liu	9	Sichuan University
6	Jianwei Li	3	Xi’an Polytechnic University	Shihui Han	9	Harbin Engineering University
7	Lingying Pan	3	Beihang University	Mary Ann B. Meador	8	National Aeronautics and Space Administration (NASA)
8	Shahriar, G.M.	3	University of Toronto	Rubing Zhang	7	Beijing Jiaotong University
9	Shiya Qiao	3	Donghua University	Andreas Greiner	6	University of Bayreuth
10	Xianbo Hou	3	Beijing Jiaotong University	Haksoo Han	6	Yonsei University
11	Xiaoming Sang	3	Hebei United University	Kai Wang	6	Beihang University
12	Yangfeng Gao	3	Dalian University of Technology	Martha K. Williams	6	Kennedy Space Center
13	E.N.Popova	2	Russian Academy of Sciences	Guangcheng Zhang	5	Northwestern Polytechnical University
14	Gaohui Sun	2	Harbin Engineering University	Shengtai Zhou	5	Sichuan University
15	Huafeng Tian	2	Beijing Technology and Business University	Shiyong Yang	5	Chinese Academy of Sciences

Table 3

Most Cited PIFs Research, including Authors, Journal Cited Count and Impact Factor in 1981-2022 (August).

Rank	Author	Title	Journal	Year	Cited Count	IF	ref
1	Yuyang Qin, Yibin Li	Lightweight, Superelastic, and Mechanically Flexible Graphene/Polyimide Nanocomposite Foam for Strain Sensor Application	*ACS NANO*	2015	528	18.03	[65]
2	Hedrick, J. L.	Templating nanoporosity in thin-film dielectric insulators	*ADVANCED MATERIALS*	1998	308	32.09	[64]
3	Meador, M. A. B.	Mechanically Strong, Flexible Polyimide Aerogels Cross-Linked with Aromatic Triamine	*ACS APPL MATER INTER*	2012	258	10.38	[66]
4	Ji Liu, Haobin Zhang	Multifunctional, Superelastic, and Lightweight MXene/Polyimide Aerogels	*SMALL*	2018	229	15.15	[67]
5	Haiquan Guo, Mary Ann B. Meador	Polyimide Aerogels Cross-Linked through Amine Functionalized Polyoligomeric Silsesquioxane	*ACS APPL MATER INTER*	2011	227	10.38	[68]
6	Yang Li, Wenge Zheng	Polyimide/graphene composite foam sheets with ultrahigh thermostability for electromagnetic interference shielding	*RSC ADVANCES*	2015	171	4.04	[36]
7	Krause, B	Open nanoporous morphologies from polymeric blends by carbon dioxide foaming	*MACROMOLECULES*	2002	166	6.06	[69]
8	Jingjing Ma, Maosheng Zhan	Ultralightweight Silver Nanowires Hybrid Polyimide Composite Foams for High-Performance Electromagnetic Interference Shielding	*ACS APPL MATER INTER*	2015	157	10.38	[70]
9	Shaohua Jiang, Andreas Greiner	Ultralight, Thermally Insulating, Compressible Polyimide Fiber Assembled Sponges	*ACS APPL MATER INTER*	2017	155	10.38	[71]
10	Inagaki, M.	Carbon foams prepared from polyimide using urethane foam template	*CARBON*	2004	151	11.31	[72]
11	Haiquan Guo	Tailoring Properties of Cross-Linked Polyimide Aerogels for Better Moisture Resistance, Flexibility, and Strength	*ACS APPL MATER INTER*	2012	149	10.38	[73]
12	Mary Ann B. Meador	Low Dielectric Polyimide Aerogels As Substrates for Lightweight Patch Antennas	*ACS APPL MATER INTER*	2012	146	10.38	[74]
13	Mary Ann B. Meador	Polyimide Aerogels with Amide Cross-Links: A Low Cost Alternative for Mechanically Strong Polymer Aerogels	*ACS APPL MATER INTER*	2015	144	10.38	[75]
14	Fei Zhang, Wei Feng	Stress Controllability in Thermal and Electrical Conductivity of 3D Elastic Graphene-Crosslinked Carbon Nanotube Sponge/Polyimide Nanocomposite	*ADV FUNCT MATER*	2019	143	19.92	[76]

15	Hu Liu, Chuntai Liu.	Lightweight, Superelastic, and Hydrophobic Polyimide Nanofiber /MXene Composite Aerogel for Wearable Piezoresistive Sensor and Oil/Water Separation Applications	*ADV FUNCT MATER*	2021	141	19.92	[77]
16	Yang Li, Wenge Zheng	Ultrathin carbon foams for effective electromagnetic interference shielding	*CARBON*	2016	138	11.31	[78]
17	Lee, Y. J.	Low-dielectric, nanoporous polyimide films prepared from PEO-POSS nanoparticles	*POLYMER*	2005	137	4.43	[79]
18	Xiaoyu Chen, Hu Liu	Highly Compressible and Robust Polyimide/Carbon Nanotube Composite Aerogel for High-Performance Wearable Pressure Sensor	*ACS APPL MATER INTER*	2019	136	10.38	[80]
19	Carter, K. R.	Nanoporous polyimides derived from highly fluorinated polyimide/poly(propylene oxide) copolymers	*CHEMISTRY OF MATERIALS*	2001	126	10.51	[81]
20	Leventis, N.	Polyimide Aerogels by Ring-Opening Metathesis Polymerization (ROMP)	*CHEMISTRY OF MATERIALS*	2011	113	10.51	[82]

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JOURNALS

The 439 included articles were published in 174 different journals, thus, an average of 2.5 PIFs articles published by each journal. However, this wasn't the case since most journals (113 articles or 64.94 %) published a single polyimide-based foam article over 40 years. Consequently, only few journals have published the majority of PIFs articles. Considering the wide variety of journal sources, it can be explained that PIFs research articles focused on both an "experiment and processing techniques" and "application and review" that can help to solve a similarly wide range of problems.

Number of published articles

Fig. 4 displays the journal outlets that have published more than 5 PIFs articles and their 2022 impact factor, above 50.34 % of articles published in the top 20 journals from 1981 to 2022 (August). According to the results, 20 bulletins (11.49 %) have issued more than 5 PIFs-related articles. Among these journals, ACS APPL MATER INTER was the most prolific journal with 31 published articles during the studied period. As a top-ranked international journal, *ACS APPL MATER INTER* mainly received articles related to materials science (nanoscience & nanotechnology) and multidisciplinary research areas. There was no doubt that PIFs articles are aligned with the journal's scope.

The *J APPL POLYM SCI* has also published 26 PIFs articles over the studied 40-year period. As a rising journal, *J APPL POLYM SCI*, which received articles interrelated to chemistry or polymer science, was very grateful among researchers due to its pro-people. Therefore, it was the most crucial support for PIFs publications. Last but not least, several other journals worth mentioning have published more than 10 PIFs articles, such as *CHEM ENG J, POLYMER, RSC ADVANCES, CARBON, COMPOS SCI TECHNOL and COMPOS PART B-ENG,* etc.

Journal impact factors

As shown in **Fig. 4**, the journals that published PIFs articles have very different impact factors (IF). *ACS APPL MATER INTER*, the top-ranked journal for PIFs scientists, had an IF of 10.38 in 2022. Eighteen journals with IF > 10.38 (*ACS APPL MATER INTER*) have published 62 articles focusing on PIFs research. The journal, *ADVANCED MATERIALS,* was the leading journal in the field with an IF of 32.09. However, only 2 PIFs articles were published in this journal in 1998 and 2004 during the studied period.

There were 6 PIFs review articles published in *ACS APPL MATER INTER* (IF 10.38) [8], *PROGRESS IN POLYIMIDE CHEMISTRY II* (IF 10.13) [83], *COMPOS PART A-APPL S* (IF 9.46) [84], *ACTA CHIMICA SINICA* (IF 2.79) [85], *J MACROMOL SCI A* (IF 2.22) [86] and *PROG CHEM* (1.04) [87], respectively. The above review articles are worth reading for any PIFs researchers because they can inspire

and provide insightful understanding on the progress of polyimide-based foam materials in recent years. On the other hand, it was found that 31 journals with no impact factor in the WOS database have published 50 PIFs-related articles, most of which were international conference articles.

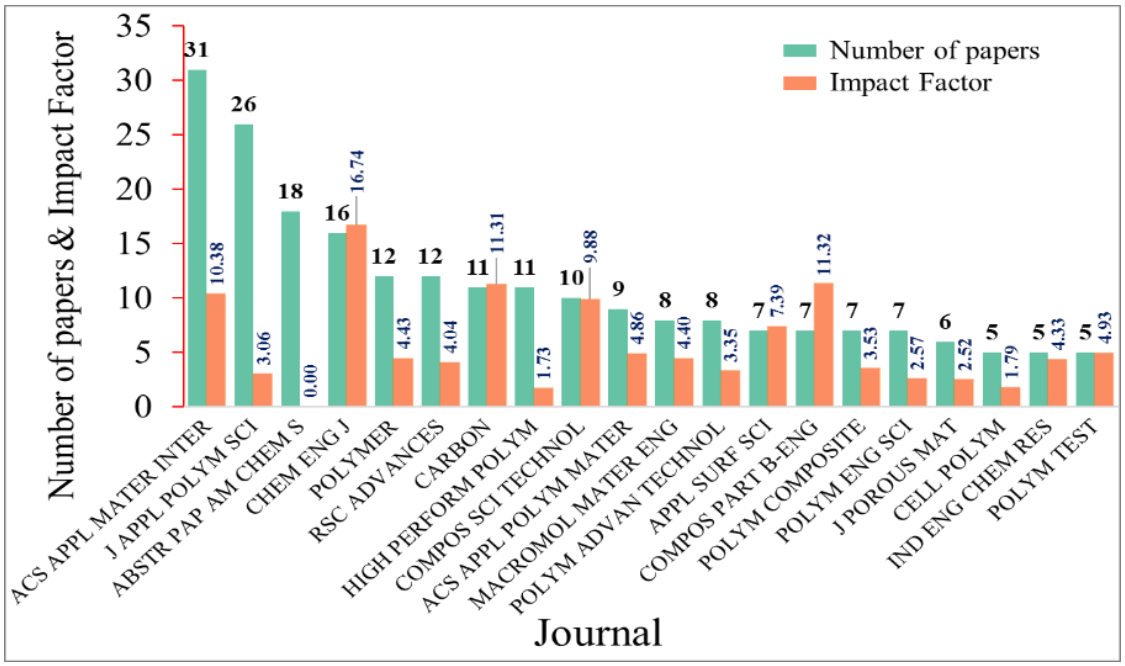

Fig. 4. The journals published more than 5 PIFs-related articles in 1981-2022 (August), along with their impact factors.

RESEARCH CATEGORIES

During the studied period, research categories related to PIFs were more diverse, and the application fields were more and more widespread. Nevertheless, the development and applications of PIFs are still emerging in recent research topics. Polyimide-based foams are used in diverse research applications because they have (1) outstanding comprehensive properties; (2) numerous approaches in the synthesis process; (3) various foaming methods; (4) potential uses in several high-tech fields; (5) challenging issues. Due to its thermosetting characteristics, almost the preparation of PIFs is via chemical synthesis foaming. To understand the whole foaming process of polyimide-based foams, this review study conducted a detailed review of the synthesis process, foaming mechanism and methods, chemical structure and physical morphologies, multifunctional properties characterization, application fields and advanced cases of PIFs.

Synthesis process

PIFs, as a kind of polymer foams with wide varieties, are mainly composed of polyimide resin and internal open or closed pore structures with different sizes [8,87]. Usually, the preparation of PIFs is as follows: (1) polyamide acid solution (PAAs) is synthesized by low-temperature polycondensation of dianhydride and diamine in a polar solvent such as dimethyl formamide (DMF), dimethylacetamide (DMAc), N-Methylpyrrolidone (NMP) or tetrahydrofuran (THF)/methanol(MeOH) mixed solvents; (2) the

PAAs is dried into precursor powders under vacuum/or PAAs is foamed directly by using the freeze-dried method; (3) the prepolymer particles are added into a container (residual solvent and minor molecule by-products as blowing agent during synthesis), whereafter, PIFs can be obtained by heating PAA precursor particles (thermal imidization process) [88,89].

The two monomers of dianhydride and diamine have vast sources of raw materials, low toxicity, and accessible synthesis compared to other heterocyclic polymer monomers, such as polybenzimidazole, polybenzoxazole, polybenzothiazole, and polyquinoline, etc. Also, the diversity in monomer types allows for unlimited possibilities to develop PIFs for different varieties and end-use purposes [90,91]. According to statistics, 200-300 kinds of dianhydride and diamine have been used to synthesize polyimide foam [58,92,93]. Therefore, there are significant differences in polyimide foam's synthetic route, foaming mechanism, and foaming process. Although these methods are relatively complex and the synthesis process takes a long time, they have merits in regulating foam density and cell structure [94,95].

