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Sciento-qualitative Study of Zinc-iodine Energy Storage Systems

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Abstract: Zinc-iodine batteries have gained attention recently as promising energy storage systems (ESSs) due to their high energy density, low cost, non-toxicity, and environmental friendliness - making them a favorable alternative to conventional energy storage systems. Even though literature abounds on zinc-iodine batteries, very few studies have attempted to map the global research on Zinc-iodine ESS to discover geographical collaboration networks and research trends. This sciento-qualitative study analyzed the research trends in zinc-iodine ESS using data from Scopus on zinc-iodine ESS. By this method, the most frequently occurring keywords, authors, papers with the most citations, and states that have made the greatest contribution to the field of zinc-iodine ESSs were examined. A total of 161 articles spanning 2003-2023 were analyzed and a total of 8 publications materialized between 2003-2008, making this period an emerging phase; followed by the tempo phase from 2009 to 2016 (a total of 26 publications), and the rapid growth phase from 2017 to 2023 having an exponential increase (R^2=0.6608) of 127 publications. China has the most publications, citations, and cross-continental collaborations. Aside from the Department of Chemistry at Ku Leuven in Belgium, all the top institutions researching zinc-iodine ESSs are in China, with Shandong University's Key Laboratory for Colloid and Interface Chemistry having the most publications and citations. More so, recent progress in the fabrication of zinc-iodine ESSs focusing on anode and cathode materials, electrolytes, and device architecture has been presented. Finally, the challenges and opportunities for the development of zinc-iodine batteries are discussed, highlighting the need for further optimization of electrode materials and device architecture to achieve high efficiency, long cycle life, and cost-effective production. This sciento-qualitative study will help researchers find institutions of interest for collaborations, create new alliances, share ground-breaking technologies, and give important direction for the advancement of zinc-iodine ESS.

Keywords: Zinc-iodine batteries; Energy storage; Electrochemical performance; Anode; Cathode materials; Electrolytes; Device architecture

1. Introduction

Energy storage technologies that are more effective, economical, and ecologically benign have attracted increasing attention in recent years [1-4]. Zinc-iodine batteries have emerged as a viable alternative to existing energy storage systems due to their high energy density, low cost, and sustainability [5, 6]. Voltage production in zinc-iodine batteries is ascribed to the variation in the redox potentials between the zinc and iodine species, which drives the flow of electrons in the external circuit, generating an electrical current that can be harnessed for practical applications [7]. Zinc-iodine battery performance and stability have recently seen considerable increases thanks to new developments in the synthesis and characterization of electrode materials, electrolytes, and device designs [6, 8]. The problems with using iodine in batteries, such as corrosion, limited solubility, and cathode passivation, have been researched extensively leading to the development of novel materials, such as zinc and carbon-based anodes and cathodes [8, 9]. Despites the significant strides, the limited solubility of iodine in the electrolyte, iodine shuttle effect, dendrite formation, and corrosion of zinc anode during cycling pose a significant challenge, which leads to the degradation of battery performance [10-12]. During discharge, zinc ions are oxidized at the anode, releasing electrons and forming zinc oxide, and at the same time, iodine is reduced at the cathode, consuming electrons and forming solid iodine resulting in a voltage [13].

To address this issue, various approaches have been proposed to develop more stable and corrosion-resistant electrode materials [14-17]. For example, carbon-based cathodes using polyaniline (PANI) as a precursor material were synthesized and the resulting cathodes exhibited high capacity and stability, with a capacity retention of over 90% after 100 cycles [18]. Another approach to improving the performance of zinc-iodine batteries is the use of zinc-based anodes. In this regard, the synthesis of zinc nanoparticles using the wet chemical method has been reported as anodes for zinc-iodine batteries and the resulting anodes exhibited good stability and high discharge capacity, with a capacity retention of 83% after 50 cycles [8].

Additionally, various strategies have been proposed to improve the stability and durability of the electrolyte and the device architecture of zinc-iodine batteries, such as the use of flow batteries and hybrid systems [19]. The performance of zinc-iodine batteries is also heavily influenced by the composition and stability of the electrolyte. One of the challenges in the development of electrolytes for zinc-iodine batteries is the low solubility of iodine in the electrolyte, which limits the capacity and stability of the battery [6]. To address this issue, various strategies have been proposed to increase the solubility of iodine in the electrolyte. One approach is the use of iodine complexes, such as 1,3-diethyl-2-thiobarbituric acid (DETBA), as additives in the electrolyte [20]. Another approach is the use of ionic liquids as electrolytes [21] [22]. The use of nanomaterials or nanoporous carbon electrodes, which can provide a confined space for iodine molecules to interact with the electrolyte has been investigated extensively [23, 24]. This confinement slows down the dissolution of iodine into species that could cause self-discharge, such as triiodide and pentaiodide. Nanoporous carbon electrodes aid in better filling of iodine in the pores, thereby increasing the capacity of the iodine-based energy storage system [24]. This strategy is effective in improving the solubility and stability of iodine in nanoporous carbon electrodes. Furthermore, electrode and electrolyte regulation can be employed to promote the coulombic efficiency and stability of iodine-based energy storage systems [25]. By controlling the rates of individual reaction steps in the electrochemical oxidation/reduction of iodide/iodine, the stability of iodine and iodide species in the electrolyte can be enhanced. This can be achieved by carefully selecting the electrode and electrolyte materials and optimizing their compositions [15].

The design and optimization of the device architecture also play a crucial role in the preparation of zinc-iodine batteries. The novel strategy adopted lately is the use of flow batteries, which can improve the stability and efficiency of the battery by separating the electrode and electrolyte compartments [26]. Flow batteries use two separate electrolyte solutions stored in external tanks. The electrolyte solutions are pumped into a reaction chamber, where they undergo an electrochemical reaction that generates electricity. Unlike conventional batteries, the energy storage capacity of a flow battery is not determined by the size of the electrodes, but by the size of the external electrolyte tanks [27]. This makes flow batteries highly scalable and adaptable for various applications, such as grid-scale energy storage, backup power for critical systems, and electric vehicle charging. Flow batteries have several advantages over other types of batteries, including the ability to decouple the power and energy capacity, long cycle life, and high efficiency [28, 29]. However, flow batteries also have some limitations, such as high cost, low energy density, and ultimately not portable enough for certain applications [27, 30]. Research is ongoing to address these limitations and improve the performance and cost-effectiveness of flow batteries for various applications. For example, a zinc-iodine battery has been development with the potential to improve the stability and efficiency of zinc-iodine ESS [31]. Another approach in design optimization involves the use of hybrid systems [32], which combines the advantages of different energy storage technologies to improve the performance and efficiency of zinc-iodine batteries [33]. In this regard, a lot of insightful reviews have been conducted in recent times [5, 8, 34, 35] to consolidate the knowledge.

In essence, recent advances in the synthesis and characterization of electrode materials, electrolytes, and device architectures have led to significant improvements in the performance and stability of zinc-iodine batteries, however, zinc-iodine ESS is still at its infant stages, hence, the need for a more focused research collaboration and alliances to expand the frontiers of knowledge. Although several studies on zinc-iodine batteries have been carried out [5, 8, 36, 37], little is known of a study that statistically maps the global research of Zinc-iodine ESS, providing insight into geospatial partnerships and research trends. Therefore, a *sciento-qualitative* study that chronicles the current state-of-the-art zinc-iodine ESS research in different countries and organizations worldwide is required to help predict future trends for the formation of research alliances and collaborations.

This *sciento-qualitative* study provides insight into research trends in zinc-iodine ESS over the last two decades, considering the top contributing authors and institutions, top contributing countries, and the intra and intercontinental collaboration as well as the direction of research over the last few years. It also highlights the challenges and opportunities in the development and commercialization of zinc-iodine batteries, including their scalability, cost-effectiveness, and environmental impact.

Research significance

Extensive research has been conducted in the past few years in energy storage systems for diverse applications, and some valuable conclusions have been reached [38-41]. In ESSs, the majority of review studies have been conducted manually, which is time-consuming and subject to biases due to the influence of data selection and interpretation of literature [41]. This article reviews the recent advances in zinc-iodine energy over vast bibliometric data using a combination of and meta-synthesis based on themes identified by Bibliometrix (biblioshiny) software. This strategy ensures the validity and reliability of results while also obfuscating the procedure for researchers and practitioners [42]. Researchers from a variety of geographical locations may benefit from the graphical representation of results based on a scientometric review when forming research alliances, forming joint ventures, and sharing ground-breaking technologies and ideas. Additionally, this study discusses the most active research areas and sustainability aspects related to zinc-iodine ESS and their large-scale production. The study also discusses the challenges associated with the application of zinc-iodine ESSs for wearable technologies and autonomous energy systems. Based on the assessment of the literature, the challenges and opportunities in the development and commercialization of zinc-iodine batteries, including their scalability, cost-effectiveness, and environmental impact have been presented, which makes it easy to see future research directions.