Table 4 shows the number (more than 2) of articles that used the same type of dianhydride and diamine during the studied period Augustamong the 369 processing and synthesis PIFs articles. The results revealed that 139 articles have used pyromellitic dianhydride (PMDA) as dianhydride, mainly due to its high electron affinity (Ea). In addition, 4,4′-oxydianiline (ODA), as the most popular diamine monomer, was used in 193 (52.3 %) articles, thanks to its high acidity coefficient (kPa). It was noteworthy that 14 articles used more than two kinds of dianhydride monomers for blending, and another 27 articles have used more than two types of diamine monomers for mixing.

Table 4

The Number (more than 2) of PIFs-related Articles Used the Same Type of Dianhydride and Diamine in 1981-2022 (August).

Rank	Dianhydride			Diamine		
	Name of monomer	Abbr	Number of papers	Name of monomer	Abbr	Number of papers
1	Pyromellitic dianhydride	PMDA	139	4, 4′-oxydianiline	ODA	193
2	3,3',4,4'-Biphenyltetracarboxylic dianhydride	BPDA	91	Polyaryl polymethylene isocyanate	PAPI	46
3	3,3',4,4'-Benzophenonetetracarboxylic dianhydride	BTDA	60	2,2′-dimethylbenzidine	DMBZ	34
4	Polyamide acid	PAA	41	p-phenylenediamine	p-PDA	27
5	4,4'-Oxydiphthalic anhydride	ODPA	24	4,4′-Diaminodiphenylemthane	MDA	18
6	4,4'-(Hexafluoroisopropylidene) diphthalic anhydride	6FDA	6	m-phenylenediamine	m-PDA	3
7	2,2-Bis[4-(3,4-dicarboxyphenoxy)phenyl] propanedianhydride	BPADA	3	Dermorphin [Phe4] (1-4) amide	TAPB	2
8	3,3,4,4-diphenylsulfonetetra carboxylic dianhydride	DSDA	2	1,3-Bis(4-aminophenoxy)benzene	TPE-R	2
9	1,4,5,8-Naphthalenetetracarboxylic dianhydride	NTCDA	2	4,4'-bis(4-aminophenoxy) propane	BAPP	2
10	Bismaleimide	BMI	2	4,4'-Diaminodiphenyl Sulfone	DDS	2
11	Blending of more than 2 monomers	-	14	Blending of more than 2 monomers	-	27

Table 5

The Foaming Mechanism, Advantages and Disadvantages of Different Preparation Methods of PIFs.

Preparation methods	foaming mechanism	advantages	disadvantages	ref
Freeze-dried foaming	(a) PAA solution is synthesized by dianhydride and diamine in polar solvent; (b) extra solvent is removed during synthesis, residual solvent and small molecule by-products as blowing agent; (c) thermal forming and imidization foaming of PAAs;	PIFs with integral 3D structure can be obtained	The shape of the foam materials is adjustable to some extent but not completely controllable	[97,98]
Precursor powder foaming		PIFs with high density can be acquired	Inhomogeneous heat transfer of powder may lead to the nonuniformity of the foam structure	[99,100]
Microsphere foaming		PIFs with high closed and uniform cell can be gained	The viscosity between microspheres has a great influence on the mechanical properties	[101,103]
3D printing method		PIFs with arbitrary shape can be achieved	The requirements for costly equipment are very high	[104,105]
Paste foaming process		PIFs with lower density can be earned	The uniformity of PIFs cell is difficult to adjust	[106,107]

Foaming mechanism and methods

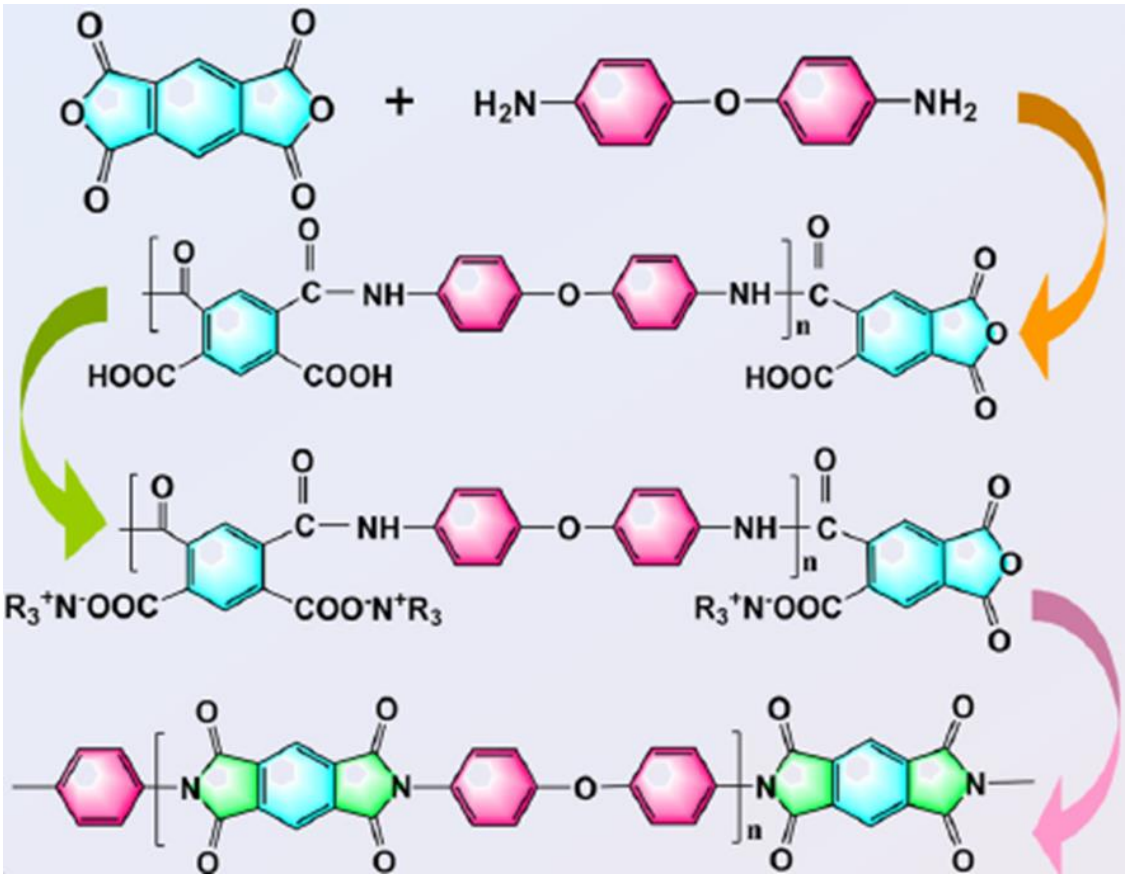

Fig. 5. The schematic illustration of fabrication strategy and reaction mechanisms for polyimide-based foams.

The research on the foaming process and mechanism was the basis and key to solving polyimide structure and cell morphology. A variety of functions of polyimide foams depend on their morphology to a great extent. How to control the morphology of polyimide foam by forming has always been one of the most concerning issues for researchers. Studying the foaming process and mechanism is the foundation and key to solving this issue. The synthesis typically involves polycondensation of diamines and dianhydrides to form polyamic acid, which is then thermally treated to form polyimide foams. The choice of monomers and synthesis conditions directly impacts the foam's properties and applications. **Fig. 5** illustrates the reaction mechanism of polyimide synthesis [96]. The schematic illustration was based on a dianhydride (PMDA) polycondensation reaction and diamine (ODA) in a dimethylacetamide (DMAc) solution. Then, poly amic acid solution (PAAs) was synthesized successfully. Ultimately, PIFs can be acquired via the thermal imidization process. To better understand the formation mechanism of polyimide foams, Lee and co-workers [97] investigated the fabrication process of polyimide foam. They believed that the forming mechanism experienced from the nucleation site to the rod-shaped structures and, finally, the spherical microparticles. The nucleation site comes from the aggregation of abundant primary particles, which the

polycondensation reaction of dianhydride and diamine can introduce. The numerous primary particles quickly aggregate together to form a rod-shaped morphology, and then spherulite can be achieved [8]. According to the state of PAA precursor polymer, the PIFs forming process can be divided into solution freeze-dried foaming [98,99], precursor powder foaming [100,101], microsphere foaming [102,103], three-dimensional (3D) printing [104,105], and paste foaming method [106,107]. Among these methods, the freeze-dried and precursor powder foaming method have been widely employed to prepare high heat and temperature-resistant polyimide-based foams [108,109]. **Table 5** shows the foaming mechanism, advantages, and disadvantages of different preparation methods of polyimide-based foams.

Freeze-dried foaming

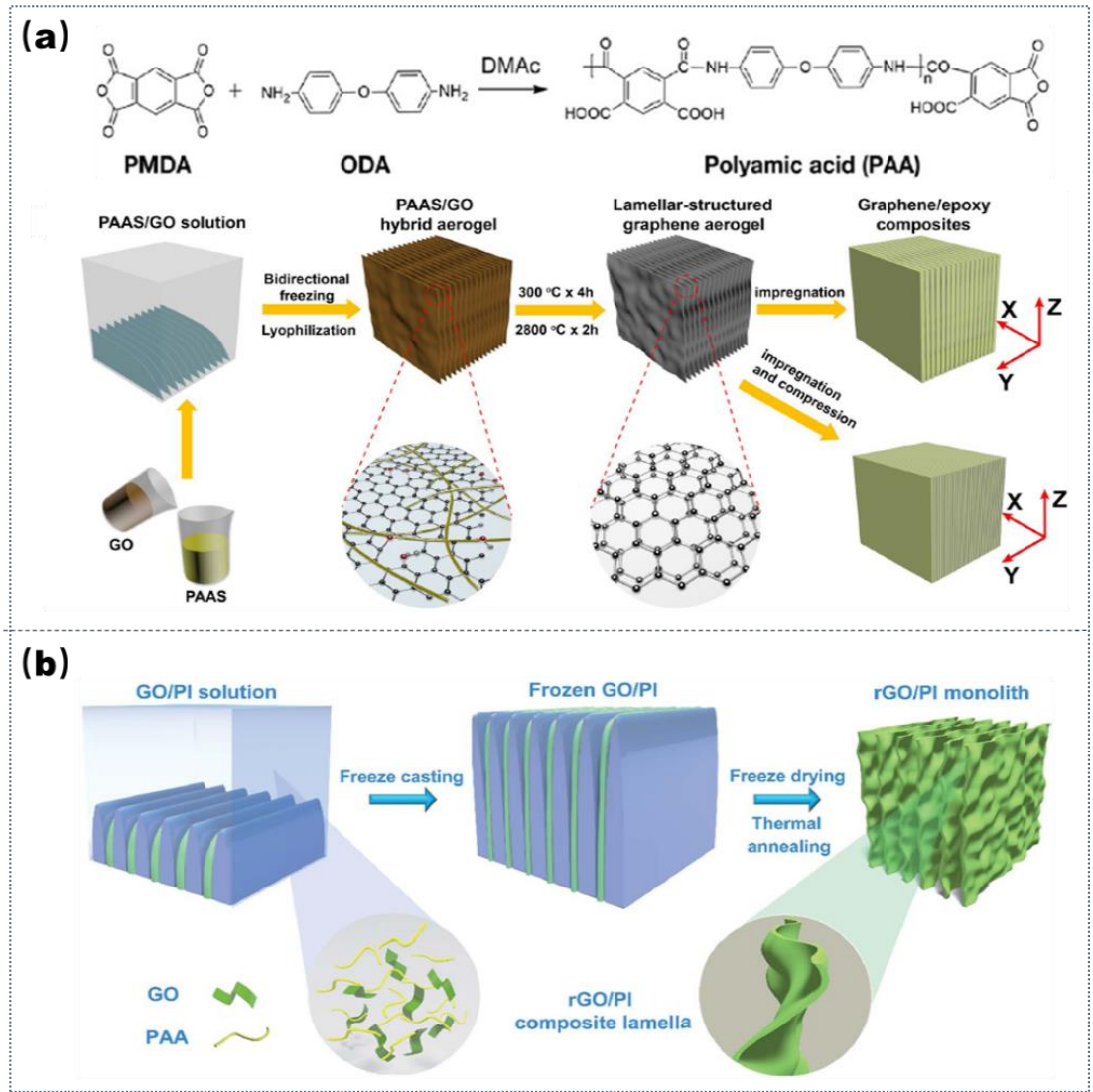

Fig. 6. (a) Schematic illustration of the fabrication of PAAS/GO aerogel using the solution freeze-dried foaming. (b) Structural design of the anisotropic rGO/PI thermally insulating nanocomposite foam with highly ordered structures. Schematic illustration of the fabrication process of the nanocomposite foam by freeze-casting and thermal annealing.

The method of PAAs freeze-dried foaming is mostly used to prepare PI aerogel. Usually, PAAs is first synthesized by dianhydride and diamine in a polar solvent. The solution is then repeatedly washed with

deionized water and dried in the air. Subsequently, PAA hydrogel can be obtained by pouring it into a mold. Finally, the hydrogel samples are put into a freeze dryer to cast different microstructures via random or unidirectional freezing, where the ice crystals grow randomly or along a generated thermal gradient. When the concentration is above 8 %, the PAAs randomly structured aerogels can be obtained [37]. Pengfei Liu et al. [110] prepared a series of lamellar-structured graphene aerogels with unprecedented thermal conduction capacity for the first time by the following steps: (1) PAAs was first synthesized via polymerization of PMDA and ODA in DMAc solvent; (2) Deionized water was then washed the PAAs repeatedly to prepare polyamic acid salt (PAAS) with the aid of triethylamine (TEA) under magnetic stirring. (3) After a graphene oxide (GO) suspension was added into the PAAS to obtain PAAS/GO suspension, the PAAS/GO hybrid aerogels were fabricated by pouring into a mold to obtain PAA hydrogel. (4) Finally, the hydrogel samples were put into a freeze dryer to cast different microstructures of PI aerogel via random or unidirectional freezing, imidization, and graphitization, as shown in **Fig. 6a.** Moreover, Yuyang Qin et al. reported that the preparation of rGO/PI foam with anisotropic, mechanically flexible, and highly thermally insulating using the freeze-drying forming method, which can be seen in **Fig. 6b** [40]. PMDA and ODA with a molar ratio of 100 to 99 were uniformly dispersed in a DMAc solution with a magnetic stirrer to obtain PAA precursors. The resultant solution was poured into excessive deionized water, and the produced precipitate was filtered, washed, and dried to remove the residual deionized water. After vigorously stirring the GO suspension and precursor solvent mixture, a three-dimensional porous structure without destroying the internal chemical bonds of the as-obtained materials was prepared by the freeze-dried method.

Powder foaming

Customarily, PAA precursor powders should be first added into an airtight mold. The mold is heated to reduce the residual solvent content to 1~10 %, and the subsequent thermal foaming treatment should be carried out. PIFs with high density can be acquired via power foaming. However, inhomogeneous heat transfer of powder may lead to the nonuniformity of the foam structure. Traditionally, thermal imidization of PAA foam at high temperatures was carried out to achieve a PI foam. This typical method possessed three main features: (1) the molar ratio of diacid diester to dianhydride could be fixed at 1:1 so that polyimide-based foams with high relative molecular weight can be prepared conveniently; (2) taking a mixture of THF and MeOH as solvent was cost-effective, and the residual solvent can be used as a foaming agent. It should be mentioned that the density and cell structure of PIFs can be effectively controlled by regulating the residual solvent content; (3) tertiary amine imidization catalyst could help to reduce the thermal imidization temperature, thus resulting in lower foaming temperature. A variety of functions of

PIFs depended on their morphology to a great extent. Determining the morphology of PIFs by the forming process has always been one of the most concerning issues for researchers. Long Ni et al. [111] prepared BTDA-derived PIFs with different diamine structures by the PAA powders foaming method, as shown in **Fig. 7a**. Similarly, Yueyi Wang and co-workers [97] fabricated the carbon nanotube (CNT)/PI foam using the traditional PAA precursor powders foaming method, which can be seen in **Fig. 7b**. The PAAs was procured by condensation polymerization of PMDA and ODA (the molar ratio was set to 100:99) in DMAc solvent. Whereafter, the obtained PAAs was poured into the deionized water to allow precipitation and washed with deionized water several times to get rid of the residual water by freeze-dried. Subsequently, the solid PAA precursor powder was dissolved in deionized water to obtain a water-soluble polyimide precursor solution. After vigorously stirring the mixture of CNT suspension and the precursor solvent, the mixture was freeze-dried to get the CNT/PAA porous structure foam. The CNT/PAA foam was initially heated to induce thermal imidization to obtain the CNT/PI foam.

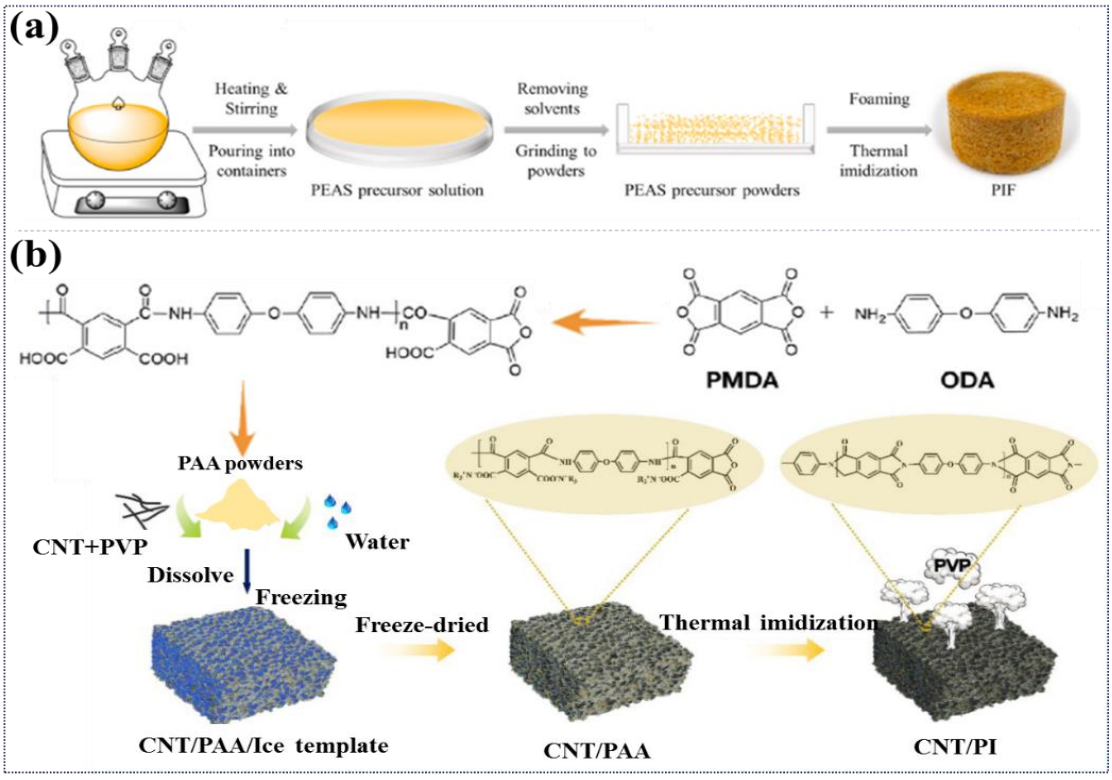

Fig. 7. (a) Schematic illustration of fabricating BTDA-derived PIFs with different diamine structures by PEAS powders foaming method. (b) Schematic diagram of the fabrication of CNT/PI porous structure foam by using the PAA powders foaming method.

Microsphere foaming

The microsphere foaming method is proposed based on the PAA precursor powder method, where microsphere refers to the product of powder pre-foaming. Microsphere foaming aims to improve the closed cell rate, cell uniformity, and structure controllability of PIFs. However, the viscosity between microspheres

significantly influences the mechanical properties. Yinfu Luo et al. [102] fabricated the three-dimensional polyimide microsphere foam. The facture steps were as follows: (1) the PEAS was synthesized by the polymerization of BPDA and MDA with an equivalent mole in THF and methanol mixed solution; (2) PIFs were prepared by the PEAS thermal forming and imidization, as shown in **Fig.8a**. The results showed that the hollow PI microspheres had outstanding heat resistance, with the bulk density varying from 56.9 to 132.2 kg/m^3. In addition, PI microspheres could be easily converted into carbon microspheres without destroying their original integrity, which displayed potential application value in preparing multifunctional composite materials.

Paste foaming

The paste foaming method used the polyimide precursor with high solid content (65~85 % (mass)) to prepare polyimide foam with a density of less than 2 kg/m^3 [107,112]. PIFs prepared by the paste foaming method had uniform cells and excellent flexibility. Nonetheless, the uniformity of PIFs cells was difficult to adjust. Zhouping Sun and colleagues fabricated the CNT/PI three-dimensional (3D) structure foam using paste foaming method, which can be seen in **Fig. 8b** [113] To obtain a PAA precursor solution, PMDA and PAPI were uniformly dissolved in a mixture of THF and MeOH solvent. Subsequently, the mixed paste solution was poured into a container for free foaming. Then, it was placed into an oven to complete the imidization reaction when the foam sample was no longer tacky. Finally, the PIF/CNT foams were prepared via a paste solution dip-coating foaming method.

Three dimensions (3D) printing foaming

3D printing foaming method has accelerated the exploration of previously unattainable functionalities of electronics via expanding the geometrical construction design of customizable building materials [59]. As displayed in **Fig. 8c**, Clive Liu et al. [114] fabricated polyimide closed-cell stochastic foams with tunable densities using the method of 3D direct ink printing filaments. Compared to other methods, the 3D printing process had an unparalleled advantage in preparing sophisticated equipment with arbitrary shapes and excellent interface bonding performances, which can be applied to aerospace, sensors, and other high-tech fields. Although the 3D printing foaming method can realize PIFs foam of any shape, the equipment can be costly.

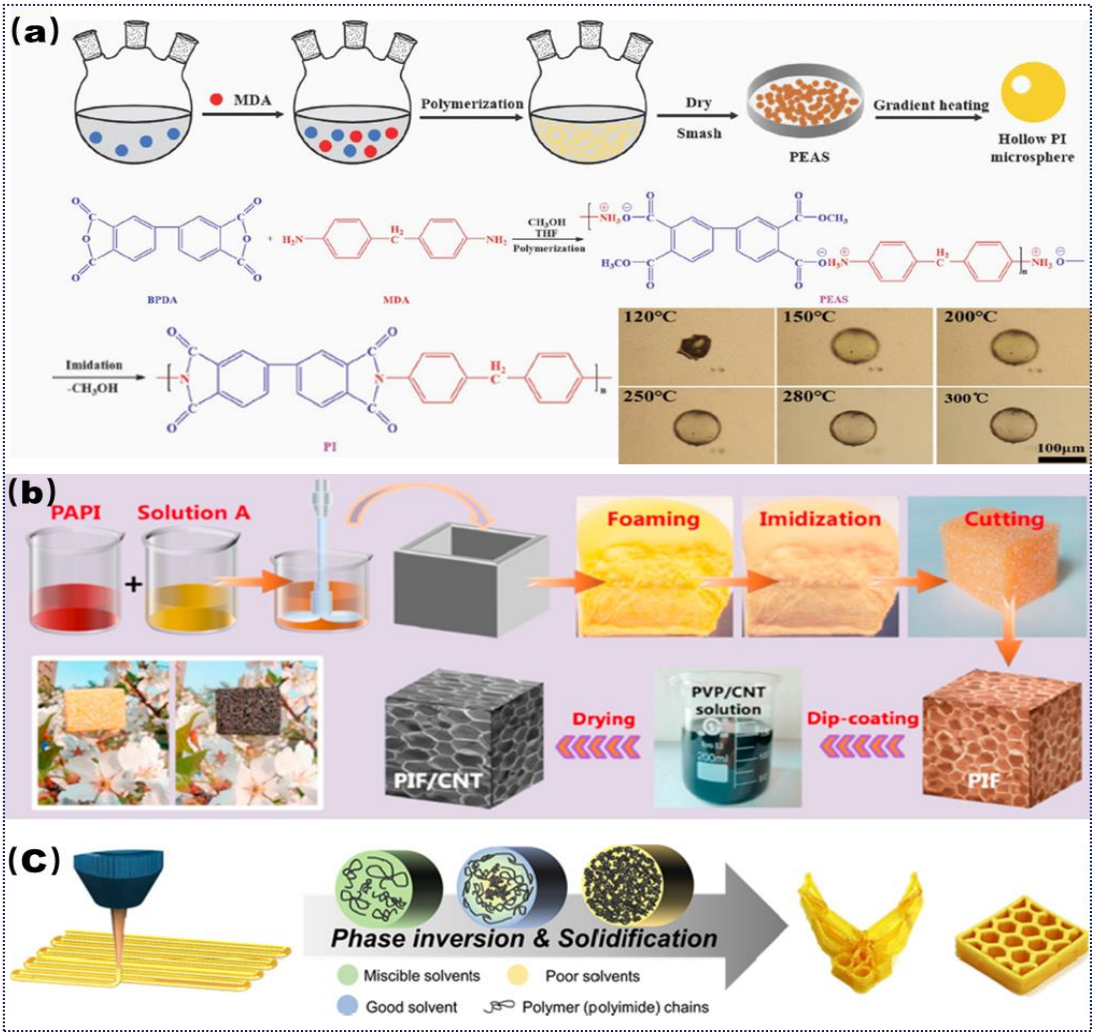

Fig. 8. (a) Schematic illustration of preparing hollow PI microspheres. (b) Schematic illustration for the simple and easy-to-scale fabrication process of lightweight PIF/CNT samples, including the chemical-foaming and solution dip-coating procedure. (c) Schematic of 3D printing and phase inversion process during solidification.