2. Methodology

Scientometric analysis of the bibliometric data on recent advances in zinc-iodine energy storage systems is presented. The primary reason for developing a scientometric review method is that subjective analyses of engineering studies conducted by researchers are prone to error [43]. Scientometrics produce a more rational and less skewed result when used alone, as it is unaffected by any individual's perspective. However, scientometric reviews often lack the contextual information necessary to interpret the meaning and significance of quantitative metrics, hence the combination of this approach with a qualitative literature study to provide a full spectrum of zinc-iodine ESS. This study summarizes and synthesizes the research conducted over the last two decades. The study quantifies research progress through the use of maps and connections between bibliometric data, resulting in a quantitative assessment, whereas a critical qualitative analysis was done based on the trend analysis generated from biblioshiny of the Bibliometrix software. Due to the volume of documents published on the current subject topic, it is vital to choose the most accurate database. The two most effective, comprehensive, and objective databases for conducting literature searches, especially for ESS are Scopus and Web of Science [44]. Scopus has broader bibliometric coverage and more up-to-date data than Web of Science [45-47]. Therefore, the Scopus database was used to compile the bibliometric data for the current analysis of recent advances in zinc-iodine ESS. The bibliometric data search in the Scopus database was conducted from the year 2003 to 13th May 2023. The searched keywords in Scopus were "zinc-iodine batteries", "zinc-iodine energy storage", "zinc-iodine battery architecture", "electrolytes for zinc iodine and "cathode anode materials for zinc-iodine battery". The document results for each search keyword are shown in Table1. Data refinement options were used to eliminate irrelevant articles. Only "articles" and "review" were selected from the "document type" drop-down menu. Also, only "journal" was chosen as the "source type", while "English" was chosen from the "language". Fig. 1 indicates the list of approaches in data analysis.

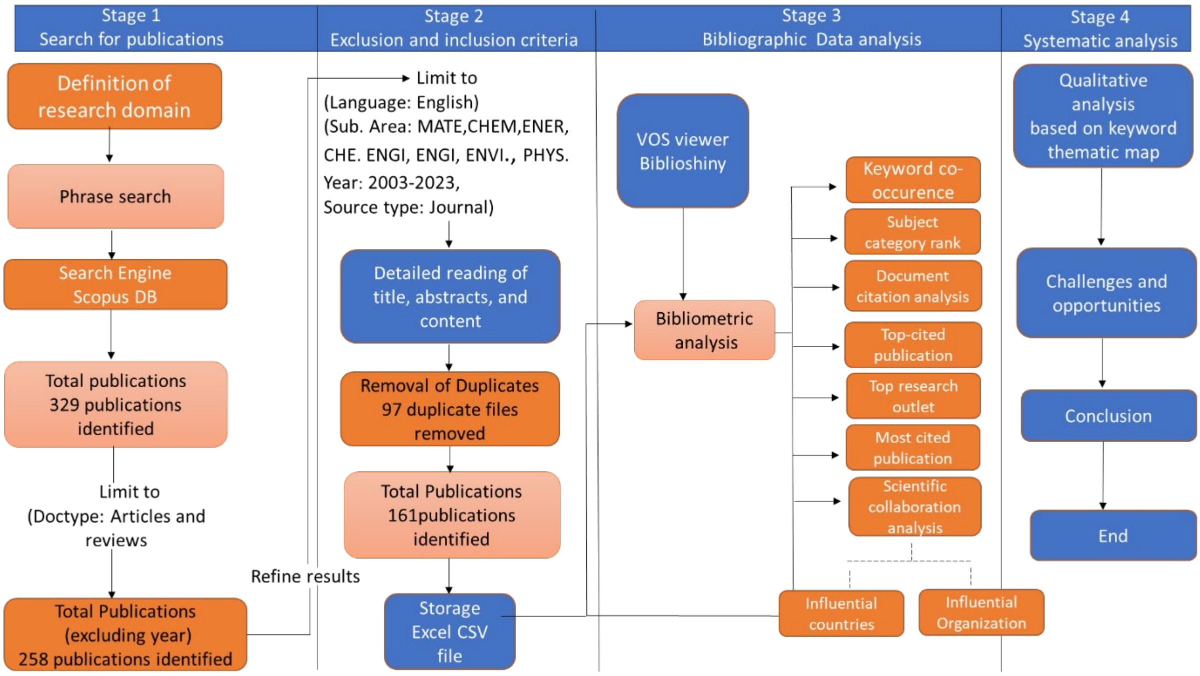

Fig. 1. Scientometric review approach towards data analysis

Before the analysis, three different files were used; the first consisted of a general search, and the second involved sorting of the files under the set parameters described above (see Fig.1). The third file involved sorting of the second by eliminating document duplicates made possible by the custom sort of feature in Ms Excel, using the sort by “title” and the sort order set to “A-Z”. The documents were carefully scrutinized, and duplicate files were removed. After applying these filters, the resulting documents 161 in total were compiled, saved, and used for analysis in the VOSviewer software developed by Nees Jan van Eck and Ludo Waltman [48], and biblioshiny by Massimo Aria and Corrado Cuccurullo [49] for scientific mapping and visualizations. Scientometrics circumvents the difficulties encountered by researchers conducting manual reviews and establishes a connection between sources, keywords, authors, articles, and countries within a given research area. The science mapping and visualization were created using VOSviewer (version:1.6.16). The analysis was carried out using VOSviewer, with the "type of data" parameter set to "create a map from bibliographic data" and the "data source" parameter set to "read data from bibliographic database files". Scopus CSV files were analyzed in a few simple steps while maintaining data consistency and reliability. As part of the science mapping review, the sources of articles, the most frequently occurring keywords, the most cited authors and articles, and country/regional participation were analyzed. Maps were used to illustrate the various parameters, their relationships, and co-occurrence, while tables summarized their numerical values.

Table 1. Documents searched in Scopus database and resulting documents as of May 13, 2023.

S/N	Searched key phrases	Total documents results	Documents results after applying filters (Limit to articles and reviews)	Documents results after applying all filters
1.	Zinc-iodine batteries	79	74	22
2.	Zinc-iodine energy storage	49	46	41
3.	zinc-iodine battery architecture	3	3	3
4.	electrolytes for zinc iodine	193	131	91
5.	cathode "AND" anode materials for zinc-iodine battery	5	4	4

3. Results and discussions of scientometric data analysis

Annual publication trend

In terms of the quantity of publications, their growth trajectory, and the geographical distribution of authors, simple descriptive statistics are ideal for illustrating this phenomenon [50]. The examination of the number of papers published each year reveals a pronounced output of scholarly works, followed by a stable and exponential growth in the two decades under review (Fig. 2). As shown in Table1, the searched keywords "electrolytes for zinc iodine" resulted in most documents (131), while "zinc-iodine battery architecture" resulted in the least documents (3). There were 8 publications in total from 2003 to 2008, making this time an emerging phase for Zinc-iodine ESS. Following that, a gradual increase in the number of publications was seen from 2009 to 2016 (a total of 26 publications), and this time may be referred to as the pick-up or tempo phase. However, from 2017 to 2023, there was an exponential increase (R^2=0.6608) of 127 total publications, and this may be considered a time of rapid growth phase in Zinc-iodine ESS research. It is interesting to note (see Fig. 1), how research on zinc-iodine energy storage systems has increased exponentially over the last four years. This phenomenon could be caused by the perception that zinc-iodine energy storage devices provide a promising alternative solution for energy storage potential compared to the existing technologies [8]. The rapid growth phase is a piece of evidence that scientists are actively researching and expanding the frontiers of knowledge in this particular field, likely in response to the demand for more efficient and sustainable energy storage solutions - consistent with UNDP SDG goal 7 [51].

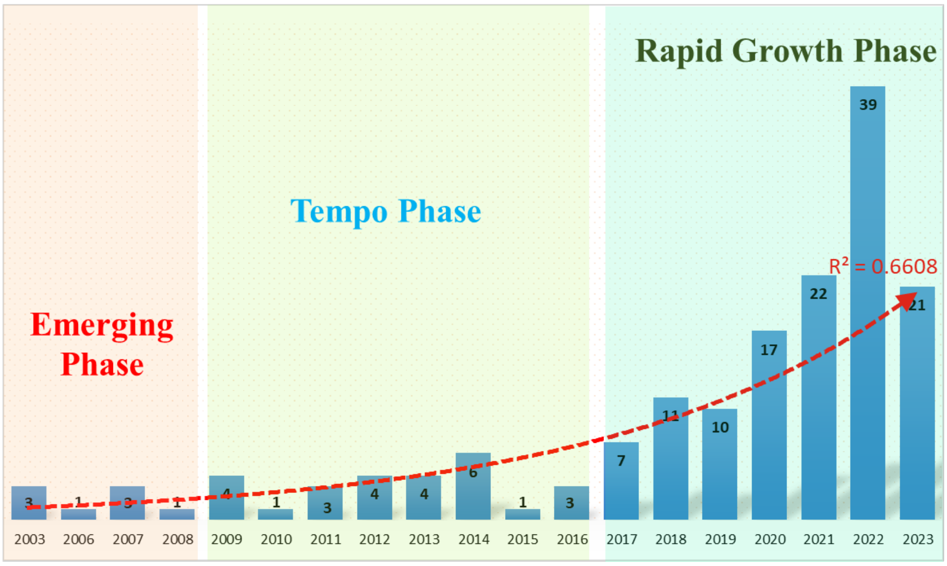

Fig. 2. Annual publication trends of articles

In recent years, the improvements in efficiency, performance, and scalability in Zin-iodine ESS may have practical implications for energy storage applications because they hold promise for addressing the challenges associated with conventional energy storage systems, such as improved efficiency, cost-effectiveness, and most importantly, meeting sustainability requirements.

Top publication sources

Based on filtered data from Scopus, the analysis of top publication sources was done using the VOSviewer. "Bibliographic coupling" was chosen as the "analysis type," and "sources" was chosen as the "analysis unit". A source's minimum number of documents was set at 2, and 22 of the 85 sources met this condition. The largest group of linking items among the 22 that met the criteria was 17, as shown in Fig. 3. Table S1 lists the top sources and journals that registered at least two publications together with their citation counts and overall link strength. The node size corresponds to the contribution of the journal's article count; a bigger node size suggests a greater contribution. Typically, ACS Applied Materials and Interface, Energy Storage Materials, and Energy and Environmental Science have bigger nodes than the other journals, indicating their higher impact in the present research area. This suggests papers in Zinc-iodine ESS are published in these journals than the others within the same cluster. Furthermore, nodes (sources/journals) of the same color show clusters of connected journals detected using VOSviewer analysis. Three clusters have been observed shown by red, green, and blue. Cluster 1 (red) comprised 8 items, cluster 2 (green) contained 7 items, and cluster 3 (blue) had 2 items. These clusters are groups of closely related research articles or terms that share common characteristics or themes. These clusters are formed based on the strength of co-occurrence relationships between articles published in these journals. It is therefore, evident that articles within cluster one (1) tend to have higher co-occurrence frequencies with each other and these articles are published mainly by ACS Applied Materials and Interface, Energy Storage Materials, Journal of Alloy and Compounds, Nature Communications, Journal of Colloids and Interface, and Current Opinions in Electrochemistry with other two Journals not listed in this category. In the same manner, articles in cluster 2 show relatively good occurrence relationship marginal differences in their node size, especially the top five journals in this cluster comprising Energy and Environmental Science, Angewandte Chemie-International Edition, Advanced Materials, and Journal of Power Source. The clusters with higher co-occurrence frequencies indicate that certain research topics or themes are more prevalent or dominant and hence are published mainly in journals within that cluster. This implies that editors of these journals within a particular cluster may have an active interest in studies that explore similar topics. More so, the strong links between journals of a particular cluster suggest a strong interconnectedness and collaboration among researchers within these domains, which may foster the exchange of ideas, sharing of methodologies, and collective efforts to address the research challenges.

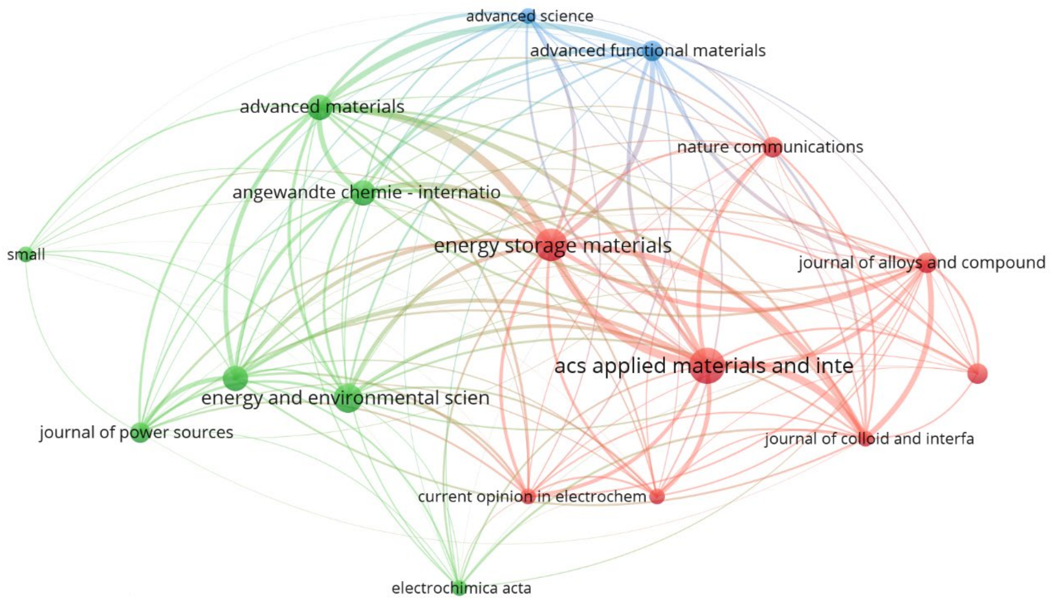

Fig. 3. Science mapping visualization of top publication sources.

Frequently occurring keywords

Keywords are important research metrics because they identify and represent the research domain’s fundamental field [52]. For this analysis, "co-occurrence" was chosen as the "kind of analysis," and "all keywords" was chosen as the "unit of analysis" in the VOSviewer. To avoid duplicate keywords, sorting was done in Excel by replacing repeated words with a single identification and analyzed as a Thesaurus file resulting in 28 keywords. In this scenario, 27 out of the 28 new items were largely connected. The 27 keywords that appeared frequently in research articles in the current subject field are summarized in Table S2 with labels containing an asterisk (*), describing removed words due to their wide gap to the study topic. Zinc-iodine battery, dye-sensitized solar cell, iodine, zinc, and energy storage devices are the top five most commonly occurring keywords. Fig. 4a shows the co-occurrence visualization of keyword networks. It can be observed that the keywords have larger nodes than the others, implying their relevance in the study of zinc-iodine batteries. Clusters of keywords have been colored differently in the network to highlight their co-occurrence in distinct publications. The colors red, light green, blue, yellow, blue, violet, red, and deep green were used to identify eight clusters. Cluster 1 (red) contained 5, cluster 2 (light green) 5, cluster 3 (blue) 4, cluster 4 (yellow) 3, cluster 5 (blue) 3, cluster 6 (violet) 3, cluster 7 (red) 2 and cluster 8 (deep green) has 2 keywords. Additionally, to analyze the most prominent keywords over the last five years of Zinc-iodine ESS research, the overlay visualization function was used to examine the yearly trends in color codes and node sizes, with the biggest node sizes representing the most emphasized keyword in zinc-iodine ESS research. Cyclic voltammetry and dye-sensitized solar cells were the predominant keywords in 2018 with a gradual shift towards flow batteries, zinc, iodine, and aqueous batteries, and then to energy storage devices around 2019 to mid-2021. Zinc-iodine batteries had an exponential rise between the years 2022 into 2023 as shown in Fig. 4(b) occurring 29 times with a total link strength of 29 followed by iodine occurring 7 times with a link strength of 7. Other associated keywords, such as heteroatom doping, oxygen-containing functional groups, and electrolyte additives also featured weakly in the most active keywords used in 2022-2023. The prominently highlighted keywords typically suggest areas of active research and scientific interest. In comparison to the weakly featured keywords, it indicates that the prominently featured keywords/topics have received significant attention in recent years and are likely developing or established research trends in the 2022-2023 period. In terms of node size average normal citation, 2022-2023 received a boost average normal citation of 1.4, thus 50% compared to 2021, 2020, 2019, and 2018, recording 1.2, 1.0, 0.8, 0.6, and 0.4, respectively.

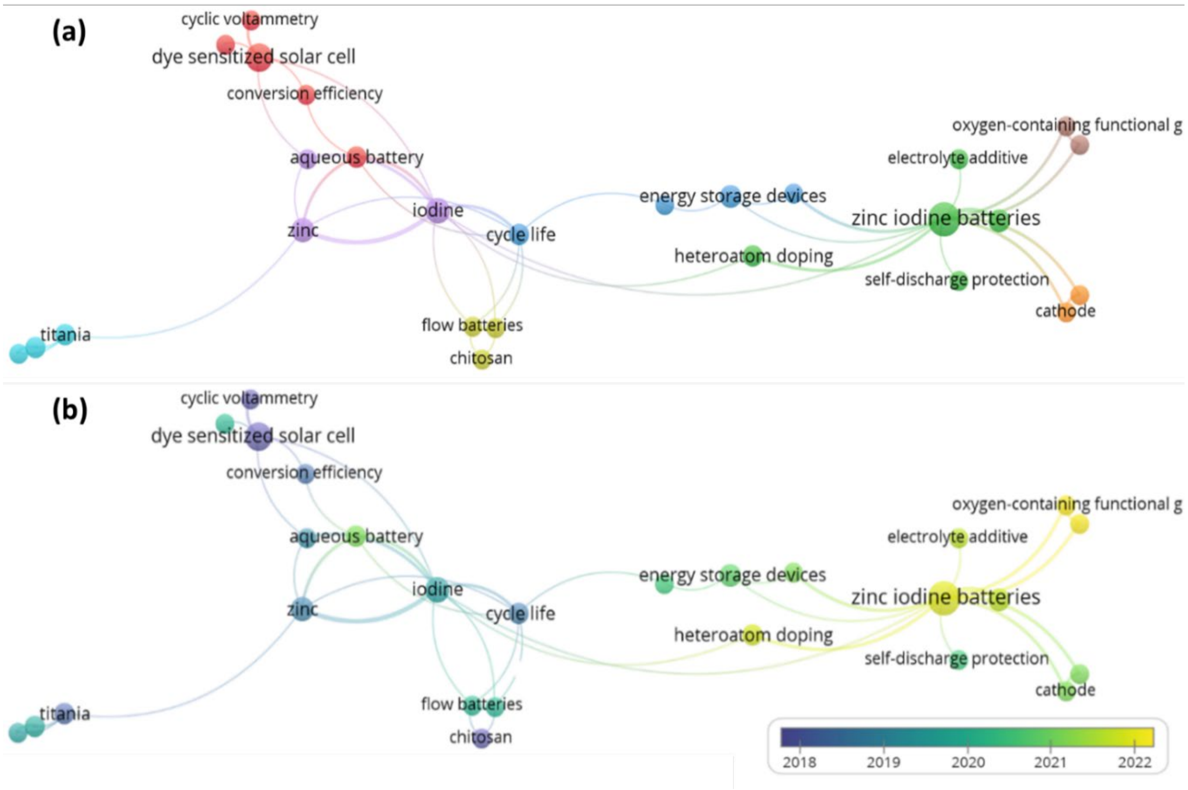

Fig. 4a. Co-occurrence visualization of keyword networks (b) Science overlay visualization and its co-occurrence in years of publications.