Chemical characterizations of PIFs

PIFs are composed of a polyimide-based skeleton and numerous internal bubbles, its own physical and chemical properties depend on internal structure [115]. From the perspective of physical form, it can be grouped into chemical and biological structures. Generally, the chemical structure of PIFs is determined by the following: (1) the type of monomer that can synthesize polyimide; (2) the molecular structure of polyimide; (3) the kind of foaming agent [61]. The chemical composition and degree of imidization reaction can be characterized by conventional description methods, such as Fourier Transform Infrared (FTIR) spectroscopy, X-ray photoelectron spectroscopy, Raman Spectroscopy, and nuclear magnetic resonance (NMR) spectroscopy. Rui Qin and colleagues [116] arranged PIFs, which were synthesized by BTDA and 4,4-bismaleimiwde diphenylmethane (BDM) in a mixture of THF and MeOH solvent. Subsequently, the precursor powders were prepared for further characterization use. In their study, the FTIR spectra were

carried out in the wavenumber range between 600 and 4500 cm^{-1} to confirm the degree of imidization. As presented in **Fig. 9a–f**, with temperature heating from room temperature (25 °C) to 140 °C, the intensity of absorption peak at 1395 cm^{-1} was rapidly decreased, owing to the O-C=N bond in BDM monomer, indicating the structure of BDM was transforming. Besides, the wavenumber of 1284 cm^{-1} and 1780 cm^{-1} represented the ester groups in PEAS and carbonyl groups in imide rings of polyimide foam, respectively. In the temperature region between 92 °C to 198 °C, the content of ester groups was decreased, and carbonyl groups in imide rings of PI were increased, demonstrating polyimide foam was acquired [62]. Furthermore, the chemical composition and the graphitization degree of porous carbon were also characterized by X-ray photoelectron spectroscopy and Raman spectrum k. Additionally, the polyimide rigid closed-cell foams were fabricated by the thermal foaming of the nadimide end-capped polyimide oligomers (NPOs) powder, which was prepared by the copolymerization of BPDA and an aromatic diamine mixture containing 2-(4-aminophenyl)-5-aminobenzimidazole (BIA) and PDA, as shown in **Fig. 9g–h** [117]. The authors used the ^1H NMR spectra to characterize the chemical structure of the NPO precursor powder. The characteristic signals of $H_1 \sim H_4$ in the end-capping agent were detected in all NPO precursors, along with the motions of C-C on the benzene ring and different diamine groups. On the other hand, the signal at 13.26 ppm was attributed to H_5 in the BIA moiety, and the signal intensity increased with the BIA content increase. These results confirmed the existence of the cis-5-Norbornene-endo-2,3-dicarboxylic anhydride (NA) end-capping groups and BIA moiety in the polymer backbones. Both the FTIR and NMR analyses have ensured that PIFs with different diamine structures were successfully prepared in this article.

Morphologies of PIFs

The properties of PIFs are strongly associated with cellular structure or cell morphologies. It usually refers to the pore size, cell distribution, foam directions, and closed-cell rate [118]. Proverbially, PIFs can be divided into closed-cell and open-cell foams. In closed-cell foam, almost all the foam bubbles are surrounded by a complete hole wall, and the holes are not connected. On the contrary, it is defined as open-cell foam plastic. However, in practice, open and closed-cell structures may appear simultaneously in foams, just happening at different times. For further detail division, sponges, porous films, and aerogels also belong to open-cell PIFs because of their high open-cell rates.

PI closed-cell foams

The melt viscosity of PAA precursors directly affects the cell morphologies and mechanical properties of PIFs. The higher melt viscosity of PAA precursors leads to stricter cell walls, which can quickly form closed-cell PIFs [63]. Generally, rigid closed-cell PIFs possess outstanding mechanical and thermal

properties owing to their high closed-cell ratios. Undoubtedly, the interaction between neighbouring foam cells is detrimental to preparing high-performance and low-density PIFs when the viscosity of PAA precursors is too high. Luo et al. [119] reported that the PAA precursors were synthesized via BTDA and MDA polymerized in a mixture of THF and MeOH solution. Subsequently, the PIFs with a nearly homogeneous spherical closed pore structure were fabricated by thermal forming and imidization. The cell morphologies and wall structures of PIFs are displayed in **Fig. 10a**. It exhibited a very stable and high porosity closed-cell structure. The triangular areas between the foam cells were also relatively smooth and free of defects. The results showed that the compressive strength of PIFs could reach 9.79 MPa. Meanwhile, the thermal conductivities of PIFs fell in a range of 0.071-0.073 W/m/K, which demonstrated promising applications in the fields of aerospace and marine as a high temperature-resistant structural material.

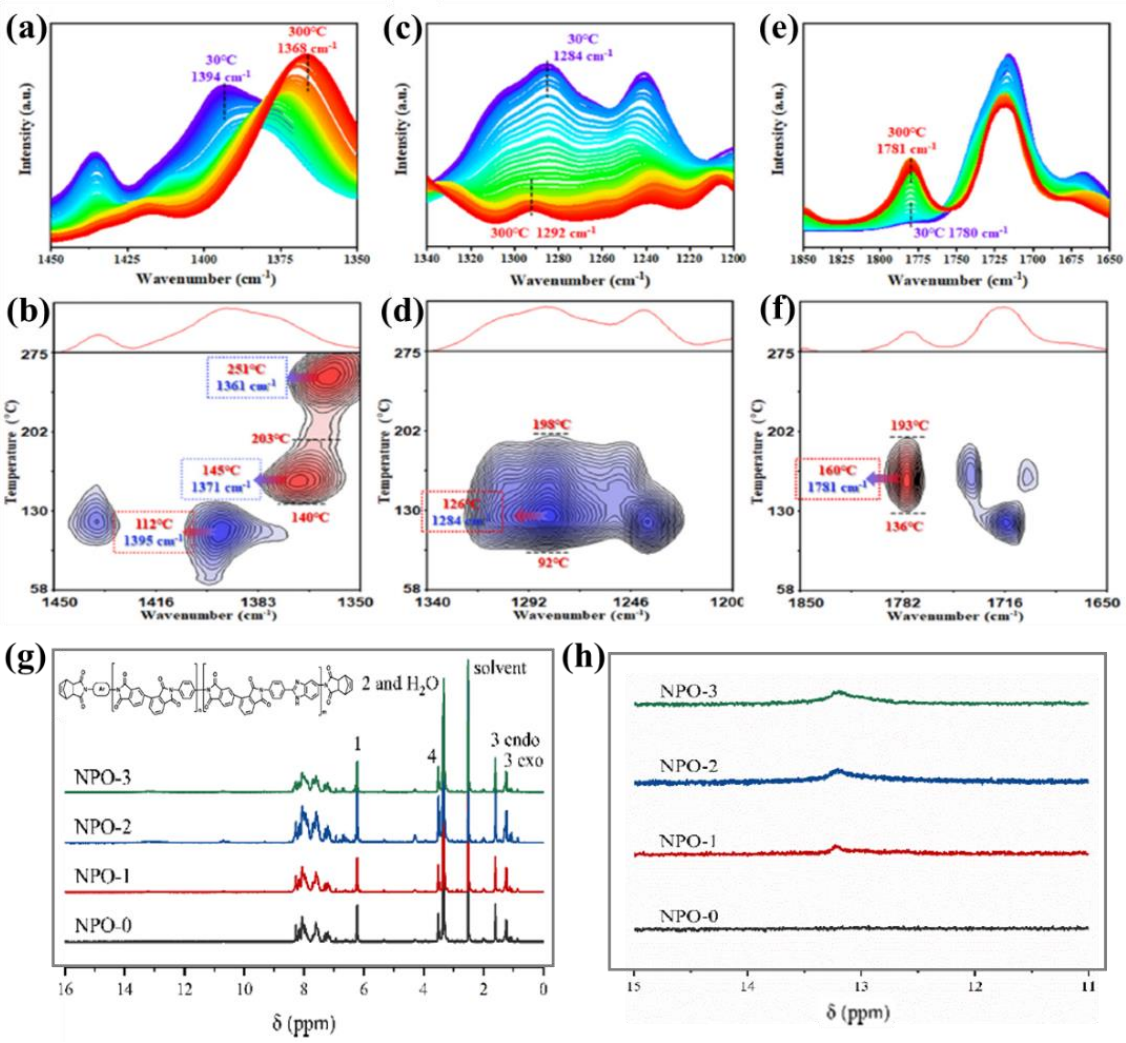

Fig. 9. (a-f) Spectrums of in-situ heating FTIR and PCMW2D of PEAS/BDM (a-b: distinct groups in BDM or BMI, c-f: distinct groups in PEAS or PI). (g-h) NMR spectra of NPOs with different BIA contents: (g: complete spectrum and (h) partial spectrum at 11-15 ppm).

PI open-cell foams

On the contrary, a lower melt viscosity endows better cell wall fluidity and fusibility, which is

beneficial in preparing PIFs with larger cell sizes or open-cell structures due to the coalescence of foam cells. Therefore, PIFs with lower density can be fabricated by the open cell structure and are more widely used in the fields of thermal insulation, sound absorption, and EMI shielding (SE). However, open-cell foam's mechanical properties are lower than closed-cell foam because of its poor melt strength and high open-cell rate. Wang et al. [120] prepared the PAAs by the polymerization of PMDA and ODA with an equivalent mole in a DMAc solution. Afterward, the PAAs was induced thermal forming and imidization to fabricate the CNT/GO/PI foams under a nitrogen atmosphere. It could be found that the foams with an open, uniform, and highly porous structure were fabricated. Further, observation showed the cell wall of the foam was a bit rough (comparison with closed-cell) because of the poor melt strength and different foam processes, as shown in **Fig. 10b**. Unsurprisingly, the resultant composite foam revealed an average EMI SE of 28.2 dB and a specific EMI SE of 7050 dB cm^2 g^{-1} at only 0.02 g/cm^3 density.

PI porous films

PAA precursors are obtained by polymerizing diamine and dianhydride in a polar solvent for most instances. Afterward, the polyimide porous films can be made by casting the PAAs on the substrate surface, and then thermally baking is carried out to form a seal-supporting gelled porous film. Polyimide permeable membrane has excellent potential in filters, thermal insulation, and conductive materials owing to its better filtration absorption and fixed microstructure. However, the complex preparation process and the difficulty of large-scale industrialization are the challenges at this stage. Kwansoo Yang and collegues [121] reported preparing the boron-nitride-(BN)/PI porous films by polymerizing PMDA and ODA in DMAc solvent. Ultimately, the PI porous films were manufactured successfully by freeze-drying foaming; the cellular structure is shown in **Fig. 10c**(left). The results displayed that the porous BN/PI composite films exhibited high thermal diffusivities (0.059-1.033 mm^2/s) and low dielectric constants (2.08-3.48) at 1 GHz for BN contents of 20-80 wt%. In particular, the BN/PI composite films had shallow dielectric loss values. Regardless of the BN content and pressing conditions, the dielectric loss value was close to zero (< 0.002) at frequencies above 10 MHz. Similarly, Xianbo Hou [48] also prepared the PI porous films, in which the cell morphology was different from Kwansoo Yang's work, as shown in **Fig.10c**(right). The results demonstrated that the hierarchical pore distribution could achieve high oil/water selectivity and separation efficiency.

PI sponges

Polyimide sponges with high porosity, low densities, flexibility, and elasticity, have been applied to thermal insulation, sound insulation, and adsorption storage in various fields. Generally, PI sponges are

prepared via two steps: (1) polymerization of diamine and dianhydride to form PAAs precursor/or PI short fiber is directly applied; (2) following by freeze-drying and thermal imidization to obtain PI sponges. Nevertheless, conventionally, the preparation of porous polymer sponges has encountered one of these two limitations: 1) It exhibits poor mechanical properties and/or attained sufficiently low density (below 25 mg/cm^3) [122,123]. 2) Other carbon foams/sponges with many mechanical properties and high porosity are usually synthesized via a series of versatile steps and subject to high-temperature heat treatment (above 700 °C). However, this may not be able to be mass-produced [124]. Jun Young Cheong et al. [31] fabricated PI/carbon nanoparticle sponges by freeze-drying and heat treatment using PI chopped fibers directly. As a result, the minimum density of the PI sponge can reach 0.03 g/cm^3. The image of cell morphology is presented in **Fig. 10d**. The results discovered that the fabricated PI/carbon sponges exhibited highly robust mechanical properties and retained low thermal conductivity even in comparison with pristine PI sponges.