Top contributing authors

It is generally accepted that the amount of citations a researcher receives in a field is a measure of their impact [52]. The "kind of analysis" parameter in VOSviewer was set to "co-authorship," and the "unit of analysis" was set to "authors." The minimum number of documents for an author was set at 2, and 124 of the 551 authors met the requirements. Of the 124-author list, 116 items were largely connected. The top 30 authors with the most publications and citations in the field of zinc-iodine ESS are included in Table 2 (the full table is presented in Table S3).

Table 2. First 30 list of the top contributing authors in the relevant field of study

S/N	Author	Documents	Citations	Average citations	Total link strength
1	Zhang Jiatao	11	251	23	40
2	Li Xianfeng	10	512	51	50
3	Chen Song	8	119	15	29
4	Liu Tianxi	8	124	16	51
5	Wang Jingtao	8	58	7	56
6	Wang Zhaohui	8	64	6	26
7	Chen Qianwu	6	61	10	23
8	Yang Yang	6	61	10	30

9	Zhang Leiqian	6	116	19	45
10	Li Yat	5	177	35	12
11	Liu Weiliang	5	41	8	30
12	Liu Yu	5	155	31	14
13	Yao Jinshui	5	29	6	31
14	Zhang Yining	5	20	4	14
15	Chen Nan	4	26	7	18
16	Chen Zhongwei	4	291	73	26
17	Ge Lingfen	4	41	10	34
18	Guo Hele	4	19	5	34
19	He Yulong	4	69	17	13
20	Huang Jiajia	4	19	5	34
21	Huang Zhaodong	4	194	49	31
22	Lai Feili	4	19	5	34
23	Liang Shuquan	4	66	17	11
24	Liu Xiaomin	4	133	33	12
25	Ren Manman	4	26	7	29
26	Sun Jiaqi	4	17	4	12
27	Sun Xueliang	4	35	9	14
28	Tian Zhihong	4	19	5	34
29	Wang Yunxiao	4	37	9	22
30	Zhang Minghao	4	19	5	36

The author with the most publications is Zhang J. (11), while Li X. (10) received the most citations (512) with a total link strength of 50 compared to 40 by Zhang J. as shown in Table 2. This suggests that the works of these two authors have been influential, recognized, and cited by other researchers in subsequent studies, indicating that the author's research has contributed to advancing knowledge and has had a substantial influence on Zinc-iodine ESS. Similarly, in terms of average citation count, which is obtained by dividing the total number of citations by each author's total number of publications, Chen Z. tops with an average citation of 73 compared with 51 by Li X. The high average citation count reflects the influence and importance of the author's publications in influencing the direction of subsequent studies in Zinc-iodine ESS and hence has become a valuable resource for other researchers. Fig. 5 depicts the visualization of authors with at least 2 documents and the linkage of the most significant author. This emphasizes that many writers from different locations are linked in 9 clusters based on citations in the field of zinc-iodine batteries.

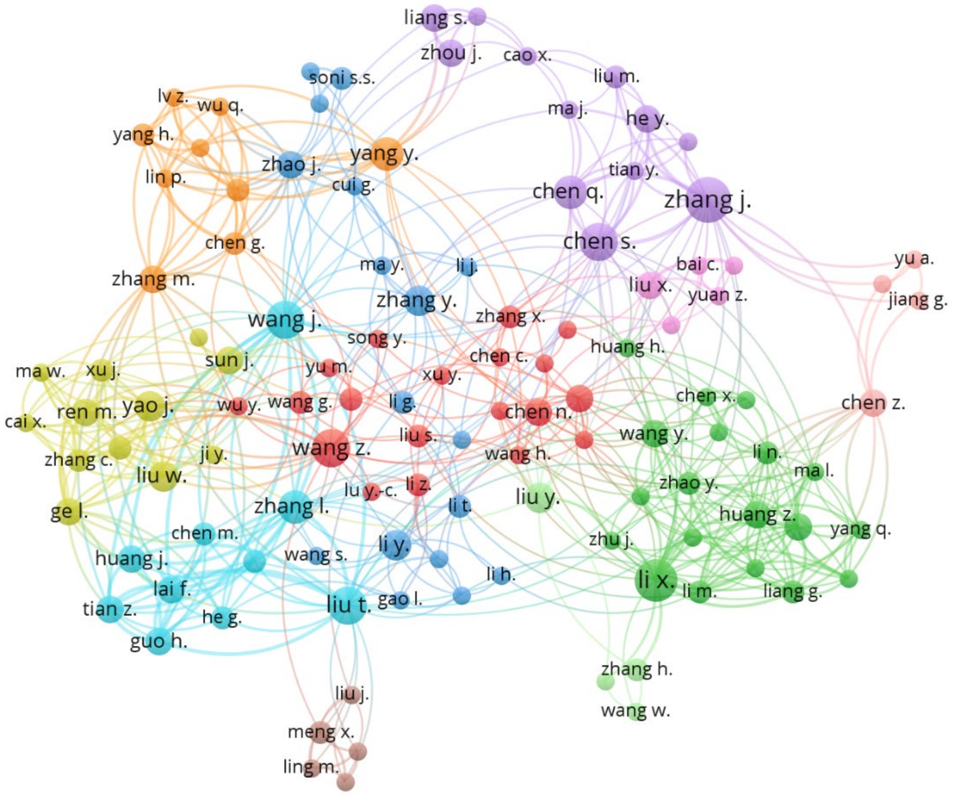

Fig. 5. Visualization of top contributing and connecting authors

Top cited articles

A research publication's influence on a certain field of study may be shown in the number of citations it has earned. In the realm of study, articles having many citations are recognized as milestones. To analyze documents based on citations, the "type of analysis" was set to "bibliographic coupling" and the "unit of analysis" to "document" in VOSviewer. A document’s minimum citation count was set at 2, and 106 of the 131 documents met this requirement (see Fig.6a). Table 3 lists the names of the authors, the year the papers were published, and the total number of citations for the top 23 research publications.

Table 3. List of the top 23 most cited research articles, authors, publication year, and total link strength

S/N	Document	Title	Year	Citations
1	Weng Guo-Ming et al. [53]	Unlocking the capacity of iodide for high-energy-density zinc/polyiodide and lithium/polyiodide redox flow batteries	2017	168
2	Guillén Elena et al. [54]	Electron Transport and Recombination in ZnO-Based Dye-Sensitized Solar Cells	2011	163
3	Yang Huijun et al. [55]	A metal-organic framework as a multifunctional ionic sieve membrane for long-life aqueous zinc-iodide batteries	2020	140

4	Pan Huilin et al. [56]	Controlling solid-liquid conversion reaction for a highly reversible aqueous zinc-iodine battery	2017	124
5	Xie, Congxin et al. [57]	A long cycle life, self-healing zinc-iodine flow battery with high power density	2018	123
6	Zhang Jing, et al. [58]	An all-aqueous redox flow battery with unprecedented energy density	2018	120
7	Xie Congxin et al. [59]	Highly stable zinc-iodine single flow batteries with super high energy density for stationary energy storage	2019	119
8	Yu Donglin et al. [60]	High-voltage and ultrastable aqueous zinc-iodine battery enabled by N-Doped carbon materials: Revealing the contributions of nitrogen configurations	2020	81
9	Dipankar Barpuzary and Qureshi Mohammad [61]	Highly Efficient One-Dimensional ZnO Nanowire-Based Dye-Sensitized Solar Cell Using a Metal-Free, D−π−A-Type, Carbazole Derivative with More than 5% Power Conversion	2014	80
10	Wang Faxing et al. [62]	A Stimulus-Responsive Zinc–Iodine Battery with Smart Overcharge Self-Protection Function	2020	79
11	Li Yixin et al. [63]	Rechargeable aqueous zinc-iodine batteries: pore confining mechanism and flexible device application	2018	78
12	Bai Chong et al. [64]	A sustainable aqueous Zn-I_2 battery	2018	77
16	Li Xinliang et al. [65]	Enhanced Redox Kinetics and Duration of Aqueous I_2/I^- Conversion Chemistry by MXene Confinement	2021b	76
17	Pugliese Diego et al. [66]	A Chemometric Approach for the Sensitization Procedure of ZnO Flowerlike Microstructures for Dye-Sensitized Solar Cells	2013	73
18	Li Xinliang et al. [67]	Activating the I^0/I^+ redox couple in an aqueous I_2–Zn battery to achieve a high voltage plateau	2021a	68
19	Swami Sanjay Kumar et al. [68]	Spray deposited copper zinc tin sulphide (Cu_2ZnSnS_4) film as a counter electrode in dye sensitized solar cells	2014	68
20	Raksa Phathaitep et al. [69]	Copper oxide thin film and nanowire as a barrier in ZnO dye-sensitized solar cells	2009	66
21	Zou Yiping et al. [70]	A four-electron Zn-I_2 aqueous battery enabled by reversible $I^-/I_2/I^+$ conversion	2021	63
22	Dokouzis Alexandros et al. [71]	Photoelectrochromic devices with cobalt redox electrolytes	2020	62
23	Hong Jessica J. et al. [72]	A Dual Plating Battery with the Iodine/$[ZnI_x(OH_2)_{4-x}]^{2-x}$ Cathode	2019	61

From the study, Weng G.-M [53] obtained 168 citations for his publication “Unlocking the capacity of iodide for high-energy-density zinc/polyiodide and lithium/polyiodide redox flow batteries”, which is considered to be one of the trend setters for the rapid growth stage starting 2017. Guillén E. [54] and Yang H. [55] received 163 and 140 citations, respectively, to make up the top three most cited papers in Zinc-iodine ESS. In is quite interesting to note that the papers that made it to the top three in terms of citation were published in 2017, 2011, and 2020 respectively. Additionally, it is intriguing to note that, over the past 20 years, only seven works in this sector have garnered more than 100 citations, demonstrating the fluidity of the concepts and principles for creating sustainable zinc-iodine ESS. In terms of foundation papers, reference can be made to papers written by Wang G-M. (2017), Gullen E. (2011), Pan H. (2017), Xie C. (2019), Zhang J. (2018), and Li Y. (2018) as shown in Fig 6(a). The author's visualization of the most referenced works is shown in Fig. 6(b). VOSviewer analysis revealed that 29 out of 131 documents were linked based on citations and grouped into 4 clusters in colors cluster 1 (red) with 12 items, cluster 2 (green) with 10 items, cluster 3 (blue) with 6 items, and cluster 4 (yellow) with 1 item. The contrast between clusters of the same color and clusters of different colors aids in locating the borders between the various study areas as well as their cohesiveness. It offers insights into the distribution of information, interdisciplinary relationships, and theme organization within Zinc-iodine ESS research. Research trends, distinctive research communities, multidisciplinary partnerships, and links between various research areas can be explored by researchers in this area. For instance, clusters of the same color typically represent closely related research articles or terms that share common characteristics or themes. When clusters have the same color, it suggests that they are more similar to each other in terms of their co-occurrence patterns. These clusters may represent subtopics within a broader research area or closely related research domains.

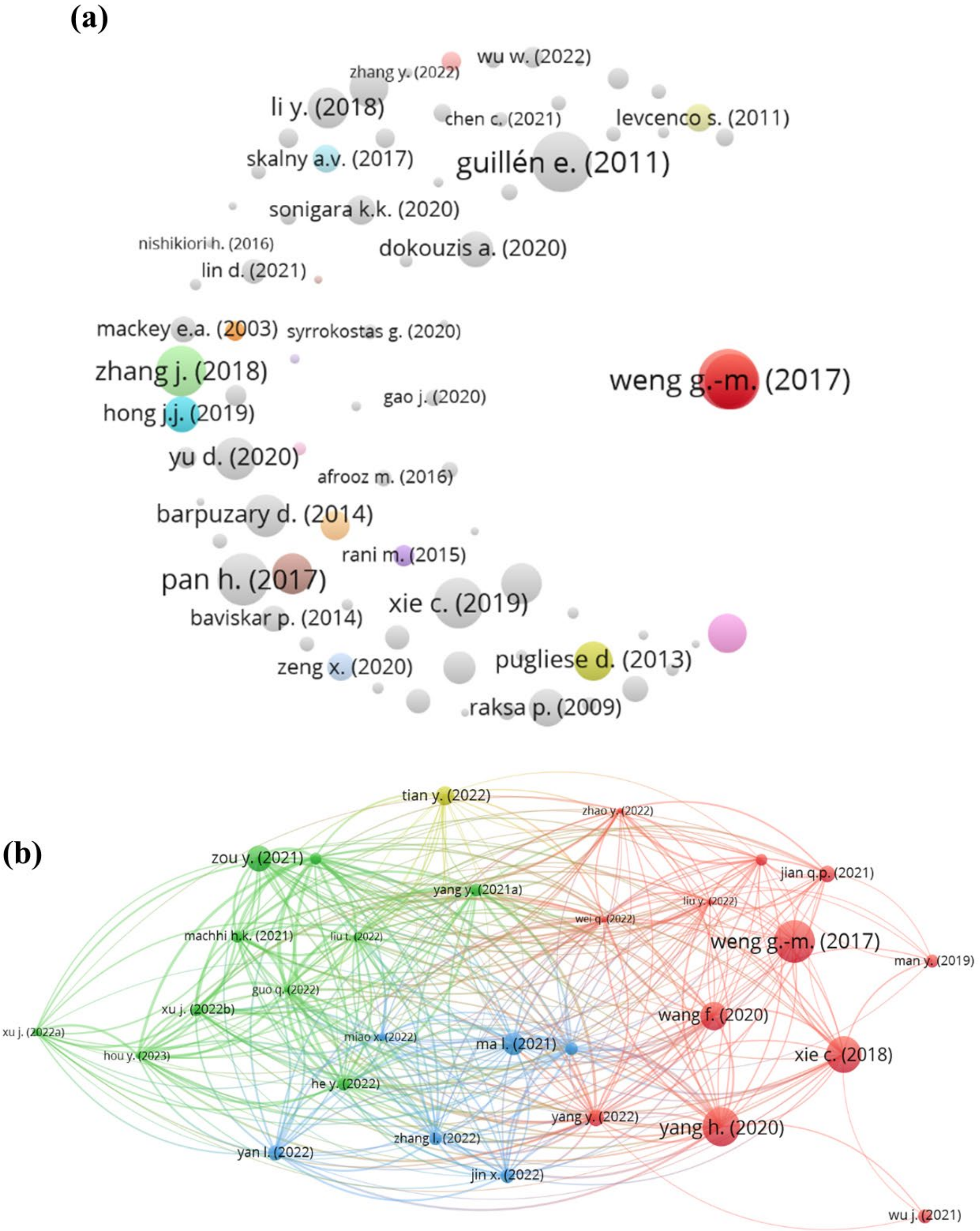

Fig. 6 (a). Visualization of cited documents, (b) Visualization of most cited authors

Top contributing countries

The present scientific field (Zinc-iodine ESS) has seen greater contributions from some nations than others over time. To view countries dedicated to zinc-iodine energy storage devices, the visualization network was created using "bibliographic coupling" as the "kind of analysis", and "countries" as the "unit of analysis". A country's minimum quantity of papers was set at 2, and 18 of 33 countries met this condition. Nine of the 18 countries were largely connected as shown in Fig. 7. Table 4 shows the list of nations that are most active in terms of publications and citations related to the Zinc-iodine ESS. The overall connection strength reveals how much a nation's publications affect those of other nations in this research area. As shown in Table 4, the overall strongest connection strength is held by China (2390), followed by Hong Kong (618), Belgium (552), the United States (498) Canada (450), etc. The VOSviewer included Hong Kong in the display of findings as a country, however, it is important to emphasize that Hong Kong is a Special Administrative Region (SAR) of the People's Republic of China. In terms of citation, China still takes the top spot (1623) followed by India (384) and then the United States (376). Also, China produced the highest number of documents (75) followed by India (9), the United States (9), Germany (9), and then Hong Kong (8). Thus, it can be said that the aforementioned countries have the most influence on research into zinc-iodine ESS at the moment.

Table 4: List of the 18 top contributing countries in the relevant field of study

S/N	Country	Documents	Citations	Total link strength
1	China	75	1623	2390
2	India	12	384	239
3	United States	9	376	498
4	Germany	9	145	448
5	Hong Kong	8	368	618
6	Japan	6	184	256
7	South Korea	6	63	0
8	United Kingdom	6	273	307
9	Canada	5	158	450
10	Belgium	4	19	552
11	Malaysia	4	37	0
12	Australia	3	83	0
13	Greece	3	86	0
14	Russian Federation	3	75	0
15	Thailand	3	68	0
16	Italy	2	135	0
17	Poland	2	55	0
18	Spain	2	203	0

China’s high contribution to zinc-iodine ESS in terms of document, citation, and total links strength could be attributed to the presence of advanced research infrastructure, highly skilled researchers and scientists, and access to government funding, especially due to the national priorities and strategic focus on alternative energy sources. The combination of these and many other factors could be the driving force behind the massive contribution of China to this area of research.

Fig. 7 shows the relationships between nations based on citations. How much a nation has contributed to the field of study is shown by the size of the frame. This graphical depiction of participating nations will aid researchers in creating scientific partnerships, producing joint venture reports, and exchanging fresh techniques and ideas. In general, 5 clusters were found, with cluster 1 (red) having 4 items, cluster 2 (green) having 2, cluster 3 (blue) having 1, cluster 4 (yellow) having 1, and cluster 5 (violet) also having 1. Again, China, Hong Kong, the United States, etc. are the hubs with the strongest connections.