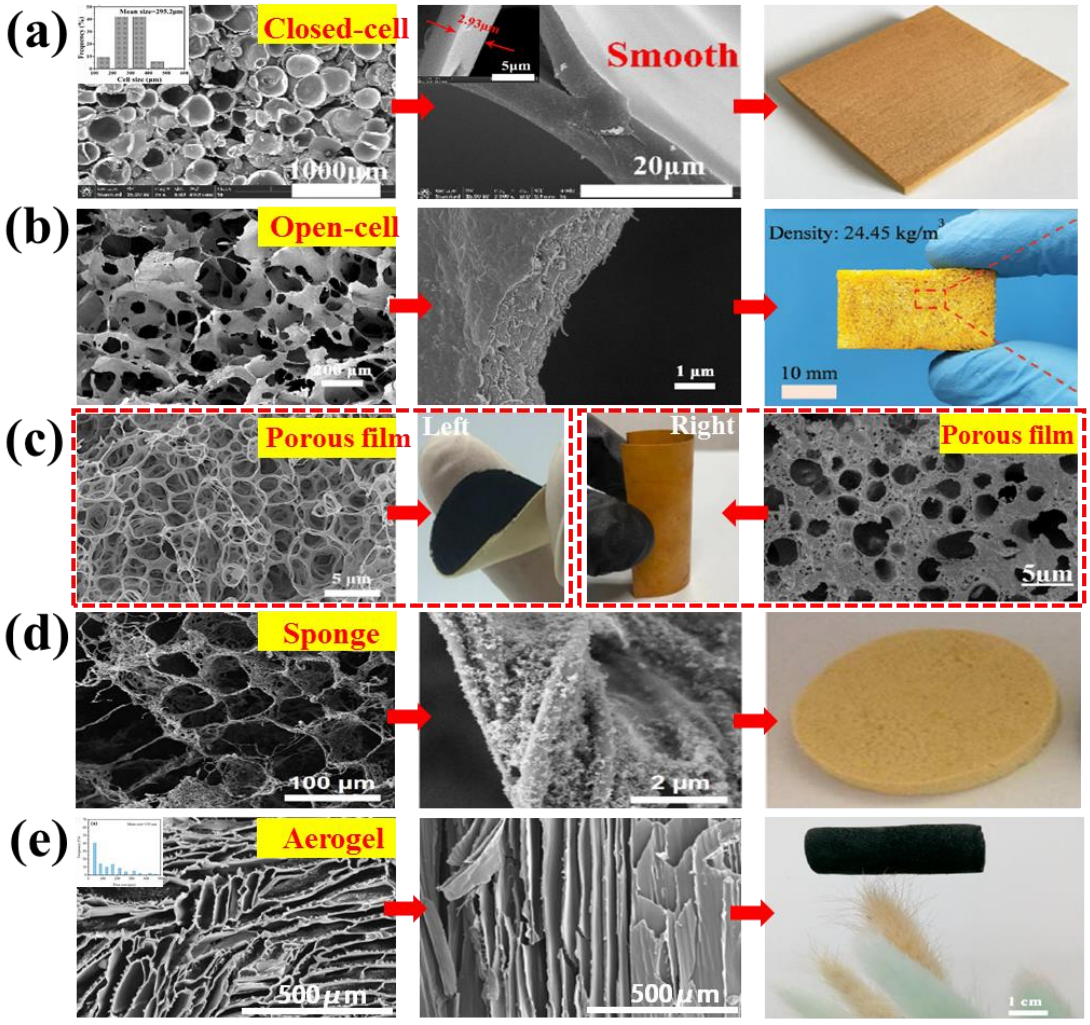

Fig. 10. SEM microstructure images of the pores structure of PIFs. (a) Closed-cell. (b) Open-cell. (c) Porous PI film. (d) PI sponge. (e) PI aerogel.

PI aerogels

Polymeric aerogels have great potential in numerous fields because of their unique combination of thermal and electrical properties and lightweight porous structures. [125] S.S. Kistler of Stanford University first prepared it via the sol-gel method and supercritical drying technology in 1931 [126]. However, polyimide aerogels were reported by chemical reaction of aromatic diamines and dianhydrides until 2004 due to its process complexity and equipment limitations [127]. PI aerogels have held excellent application potential in sound absorption, filter adsorption, oil/water separation, etc., owing to their ultralow density, ultrahigh porosity, and ultralow thermal conductivity [128,129]. As one of the best materials with outstanding comprehensive properties under harsh temperature environments, PI aerogels can play essential roles in the high-tech field in the future [84]. PI aerogels can be divided into linear and crosslinked PI aerogels according to chemical structures. Linear PI aerogels were formed by physical crosslinking among molecules, resulting in significant shrinkage during drying. Linear PI aerogels were first synthesized by Rhine et al. from the U.S. in 2004 [130]. Nevertheless, Linear PI aerogels generally face problems of shrinkage and collapse of pore structure during preparation, resulting in poor macro-structural integrity and mechanical properties. Towards these problems, crosslinked PI aerogels with a three-dimensional network structure by adding crosslinking agents have been widely studied to improve mechanical properties and decrease shrinkage rate during drying compared with linear PI aerogels. Crosslinked PI aerogels were first prepared by Kawagishi et al. at the University of Tokyo by using the non-equimolar ratios of dianhydride and diamine and crosslinking agent 1,3,5-tris (4-aminophenyl) benzene by thermal imidization and supercritical drying in 2007 [131]. In addition, the addition of carbon nanotubes (CNT) and graphene in PI aerogels to increase the conductivity and electromagnetic shielding (EMI) properties of gel had a great challenge and have been widely discussed. Long Ni and colleagues [132] reported the preparation of the crosslinked CNT/PI aerogels with a three-dimensional network structure via the freeze-drying foaming method. Firstly, ODPA and ODA were uniformly dissolved in DMAc solution to obtain PAA precursors, and then the samples were put into a freeze-dryer via unidirectional freezing. Finally, the CNT/PI aerogels were fabricated by thermal forming and imidization at elevated temperatures. As shown in **Fig. 10e**, the freeze-drying and thermal imidization were completed, and the ice crystals were eliminated to form aligned slit-like channel structures and irregular similar honeycomb-like pore structures. The results showed that the CNT/PI aerogel had anisotropic EMI shielding performance (the minimum reflection loss (RLmin) of -52 dB at 12.8 GHz and adequate absorption bandwidth of 6.7 GHz with a thickness of only 2.5 mm in the perpendicular direction to the aligned slit-like channels. In comparison, its RLmin was only -11 dB at 15.9 GHz in the orientation direction of slit-like media) and had remarkable compression performance. There

was hardly any shrinkage or collapse of the pore structure during the preparation.

Multifunctional properties of PIFs

During the past decades, multifunctional properties characterization related to PIFs has become more disparate, and the application fields are more extensive. Herein, the most relevant multifunctional properties characterizations were discussed based on the use of PIFs or their potential applications in high-tech fields. An overview of the central multifunctional properties characterization of PIFs during the studied period included Augustmechanical properties, thermal properties, EMI Shielding, flame resistance, sensing performance, oil-water separation, and others, etc.

Mechanical properties

The mechanical properties are crucial for the practical application of PIFs in modern high-tech manufacturing. Ordinarily, the mechanical properties of PIFs mainly include tensile, compression, flexural, elasticity, and fatigue properties, which are primarily affected by their chemical structure, density, cell structure, test temperature, and many other factors [133]. There are three main strategies to enhance the mechanical and thermal properties of PIFs. One approach is to adjust the foaming process of PAA precursors to boost the density [47]. An alternative strategy is using the inorganic fillers as the reinforcements; Moreover, molecular structure modification has been recognized as another effective method to improve the mechanical and thermal properties of PIFs [134]. Some researchers have also reported another method for preparing PIFs by introducing crosslinking points into the polyimide matrix [135]. We could see from **Fig. 11a** the PI aerogel with a porosity of 96.0 % and density of 21 mg/cm^3 was obtained via the polymerization of ODPA and 1,3-bis (4′-aminophenoxy) benzene with an equivalent mole in DMAc solution, which can stand on a Setaria grass with a lot of burrs. Meanwhile, it also could support a bottle of mineral water with a total weight of 300 g without being damaged, as shown in **Fig. 11b–d**, indicating that the hybrid foam had the load-bearing capacity to a certain extent despite its lightweigh [136]. In addition, the fatigue behaviors of the PI aerogel under a 10,000-cycle compression at different strains were checked, as shown in **Fig. 11e**. The result revealed that the PI aerogel's excellent elasticity and fatigue resistance with a high negative Poisson's ratio at a high compression (<50%) has potential application in many fields. Yang Cheng and colleagues [137] prepared PI aerogels with covalent crosslinking to improve mechanical performance. The PI aerogel exhibited growing mechanical robustness due to the crosslinking points, and the flexible PI network functioned well in endowing the aerogel with compressive resilience [138]. As shown in **Fig. 11f**, the PI aerogel displayed excellent resistance to bending, twisting, and cutting. This means that it could be turned into a large deformation and restored to its original size without damage

or rupture as the external loading was removed. Moreover, benefiting from the excellent mechanical properties above, PI aerogel could be cut into any desired shape by a knife, such as a triangle, hexagon, or forms of star and flower, due to their low shrinkage rate and good structural formability.

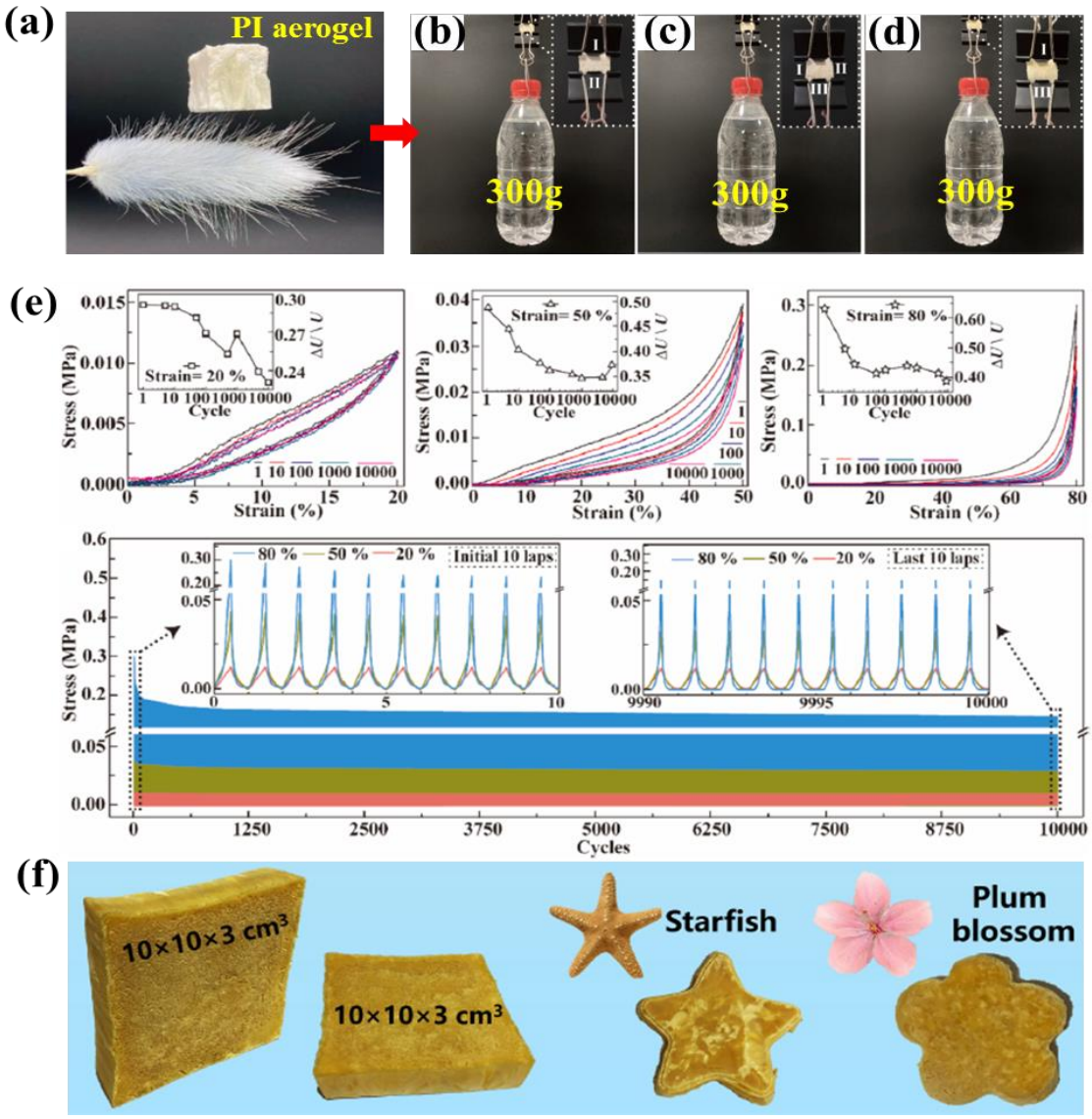

Fig. 11. (a) Intuitive lightweight representation of the PI-based foam on a setaria grass (b-d) Infrared images of the carrying capacity of the PI-based foam. (e) Fatigue resistance of PI aerogels. Loading–unloading curves of 10,000-cycle compression of PI aerogels and stress change in 1–10 cycles and 9900–1000 cycles. (f) The string of digital photos of PI aerogel cutting into starfish and plum blossom.

Thermal properties

PIFs are a perfect thermal insulation material at extremely high and shallow temperatures thanks to their extraordinary physical and chemical structure. The thermal decomposition temperature of PIFs can reach 600 °C, which is one of the most thermally stable foams [139]. Generally, the thermal insulation performance of the foam is characterized by thermal conductivity and thermal diffusion coefficient (the lower the thermal conductivity, the better the thermal insulation effect). Traditionally, there are four heat-

conduction forms of foam materials: (1) heat conduction of solid polymer matrix; (2) heat conduction of gas in foam pores; (3) radiant heat conduction of cell wall and cell gap; (4) thermal convection. Gibson [140] reported that the heat conduction of gas in foam pores contributed the most to the heat conduction of foam. In addition, the main factors affecting the thermal insulation performance of polyimide foam included density [141], cell structure [34] and external factors (such as environment temperature and pressure) [16]. To further reveal the thermal performance of PIFs, the high-temperature resistance and thermal insulation were comprehensively investigated in this chapter. Jing Tian et al. [142] fabricated the PI/silica (PSi) aerogel with a low density of 0.047 g/cm^3 and an ultra-high porosity of 97.5 % via freeze-dried foaming method, which displayed excellent thermal insulating performance with an ultra-low thermal conductivity of 21.2 mW/m K in the radial direction at 25 °C (**Fig. 12a**). The thermal conductivity schematic explanation of polyimide-based aerogel at different length scales during the heat conduction process prepared by unidirectional freezing was demonstrated in **Fig. 12b**. The thermal insulation performance of PSi aerogels was further verified by placing them on a hot stage with different surface temperatures, and a series of thermographic images were recorded. The two typical photos with the stage temperature and the temperature difference between the sample's surface and the hot stage are shown in **Fig. 12c–d**. In addition, the radial thermal conductivities of PSi aerogels under different temperatures and relative humidity were investigated. It could be seen from **Fig. 12e-g** that the PI-based aerogels exhibited a relatively low thermal conductivity despite the high temperature. The polyimide-based foam preserved the original structure, revealing its prominent high-temperature resistance, which made it widely used in aerospace and other fields [143,144]. Similarly, Ruxun Yuan et al. [145] reported the preparation of PI aerogel with the highest porosity of 96.8 % via directional freezing. As shown in **Fig. 12h**, the thermal insulation performance of PI aerogel was assessed by putting it on an open-air hot plate (300 °C) and imaged with an infrared camera. For better visual observation, a flower was placed on the 20 mm thick PI aerogel, and the flower temperature was only 55.4 °C after 10 min, indicating the PI aerogel's excellent thermal insulation performance. In addition, the PI aerogel placed on a Dewar tank with -196 °C was investigated in **Fig. 12i**. The temperature at the top of the PI aerogel only dropped from 24.3 to 21.6 °C after 30 min, suggesting that PI aerogel had excellent heat insulation at low temperatures. Furthermore, the thermal conductivity of the PI aerogel was measured at only 31 mW/m K; it showed not only good thermal insulation but also a high compressive strength compared with other PI-based foam, as shown in **Fig. 12j**.

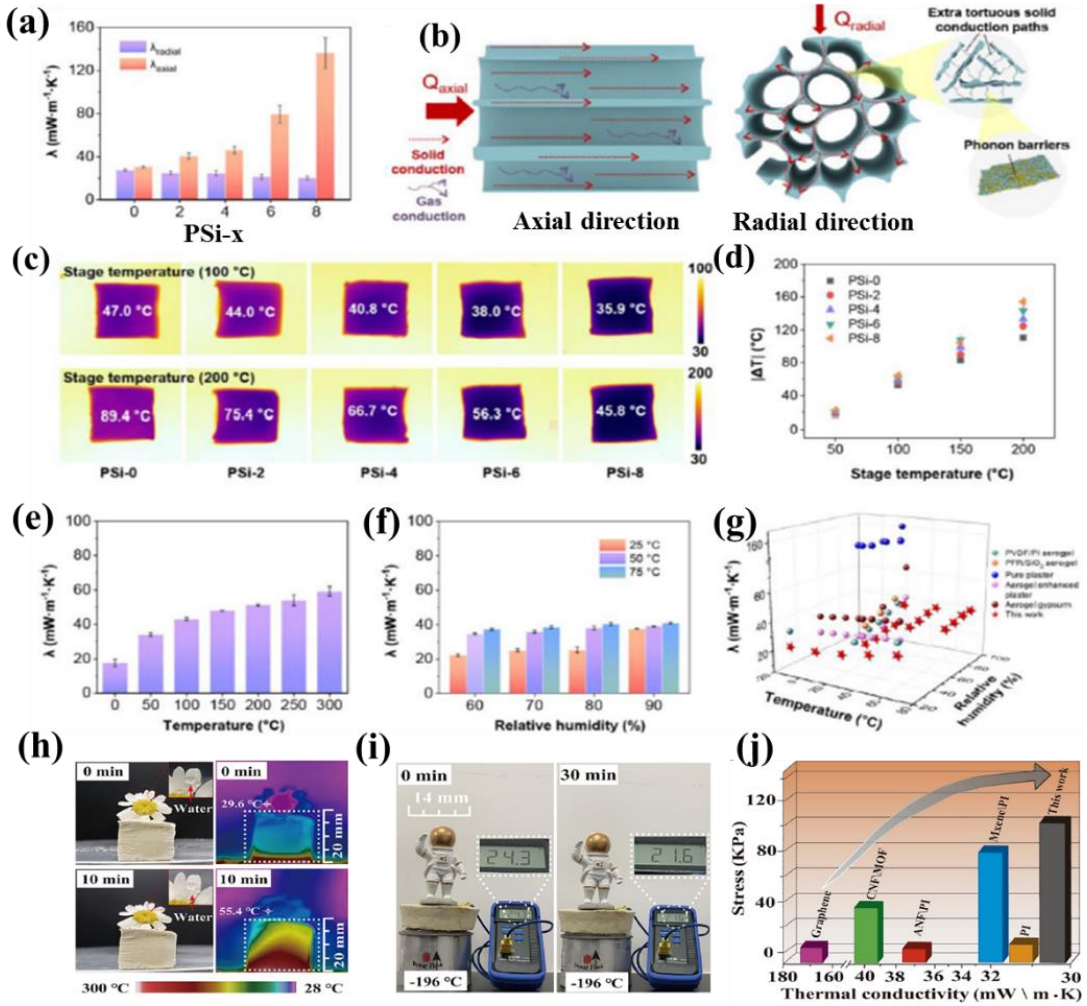

Fig. 12. (a)The thermal conductivities of the PSi aerogels in axial and radial directions. (b)Schematic illustration of the thermal insulation mechanism. (c)Thermographic images of the PSi aerogels on a hot stage. (d)The temperature difference between the surface of PSi aerogels and the hot stage with temperatures ranging from 50 to 200 °C. (e-g) The thermal conductivity of the PSi aerogels at different temperatures and relative humidity. (h-i) Infrared thermal images of the thermal insulation performance of the PI aerogel on a hot and cold stage. (j)Comparison of the compressive strength and thermal conductivity of the PI aerogels.

EMI Shielding performance

High-performance EMI shielding materials with lightweight and superior mechanical flexibility have sparked considerable attention in almost every electronics industry for attenuating electromagnetic radiation of complex electronic systems [146,147]. The mechanism of EMI shielding materials is to limit the transmission of electromagnetic energy from one side of the material to the other side of the space. EMI shielding foams prepared with carbon nanotube, carbon fiber, stainless steel fiber, graphene, metal carbide coated graphene/or nitride as a conductive medium had excellent electromagnetic shielding effectiveness because of the advantages and porous morphology of carbon materials. Generally, the EMI shielding efficiency of high-temperature-resistant PI foam can reach 17-21 dB. However, according to literature reports [148], the electromagnetic shielding efficiency of metal-based PI foam is more than 60 dB, which

will be applied to many high-tech fields. Zhi-Hui Zeng et al. [149] reported a novel type of lightweight, ultra-flexible and sturdy crosslinking transition metal carbide (Ti_3C_2 MXene) (MXenes) coated PI (C-MXene/PI) foams (**Fig. 13a**), which prepared via a scalable dip-coating followed by the chemical crosslinking method. The schematic illustration of the EMI shielding mechanism of the C-MXene/PI foams is shown in **Fig.12b**. The EMI SE performance of pure PI, MXene/PI, and C-MXene/PI foams at the same frequency, EMI SE of the MXene/PI and C-MXene/PI composite foam after stored at 95 % room humidity and 60 °C for different days. The corresponding change of SE was compared to **Fig. 13c–f**. The result demonstrated the potential of the C-MXene/PI foams as high-performance EMI SE materials. In addition, EMI SE of C-MXene/PI foams with various dip-coating layers could be obtained by adjusting the coating layers/times with MXene suspensions. The results are shown in **Fig. 13g**. Compared with other typical porous architectures such as commercial carbon foams, metal-based foams, and other MXene-based porous composites, the EMI SE performance of the C-MXene/PI composite foams was superior to that of others (**Fig. 13h**), which displayed a satisfactory EMI SE effectiveness of 22.5 to 62.5 dB at a density of 28.7 to 48.7 mg/cm^3. The brilliant EMI SE performance of the C-MXene/PI foam, along with the facile and novel manufacturing technology, meant that it had broad perspectives in the next generation of flexible aerospace, electronics, and intelligent devices.

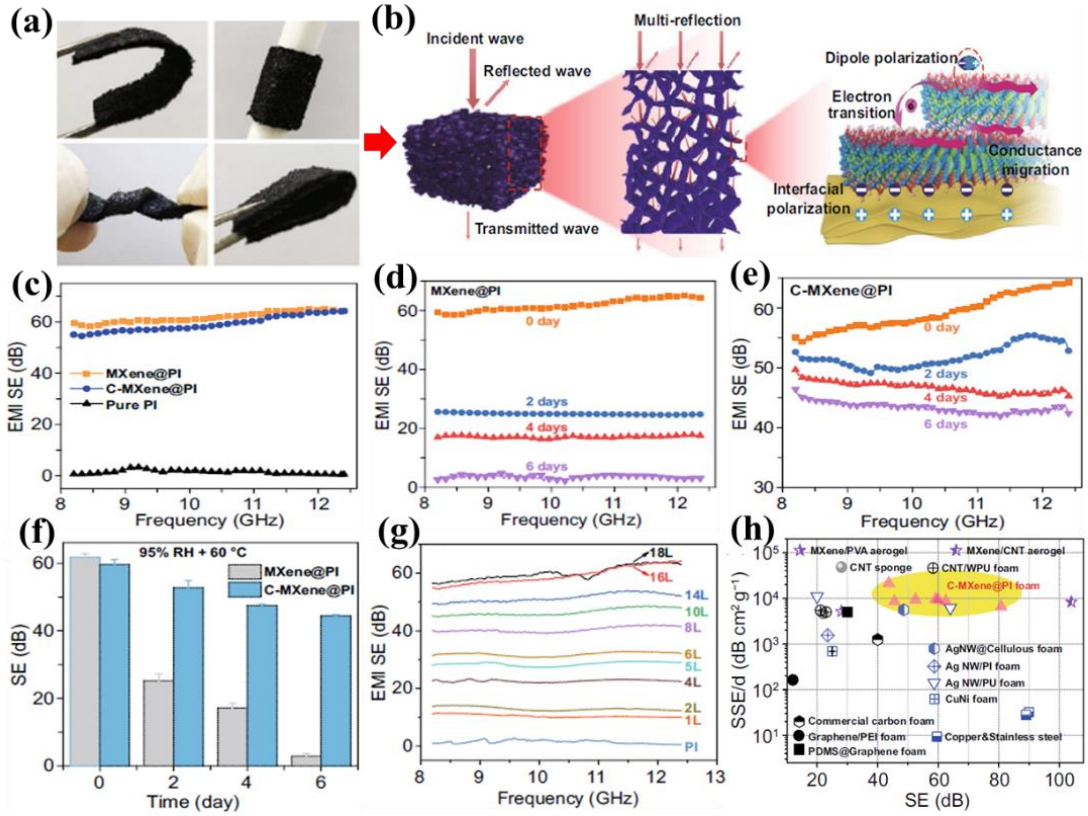

Fig. 13. (a) Photographs of the C-MXene/PI foams. (b)Schematic illustration of EMI shielding mechanism of the C-MXene/PI foams. (c)EMI SE of the pure PI, MXene/PI and C-MXene/PI foams. (d-f) EMI SE of

the MXene/PI and C-MXene/PI composite foams after being stored in a 95% RH environment and a temperature of 60 °C for different days and the corresponding change of SE as a function of time. (g) EMI SE of C-MXene/PI composite foams with various dip-coating layers. (h)EMI SE performance comparison of the C-MXene/PI composite foams with other typical porous architectures.