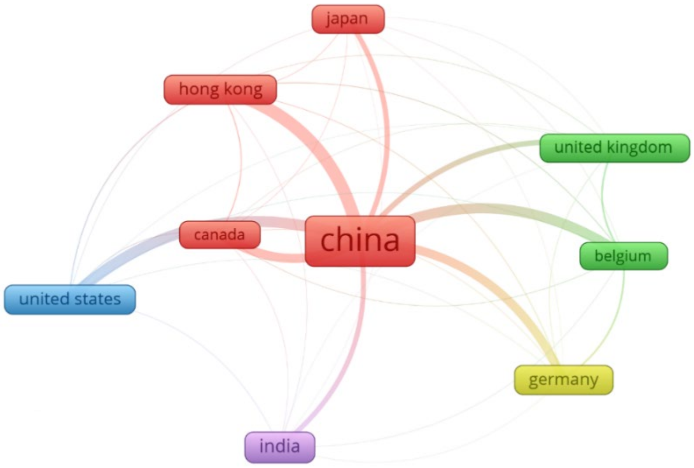

Fig. 7. Countries science mapping: Network visualization

Top contributing organizations

The present research field has greatly benefited from the contributions of several organizations as shown in Table S4. Considering this, "Bibliographic coupling" was selected as the "kind of analysis" and "organizations" as the "unit of analysis." The minimum number of papers required for an organization was 2, and 22 of the 279 groups satisfied this requirement. 19 out of the 22 groups were substantially related as shown in Fig. 8. The connecting linkages to the Key Laboratory for Colloids are also shown in Fig. 8b, while Figure 8c shows the density visualization of the organizations most crucial to the study's topic. The most active organization in terms of publications and citations related to the current study subject is listed in Table S6. An organization's effort may be shown in the overall number of documents, citations, and relationships. On this basis, the Key Laboratory for Colloid and Interface Chemistry, Ministry of Education, School of Chemistry and Chemical Engineering, Shandong University, Jinan, China, Department of Chemistry, Ku Leuven, Belgium, Engineering research Center for Nanomaterials, Henan University, Kaifeng, China, School of Chemical Engineering, Zhengzhou University, Zhengzhou, China and Department of Materials Science and Engineering, City University of Hong Kong made the top five (5) lists with documents of 6, 4,4,4 and 3 respectively. The total link strength indicates how much an organization's papers influence other organizations in this study field. In this regard, the organizations with the strongest link strength are the Research Center for Eco-Environmental Sciences, Chinese Academy of Science, China, School of Chemical Engineering, State Key Lab of Polymer Materials Engineering, Sichuan University, China, Qilu University of Technology (Shandong Academy of Sciences), Jinan, China, all having a total ink strength of 682 apiece followed by the Department of Chemistry, Ku Leuven, Belgium, Engineering Research Center for Nanomaterials, Henan University, China and School of Chemical Engineering, Zhengzhou University, China, also with 587 total link strength apiece. It can be concluded that these organizations are well-established and have a strong research infrastructure, expertise, and funding. This could help researchers identify key institutes in zinc-iodine ESS research to drive collaboration and research efforts within the field.

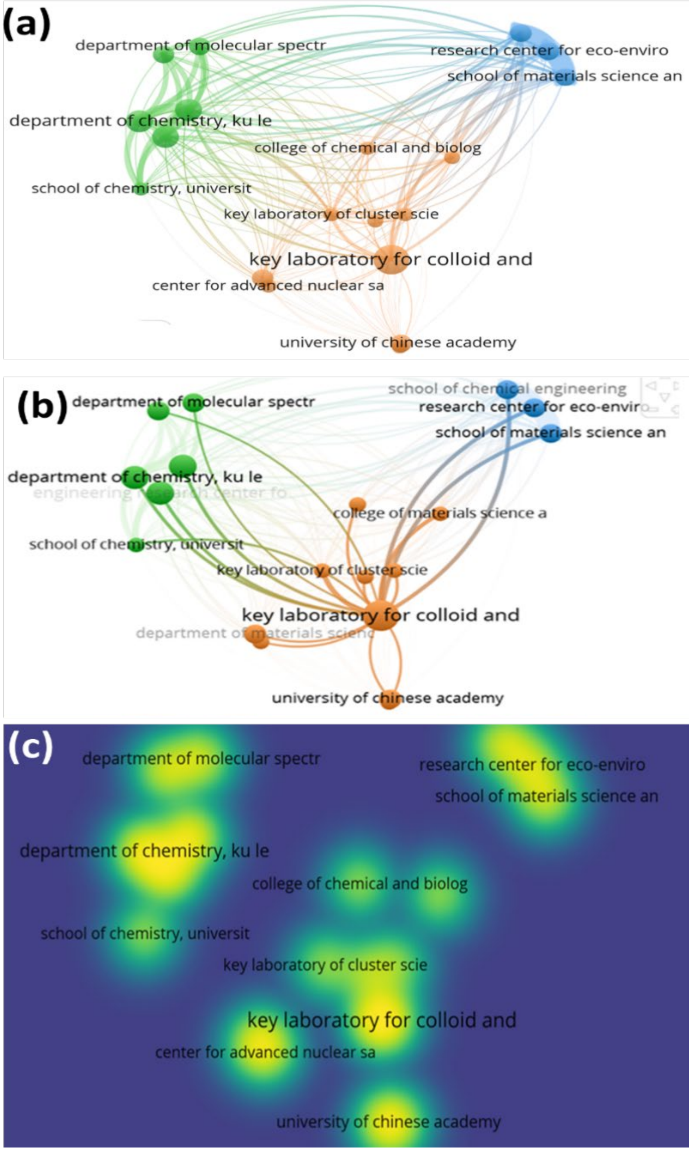

Fig. 8 (a) Network visualization of organization science mapping (b) Connecting links to the key laboratory for colloids (c) Density visualization.

World Collaboration Studies

A world collaboration map analysis (Fig.9, Table 5, the full list can be found in Table S5) indicates significant collaboration between China and other regions/countries such as Hong Kong SAR, Australia, the United States, the United Kingdom, Germany, Belgium and with China serving as the country from which these collaborations emanate. There has been some form of cross-continental collaboration emanating from Australia to Vietnam, Canada to Japan, and India to Australia, however, the frequency is low compared to collaborations emanating from China to other continents.

Table 5: Summarised World Collaboration Map of Country Involvements "From" and "To"

S/N	From	To	Frequency
1	Australia	Vietnam	1
2	Canada	Japan	1
3	China	Australia	5
4	China	Belgium	4
5	China	Canada	3
6	China	Germany	4
7	China	Hong Kong	6
8	China	India	2
9	China	Japan	1
10	China	Moldova	2
11	China	Norway	1
12	China	Singapore	1
13	China	United Kingdom	5
14	China	USA	5
15	China	Vietnam	1
16	Germany	Belgium	3
17	Germany	Israel	1
18	Germany	Thailand	1
19	Germany	United Kingdom	1
20	India	Australia	1

Also, there has been a significant cross-continental collaboration from Germany to Israel and Thailand. key alliances happened between Asian countries, China and Hong Kong (though Hong Kong is considered a Special Administrative Region of China), Europe, specifically, Belgium, and Germany. It is worth noting that India collaborated more fairly with other countries (Australia, Cuba, Germany, United Kingdom, and Vietnam) than its closest neighbors in this field of research. Interestingly, although the United States of America and Canada are neighboring countries, there appears to be no collaboration between authors researching zinc-iodine ESS. Likewise, Russia and China appear to have no collaboration now, specifically on the topic of study. Similarly, in Africa, only Egypt has had a collaboration with researchers from Saudi Arabia on zinc-iodine ESSs as per this finding. This phenomenon calls for enhanced cross-continental collaborations between authors to advance the frontiers in the Zinc-iodine ESSs field.

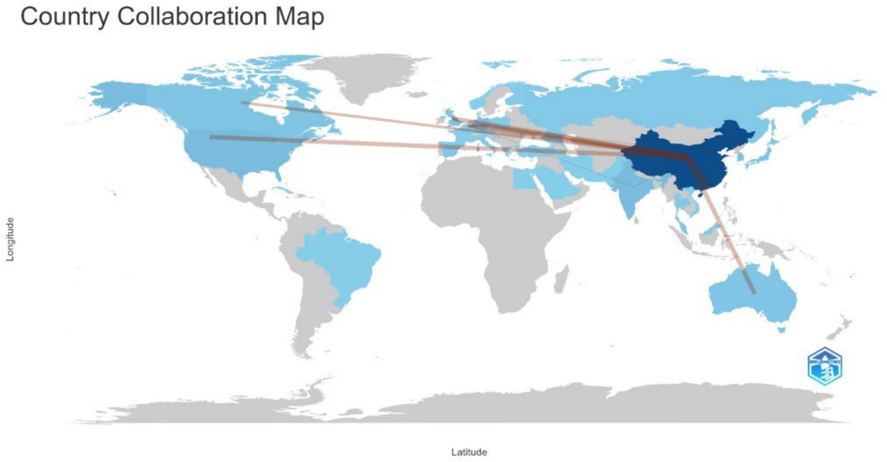

Fig. 9. Biblioshiny country collaboration map

The outcome of the global collaboration map on zinc-iodine ESS have shown the scope of worldwide relationships and identified important collaborating countries/regions. This will allow academics and policymakers to make informed decisions and support effective international collaborations.

Three-fields plot

The three-fields plot sheds light on the interactions and connections among several study areas. Field placement within the plot reveals how close or far apart they are from one another. While disciplines that are farther away reflect greater uniqueness or disciplinary boundaries, those that are closer together imply a higher level of overlap or interdisciplinary links. The three-fields plot analysis was conducted using the biblioshiny of the Biblimetrix software [49] to investigate the interconnections between journals, authors, and keywords. The interaction between top sources (left), authors (middle), and keywords (right) is illustrated in a three-field plot (Fig 10). The visualization demonstrates that most keyword publication sources for zinc-iodine batteries are recorded in Energy Storage Materials with credit to Liu T. Again, the three top keywords in this field zinc-iodine battery, zinc-iodine batteries, and heteroatom doping illustrate the keyword lead in the investigated field. With regards to the leading writers in the field, it is reasonable to assume that China is the driving force behind research in Asia and internationally in the hotspot study area of zinc-iodine batteries observed from the authors' (middle) point of the three fields plot.