Sensing Performance

Pressure sensors can convert the received forces into electrical signals and are widely used in artificial intelligence, autonomous vehicles, industrial manufacturing automation, and medical monitoring devices, etc. [150,151]. Based on different working mechanisms, pressure sensors are mainly divided into piezoresistance, capacitance, piezoelectricity, and triboelectricity [152-154], among which the piezoresistive pressure sensors have been proven to be the promising approaches because of their merits of easy processing, simple structure, good flexibility, and convenient signal collection according to literature [155,156]. Conventional piezoresistive sensors are mostly made of metal or conductive polymer/filler materials via appropriate processing technology. They have received serious attention in electronic skin and robots due to their excellent flexibility, wear-resistance, and real-time monitoring ability [157,158]. With the mechanical properties of PIFs, researchers can fabricate sensors derived from polyimide composites [159,160]. Generally, piezoresistive sensors' mechanisms and foams' electrical properties are similar. Both transfer electrical signals by adding conductive particles to form macro-porous conductive network structures [161,162]. Lei Pu and colleagues [162] prepared a radial architecture PI/MXene (RPIMX) aerogel via the interfacial enhancement of PI and radial ice-templating strategy (**Fig. 14a**). Obtained RPIMX aerogel connected in a circuit could be effectively adjusted the brightness of a LED bulb by pressing and releasing (**Fig. 14b**), indicating its great potential in piezoresistive sensors. The cyclic response behavior of the RPIMX composite aerogel under different compression strains, from which stable and reproducible response patterns were observed in **Fig. 14c–d**. The results showed that the RPIMX sensor has significant resistance change in the whole strain range of 0.5-80 %, further verifying its excellent detectability and stability in piezoresistive sensing. In addition, the application of RPIMX aerogel sensor in real-time human motion monitoring and the large motion signals of finger bending are shown in **Fig. 14e–g.** The results indicated that the high sensitivity and wide detection range of the RPIMX composite aerogel made it a prospective wearable sensor for future real-time detection of human motions.

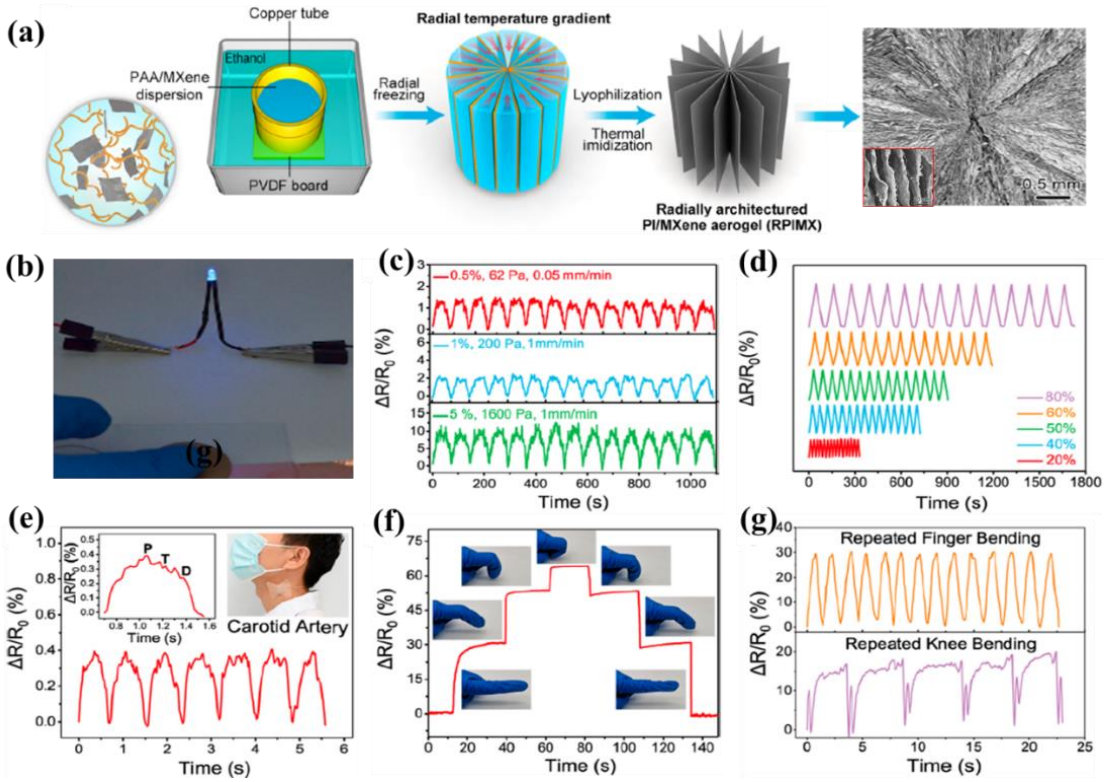

Fig. 14. (a) Preparation process of the radially architectured RPIMX aerogels. (b) A demonstration showing the conductivity of RPIMX composite aerogel during compressing. (c-d) Multicycle response curves of the RPIMX sensor under various compressive strains. (e) Application of RPIMX aerogel sensor in real-time human motion monitoring. (f-g) Large motion signals of finger and knee bending.

Flame retardant performance

Environmentally friendly materials with lightweight and fantastic fire resistance are highly required in aerospace, rail, energy-efficient buildings, etc. [163,164]. However, the widely researched polymer foams confront the problems of weak thermal stability, easy degradation, and flammability, which limits their application for the above domains [165,166]. The flame retardant mechanism includes (1) reducing the flammability of the material in the flame, (2) slowing down the flame-spreading speed, and (3) quickly self-extinguishing when the flame is removed. Various types of flame retardants, for example, hydroxyapatite (HAP), graphene oxide (GO), silicon dioxide (SiO_2), and inorganic layered compounds, have been successfully compounded with polymer aerogels to minimize the flammability of polymer foams. For instance, Zhu et al. [167] fabricated composite aerogel based on a combination of HAP and chitosan, exhibiting excellent fire resistance. However, aerogel density increased dramatically due to a large amount of HAP. Liu et al. [20] prepared PI/GO composites by the in-situ polymerization method. Nevertheless, a large amount of smoke will be generated during the combustion process of PI, which may cause the suffocation of personnel at the fire site. Therefore, PI-based foams with low density, high strength, as well as fire-retardancy are highly desirable for practical applications. For the issues above, Xue and colleagues [168] reported that preparation of layered double hydroxide (LDH)-GO synergistically enhanced PI aerogel

(PLG) with ultralight, outstanding thermal insulation, and excellent flame retardancy was designed and synthesized via a pollution-free freeze-drying method and thermal imidization process. To further explore the flame retardancy of aerogels, the cone calorimetry tests on PI aerogels and PLG-33 (3 wt% LDH and 3 wt% GO) aerogels were conducted, respectively. Corresponding heat release rate (HRR), total heat release (THR), total smoke release (TSR), and CO production (COP) were obtained, as shown in **Fig. 15a–d**. Compared with PI aerogel, the LOI value of PG-6 (6 wt% GO) aerogel, PL-6 (6 wt% LDH) aerogel and PLG-33 aerogel rise to 40 ± 1.3 %, 42 ± 1.1 % and 43 ± 1.2 %, displaying that combustion condition for PLG-33 aerogel is the most demanding (**Fig. 15e**). The TGA curves of PI/GO and PI aerogels are shown in **Fig. 16f**. Through vertical combustion test, the pure PI aerogel burns violently. It exhibits considerable shrinkage after burning (**Fig. 15g**). In contrast, PLG-33 aerogel has excellent self-extinguishing ability in the air and does not produce smoke (**Fig. 15h**). The morphology of PLG-33 aerogel after vertical combustion is shown in **Fig. 15i**. In general, PI-based foam with lower density, great mechanical properties, lower smoke, and excellent flame retardancy will be widely used in the future.

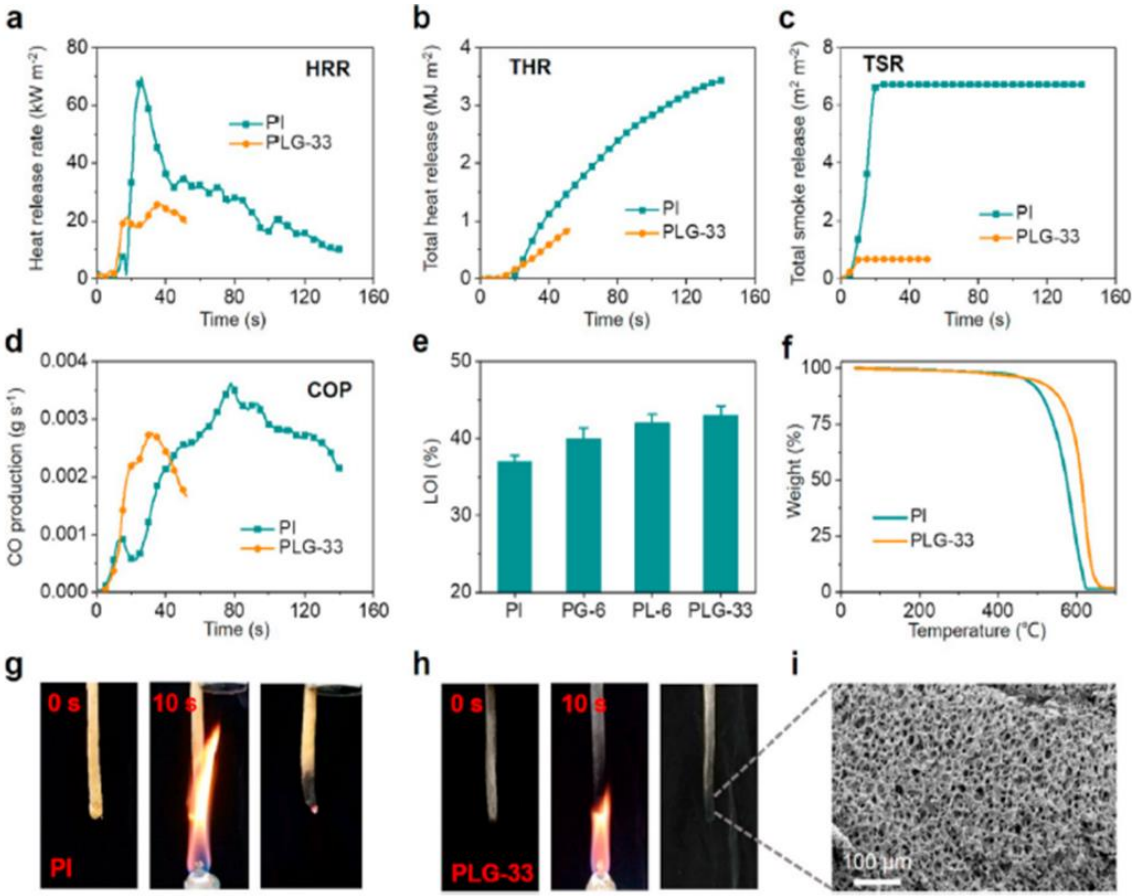

Fig. 15. Flame retardant and thermal stability of PI/GO aerogels. (a–d) Combustion behaviors of PI/GO and pure PI aerogels by cone calorimeter test. (e) The LOI values of aerogels. (f) TGA curves of PI/GO and PI aerogels. (g-h) Digital photographs of vertical combustion of pure PI and PI/GO aerogels in 10 s. (i) SEM image of corresponding PI/GO aerogel after combustion.

Oil-water separation performance

Oil leakage, sewage treatment, and organic solvent discharge have recently generated severe oil pollution in rivers and oceans. These pollutants are difficult to decompose and degrade in nature, threatening the survival of lives in nature and human beings [128,169]. Currently, the methods for oil-water separation include chemical, biological, and physical methods [170,171]. In contrast, filtration methods have drawn significant attention because of their cost effectiveness, low energy consumption, less secondary pollution, and broad applicability [172,173]. In practical application, oil-water separation polymer foam needs not only excellent separation efficiency and brilliant mechanical and thermostability properties but also needs adjustable structure to achieve the separation of oil-water mixtures in different states [174]. The polyimide-based rigid foams hold good mechanical robustness and high-temperature resistance. Nevertheless, it is challenging to achieve oil-water separation due to its small pore size, shallow pore depth, and poor oil absorbency [49,175]. The PI-based sponge and porous film possess the advantages of outstanding separation efficiency and corrosion resistance. However, their poor mechanical properties and oil resistance limit their practical applications. In particular, the PI porous film requires a secondary treatment to achieve the separation function of heavy oil [176,177]. Compared with the above PI-based foam materials, PI aerogels are promising for much oily wastewater separation and filter applications thanks to their inherent ultrahigh porosity, pore volume, large specific area, and good mechanical properties. As shown in **Fig. 16a**, a saline droplet could sit on the surface of the polydimethylsiloxane (PDMS) treated three-dimensional PI/GO sponge, indicating excellent intrinsic hydrophobicity with a contact angle of 159.9 ± 3.1° [178]. Analogical, it could be seen that a water droplet can stand on the surface of the PI nanofiber-based aerogel after close contact as shown in **Fig. 16b** [169]. Surprisingly, an oil droplet penetrated the interior immediately, thus confirming extremely low water adhesion and high oil absorptiveness for the PI aerogel. Apart from water droplets, juice, coffee, and milk droplets could also remain sphere shape on the surface of PI/MXene aerogel, showing the hydrophobicity performance (**Fig. 16c**) [77]. **Fig. 16d** illustrates the adsorption capacity of the PI/MXene aerogel for various oils and organic solvents. The oil absorptiveness was highly dependent on the density and viscosity of liquids, and the weight gain of the composite aerogel ranges from 55.85 to 135.29 times the weight of dried samples. As a result, the PI/MXene aerogel appeared to have a much higher adsorption capacity (**Fig. 16e**). As shown in **Fig. 16f–g**, the composite aerogel could completely absorb a drop of red carbon tetrachloride from the bottom of the water in a short time of 0.12 s without water absorption. Additionally, the diffused soybean oil on the surface of the water could also be well absorbed within 6.55 s. All the cases above indicated that PI-based aerogel manifested its great potential in oil/water separation. Therefore, polyimide-based aerogel has an

incomparable advantage over polyimide-based and other polymer-based foam, which has attracted the close attention of professional researchers.