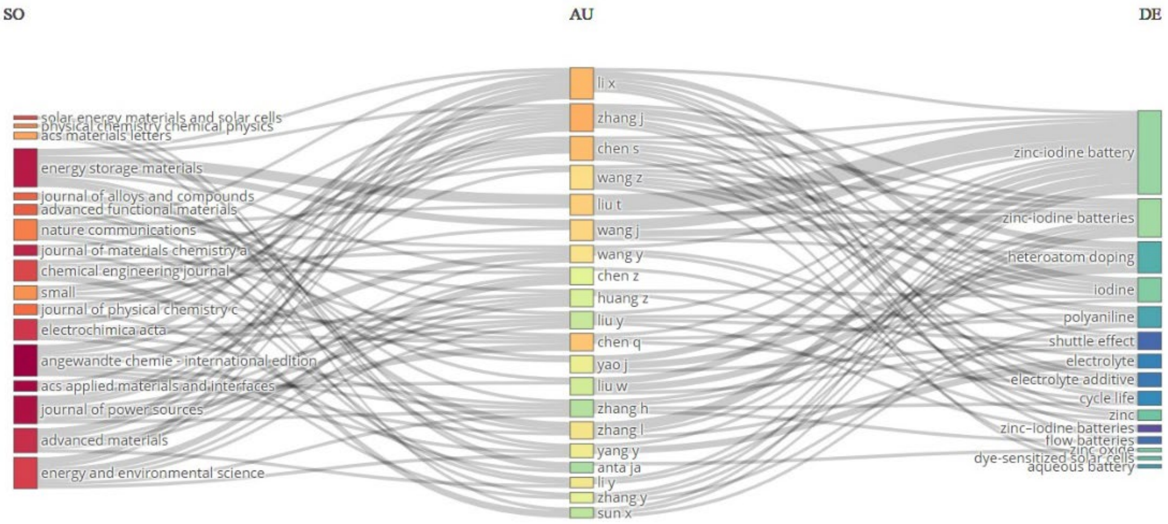

Fig. 10. The three-fields plot for the study based on institutions (left), journals (middle), and country (right).

Keyword Thematic Map

The keyword thematic map helps to identify the main research themes and topics within a specific field. It allows for a quick overview of the dominant areas of research and the distribution of keywords across different thematic clusters. In this map (Fig.11), the keywords “energy storage, cathodes, anodes, electrodes, and redox reactions” are placed in the lower-right quadrant representing a basic theme that has a high centrality and low density and is considered important and not yet developed for the research field. Similarly, “electrolyte, article, and iron” are niche themes with high density and low centrality, considered as well-researched with marginal importance in the study area. In the upper-right quadrant are the motor themes with high centrality and density considered well-researched and important. In this quadrant, are “zinc, iodine, electrolytes, zinc compounds and secondary batteries” illustrating the authors’ attributions and are believed to have evolved from the basic themes. Finally, themes emerging in the lower left quadrant are declining or emerging themes with low centrality and density and are considered not well-developed with marginal importance. In this case, no records of emerging or declining themes appeared.

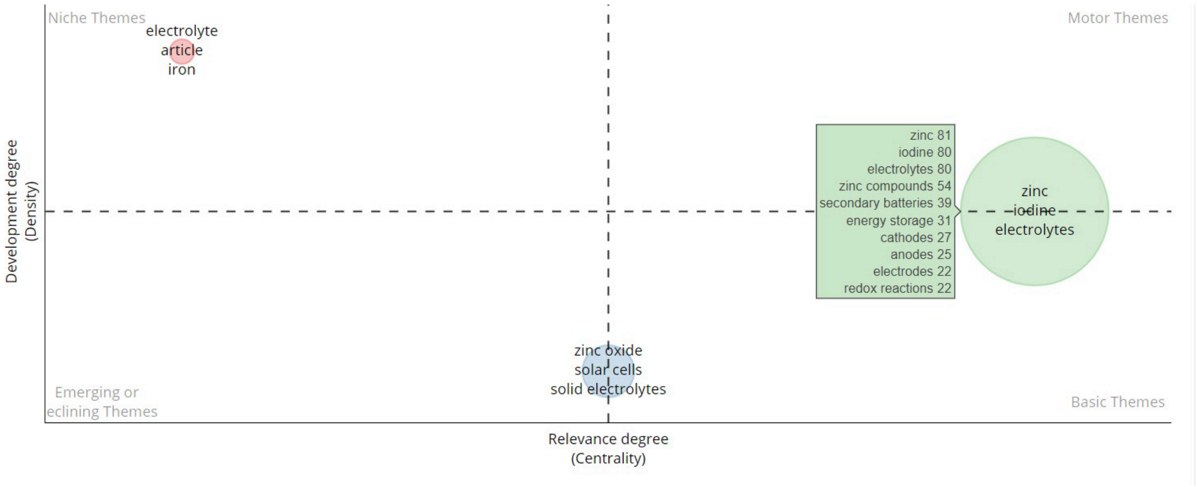

Fig. 11. Biblioshiny visualization of keyword thematic map.

Based on the motor themes (see Fig. 11) with high centrality and low density, a mini qualitative review of advancements in zinc iodine ESS is written. It can be observed that Zinc, iodine, electrolytes, and zinc compounds had the strongest centrality scores of 81, 80, 80, and 54, respectively, hence, the qualitative review is organized around these terms.

3.1 Advancements in zinc-iodine batteries

3.1.1 Electrode Materials

Iodine has drawn more interest as a positive electrode material because of its special qualities, including its enormous supplies in the ocean and its affordable cost, high theoretical specific capacity (211 mA h g^{-1}), and energy density (223 Wh kg^{-1}). According to Kang et al. [73], the top priority of storage batteries is no longer the energy density, but the levelized energy cost (LEC), that is, the cost per kWh delivered as output over the device's entire lifetime. Therefore, important considerations such as the efficacy of the electrode materials, their availability, cycle life, and the specific energy capacity are factored in to achieve LEC. In this regard, porous carbons having large specific surface area, many nanopores, superior chemical stability, and ease of manufacture are used as iodine hosts to create conductive supports to increase the efficiency of electron transmission and boost redox conversion [74] [75]. Following these precepts, Xu et al. [76] investigated pyrolyzed biomass porous carbon for $Zn\text{-}I_2$ battery and obtained a discharge capacity of 100 mA g^{-1} over 150 cycles at a density of 1000 mA g^{-1}. The performance of the electrochemical catalysts as anode and cathode materials in the creation of zinc-iodine batteries has also been shown to be effectively enhanced by heteroatom doping [73], however, the issue of polyiodide ions shuttling remained a challenge. Therefore, to suppress the shuttling of polyiodide ions, researchers have studied and developed a variety of effective methods involving ionic sieve membrane separators, novel hydrogel electrolytes, and modified electrodes [77]. On the premise of this, Liu et al. [78] evaluated the fundamental catalytic activities of various N heteroatom-doped porous carbon for iodine redox reaction /shuttling and its correlation to the performance of the aqueous zinc iodine batteries. They claimed that by using nitrogen-doped porous carbons to catalyze the iodine conversion, it was possible to prevent the creation and crossing of the triiodides, which results in a durable zinc iodine battery with a high energy density (320 Wh kg^{-1}) and very long cycle life of 10,000 cycles. Additionally, newly developed separator mediums modified by carbon composites are increasingly on the rise with common ones being graphene@carbon nanotube, graphitic carbon nitride/carbon nanotube, porous carbon/multi-walled carbon nanotube, and N, O co-doped chlorella-based biomass carbon as cathodes and anodes [79]. The recent developments in this modified separator mediums are presented under the subsection cathodes, anodes, and their method of fabrication.