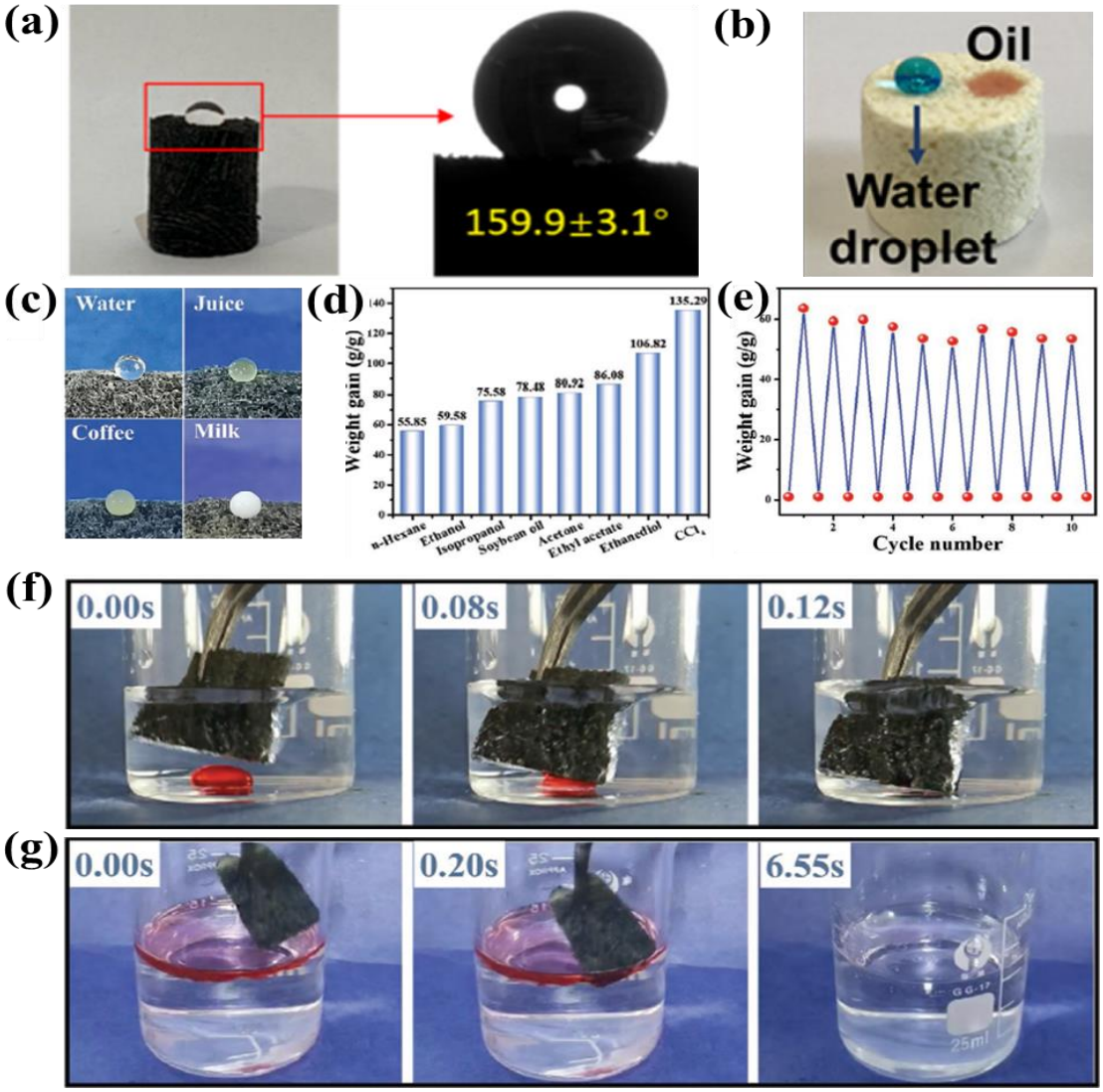

Fig. 16. Digital pictures showing (a) a saline droplet on the PI/GO sponge and contact angle testing. (b) The dynamic measurements of water and oil droplets on the surface of PI aerogel; (c) The state of different liquid droplets on the surface of PINF/MXene composite aerogel. (d) The adsorption capacity of PI/MXene aerogel for various oils and organic solvents. (e) Cyclic adsorption property of the PINF/MXene composite aerogel for ethanol. (f) The absorption of carbon tetrachloride from the bottom of the water and (g) soybean oil from the surface of the water.

Other functional properties and applications

In recent years, many scholars have listed their research, development, and utilization of PIFs as one of the development priorities of new chemical materials in the 21st century. PIFs, thanks to its outstanding characteristics in synthesis and comprehensive properties, has excellent application prospects as both structural materials and functional materials. Overall, in addition to the above functional properties characterization, PIFs also have other excellent functional properties, such as conductivity [174], sound absorption [179], wave absorption [67], radiation resistance [180], bacteriostasis [181] and among others. Although PIFs have the characteristics of high foaming technical barriers and relatively high prices, PI-based foam materials have played an important role in some advanced fields and special environments, including aerospace [182], military [183], microelectronics [184], energy [185], fuel cells [186], sensors [187], protective clothing [188], filters [189], biomedicine [50], marine [190], bionics [191], buildings [192], etc. The first large-scale application of rigid PIFs was on the United States Navy CG-47 frigate missile cruiser. At present, the U.S. Navy has used PIFs as heat insulation and sound absorption materials for all warships and submarines [22,193]. As sound absorption and heat insulation materials, applying PIFs reduces the acoustic radiation of military ships, especially submarines, and improves the stealth performance of ships. As a wave transmitting material, polyimide foam meets the stringent requirements of low dielectric (dielectric constant 1-3, dielectric loss 10^{-3}~10^{-1}), radiation protection and has been widely used in spacecraft radomes and microelectronics [194,195]. More than 15 countries have developed solimide® polyimide foam produced by INSPEC for the thermal insulation and sound insulation system of naval ships. Meanwhile, it is also widely used in civilian ships, such as luxury cruise ships, speedboats, and liquefied natural gas ships [196]. Airbus uses Rohacell® IG and Rohacell® S self-extinguishing PIFs to fabricate ailerons and air-tight diaphragms while using Solimide® flexible PIFs as insulating materials for the cabin interior [197,198]. Solimide® polyimide open-cell foam was developed as a filler on the wheels of "the Sojourner rover" robot to slow down the vibration caused by the rotation of the wheels [199]. PIFs prepared by MIS Technologies Corporation were applied as multilayer heat insulation material for the interior of the space shuttle [200]. The polyimide foam developed by NASA was used in the solar sail of the Hubble telescope as a filled foam, which can reduce the vibration produced by photoelectric solar cells during launch and play the role of buffer protection [201,202]. Boeing applied polyimide foam to the cabin wall of Boeing 767 in the 1990s, and it continues to this day.

Conclusions and prospects

This review study conducted a bibliometric analysis of the polyimide-based foam articles from 1981 to 2022 (August). The results showed that there was a rapid development of polyimide-based foams in the past 40 years. One hundred forty-two research institutions from 20 countries conducted PIFs-related research worldwide. Regarding the countries' impact, China was leading in the number of published articles. In addition, Sichuan University (institution) and Tianxi Liu (corresponding author) from Jiangnan University contributed the most prolific articles. The number of articles published by *ACS APPL MATER INTER* was the highest among 174 different journals. The research institutions from the United States made more contributions to polyimide-based foam before 2010 than other countries. Research categories and the main application areas presented the update of polyimide-based foam theory and practice. Among the 439 retrieved articles, 369 processing and synthesis type articles, 60 application type articles, six reviews, and four 3D printing were included, respectively. The literature co-citation analysis indicated that the knowledge base of the PIFs research field mainly focuses on chemical synthesis foaming, mainly (a) condensation polymerization of dianhydride and diamine in a polar solvent at low temperature; (b) thermoforming and imidization process. Furthermore, the results of the co-occurrence analysis of article keywords showed that PMDA (dianhydride) and ODA (diamine) were the most popular monomers. The academic hotspots were mainly concentrated on studying the reaction mechanism, foaming method, microstructure in the chemical process, multifunctional properties characterization, and application research. Among them, the foaming methods have a specific solution freeze-dried foaming, precursor powder foaming, microsphere foaming, paste foaming, and 3D printing method, as well as their advantages and disadvantages of different preparation methods of polyimide-based foams, which have been systematically discussed above respectively. Researchers and industrial practitioners can choose appropriate preparation methods to achieve polyimide-based foams according to different practical applications. From the perspective of cell morphology, PIFs can be divided into closed-cell foams, open-cell foams, sponges, porous films, and aerogels. Herein, this review study discussed the most relevant multifunctional properties characterization of PIFs based on their potential applications in high-tech fields owing to the high porosity feature of the polyimide-based foams and their inherent characteristics, such as the mechanical properties (tensile and compression resistance, bearing capacity, flexural resistance, elasticity, and fatigue properties), thermal properties (thermal insulation and heat resistance), EMI Shielding, flame resistance, sensing performance, oil-water separation and others

(conductivity, sound absorption, wave absorption, radiation resistance, bacteriostasis).

PIF, which involves research on molecular structure, cell morphology, molding process, functional design, characterization, and visualization in the foaming process, has been one of the research hotspots in the field of high-temperature resistant, lightweight polymer materials in recent years. However, its high cost is the main reason why PIFs have not been produced in different varieties after half a century of development. Committed to the "low-cost technology" research of PIFs, including the low-cost PIFs monomer and foam molding industrialization technology, has always been the top priority. From the perspective of chemicals, the cost of PIFs can be reduced in three ways: (1) looking for novel methods in monomer synthesis and polymerization; (2) developing a new kind of polymer whose properties are slightly lower than those of PIFs but higher than those of the modified polymer via copolymerization or blending; (3) by adopting thermoplastic PIF, it can directly polymerize dihydrogen and diamine in the extruder without any solvent and introduce supercritical physical foaming agents (carbon dioxide, alcohol, nitrogen), which cannot only significantly improve efficiency and reduce costs but also will prevent environmental pollution.

In addition to the above cost-related issues, the research of isomeric PIFs have attracted more attention, and it is referred as "second-generation polyimide foams." Compared with traditional PIFs, the PIFs obtained from the positional isomers of dianhydride and diamine have higher glass transition temperature and lower melt viscosity while maintaining high thermal oxidation stability, outstanding mechanical properties and electrical insulation, and so on. Some isomers also tend to form macrocycles. The research work in this area is only in the initial stage. The synthesis of polyimide and the research on the comprehensive properties of PIFs will make more significant progress shortly. It can be expected that more new polymer materials will be developed from isomeric polyimide. The results showed that the development of the comprehensive technology of PIFs are the fundamental guarantee of realizing its low cost.

Admittedly, there are still many problems to be solved in the research of PIFs, such as developing low-temperature curing methods, synthesizing polyimide-based foam with low density and high performance, high thermal conductivity, and among others. To promote the industrialization development and practical application of PIFs and solve many critical technical problems, it is imperative to summarize and refine the primary data and information on PIFs. With further improvement of synthesis technology and processing technology, as well as the substantial cost reduction, PIFs with its superior

performance will occupy a more prominent position in the field of composite materials. In the end, this review study conducted a state-of-the-art review of the research on PIFs, as well as provide information on their technological developments, prospects and opportunities, which will be helpful for other researchers interested in PIFs research.

CRediT authorship contribution statement

Haiming Liu: Conceptualization, Data curation, Formal analysis, Methodology, Investigation, Methodology, Writing –original draft, and, Writing – review & editing. **Xiangdong Wang**: Conceptualization, Supervision, Validation, Writing – review. **Maxwell Fordjour Antwi-Afari**: Validation, formal analysis, Writing – review & editing. **Haoyang Mi**: formal analysis, Investigation, Supervision, Validation, Funding acquisition, Writing – review & editing. **Chuntai Liu**: Investigation, Supervision, Validation, Funding acquisition, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgments

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