3.1.2 Cathodes

The greatest performance of a cathode may be reached by understanding material properties such as bulk and surface crystallinity, band structure, and surface morphology [80]. Understanding these properties offers suggestions for improving cathode processing and growth by supplying performance data to verify cathode behavior. A cathode is one of the two electrodes involved in an electrochemical cell or system. It is the electrode where reduction reactions occur during the electrochemical process. Reduction is the increase of electrons or the lowering of a species' oxidation state. In zinc-iodine batteries, cathode reduction is a complicated process that involves the conversion of iodine (I_2) to iodide ions (I^-) during discharge and the reverse reaction during charge. The cathode's reduction of iodine to iodide is a multistep process, and the pace of this reduction reaction impacts the battery's overall performance [80, 81]. The most reliable strategy to avoid cathode reduction in Zn-I_2 batteries is to use a cathode material that is stable in the presence of iodine and zinc ions. Carbon, graphite, and titanium have all been discovered to be effective [82]. To prevent iodide ions in electrolytes from forming soluble I_3^-, an iodine cathode coated over a carbon fiber paper (CFP) current collector having a high areal capacity of 4 mAh cm^{-2} was coupled with a zinc metal anode (ZMA) [72]. The fabricated Zn-I_2 cathodes had a high capacity of 174.4 mAh g^{-1} at 1C and a cycle life exceeding 3000 with 90% capacitance retentions as shown in Fig 12a [56]. The remarkable cyclic stability and reversibility were attributed to the rational regulation of the competitiveness between iodine species' solvation in the electrolytes and the adsorption rate in carbon as illustrated in Fig 12 b. A study by Miao et al. [77] investigated the electrodeposition of PPy and PANI onto pristine carbon felt (p-CF) as electrodes for the fabrication zinc-iodine battery. The highly conductive electrode achieved a specific capacity of 235.9 mAh g^{-1} at 0.35 A g^{-1} (~1.7 C), and 10,000 cycles with 75.7% capacity retention (Fig 12 d). In furtherance to this work, Zeng and co-workers prepared a high-performance polyiodide doped PANI cathode charge storage by iodine redox couple. Iodine can be strongly confined in the PANI backbone during the redox process in the form of I_5^-, I_3^-, or I^- by coulomb force to suppress iodine sublimation and polyiodide dissolution during cycling. This resulted in a high reversible capacity of 160 mAh/g and remarkable cycling stability with capacity retention of 79% after 700 cycles at a current density of 1.5 A/g [83]. Similarly, the application of metal-organic framework-5 (MOF-5) and cotton fiber modified with ketjen black (KB@CF) as a separator has been reported to have high capacity as high as137 mAh g^{-1} after 300 cycles at 0.1 A g^{-1} (see Fig. c) [79]. In another study, heteroatomic functional porous carbon-based cathodes derived from zeolitic imidazolate framework precursors and glass fiber membrane as a separator were investigated [84]. The high specific surface area and pore size distributions of the heteroatomic functional ZIF8 carbons served as iodine hosts for zinc-iodine aqueous batteries, which transformed the weak van der Waals force between the hosts and the iodine species into a strong chemical interaction ensuring maximum specific capacity of 313.6 mAh g1 (at 0.5 C) at 100 C capacity retention. Also, I_2 cathode halogen bonds of 285 mAh g^{-1} output capacity and salient rate capabilities (101 mAh g^{-1} at 12 A g^{-1}) with good cyclic durability (86.8% capacity retained after 103 cycles) have been reported [85] to eliminate the problem of I_2 sublimation and dissolution into aqueous electrolyte media.

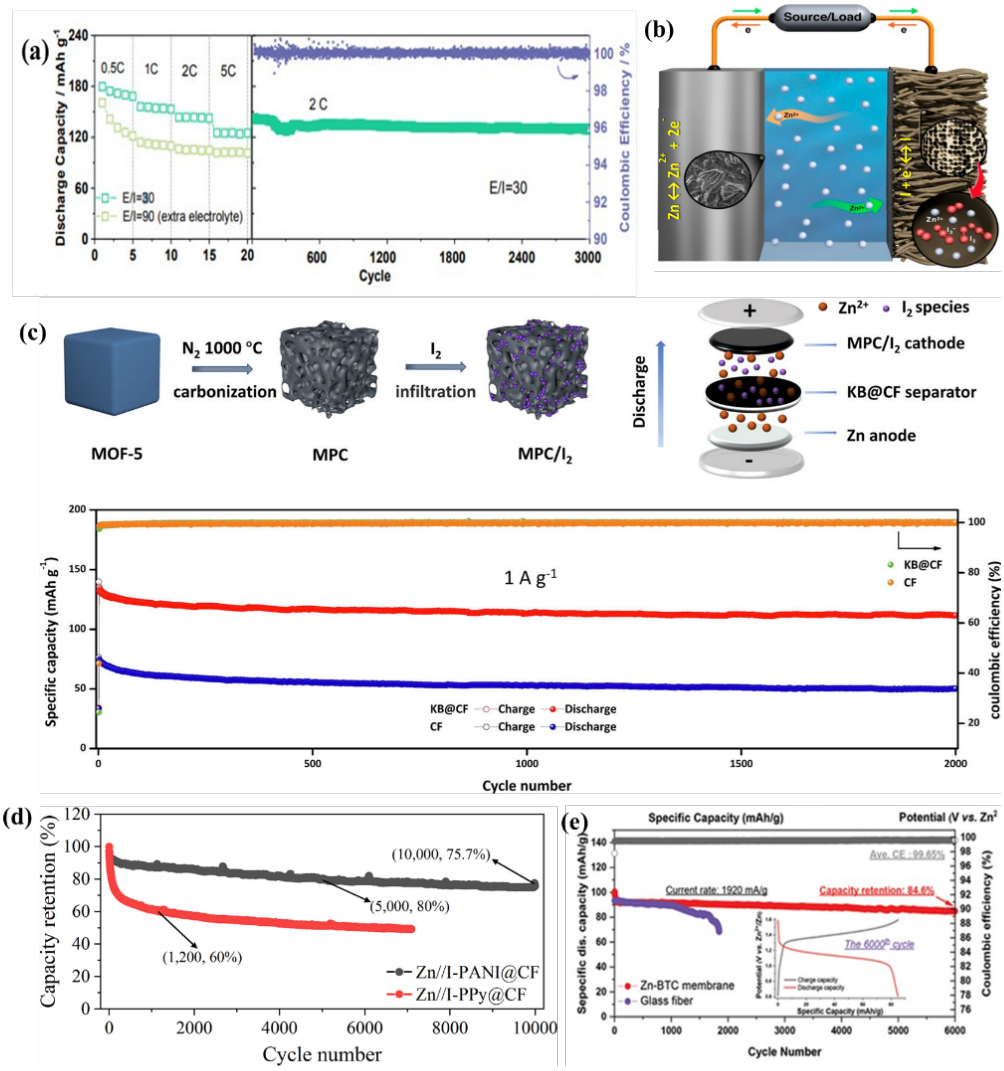

Fig. 12. (a) Rate and long-term cycling of 40I$_2$/ACF electrode with a controlled electrolyte to iodine (E/I) ratio of 30 mlE gI^{-1} at 2C, (b) An illustration of the rational regulation of competitiveness between iodine species' solvation in the electrolytes and the adsorption rate. Reprinted with permission from ACS Publications [56], (c) ZIBs constructed with the MPC/I$_2$ cathode and the KB@CF separator with excellent electrochemical performance. Reprinted with permission from Elsevier [79], (d) Capacity retention of Zn//I-PANI@CF and Zn//I-Ppy@CF batteries during GCD cycle process at 20 Ma. Reprinted with permission from Elsevier [77], (e) Long-life Zn–I$_2$ cell at current density of 1920 mA g^{-1} with GF and Zn-BTC membrane. The inset curve is charge/discharge profile of the 6000th cycle with Zn-BTC membrane. Reprinted from Wiley and Sons [55].

For further improvement, Ji et al. [86] proposed a nitrogen-doped litchi shell porous carbon cathode host material that performed well at a capacity of 127 mAh/g at 100 mA g^{-1} as a matrix for zinc-iodine batteries, whereas Chen et al. [87] developed a pouch cell with W$_2$N/N-C cathode of 137 mAh g^{-1} specific capacity and retention of 94.8% over 300 cycles. It has been reported that graphene cathodes promote the kinetics of I_3^-/I^- redox reaction to achieve high coulombic efficiencies of nearly 100% with stable capacities [88]. In this case, iodine species were hindered from dissolving in an electrolyte using an I_2-loaded oxidized salt-templated carbon (I_2/OSTC) cathode and glass microfiber filter membrane separator. After 10,000 cycles, there was an 85.04% capacity retention for the battery [89]. I_2 cathode served as a sieve membrane for long-life aqueous zinc–iodide batteries, and metal-organic framework (MOF) membrane as a separator, achieving >6000 cycles, high capacity retention (84.6%), and high reversibility (Coulombic efficiency: 99.65%) as shown in Fig 12 e [55]. The successful use of tungsten nitride-decorated porous carbon composite as a cathode in a Zn-I_2 battery produced 186.5 mAh g^{-1} of discharge capacity and a sustained cycle performance over 100 cycles [75]. Based on this improvement, Tian et al. [90] described the use of an iodine cathode in the fabrication of a smart self-protection ability in batteries with excellent performance. To further enhance the electrochemical performance, a cation exchange membrane that serves as a separator and a hierarchically hollow carbon nanostraw (HCNS) developed by Chai et al. [91], to serve as the cathode to prevent a reduction in battery capacity while enhancing cycle stability. The zinc-iodine batteries had a high capacity of 234.1 mAh g^{-1} at 1 A g^{-1} and ensured both iodine species adsorption and dissolution in the electrolyte reached quick equilibrium. The zinc-iodine batteries had higher capacity retention rate and electrochemical exchange capacity than the standard unidirectional carbon nanotubes.

It has been discovered that a composite material with iodine and microporous carbon can function as a cathode for zinc-iodine batteries with a CE of 97%, and EE of 81%, and can run continuously for more than 500 cycles [59]. Similarly, Machhi et al. [12] disclosed a multipurpose porous metal-organic gel (MOG) cathode and cellulose separator with a capacity of 184.9 mA h g^{-1} and a superior retention of 95.8% after 1500 cycles at 1 C rate. Furthermore, the creation of a four-electron transfer with quick kinetics in a zinc-iodine battery cathode to achieve 6000 cycles and 750 Wh kg^{-1} energy density has been reported [70]. Also, an experimental analysis of quasi-solid Zn/I_2 batteries using alginate-based hydrogel electrolytes is reported to have a high-capacity retention of 83.1 mAh g^{-1} (72% remained) after 100 h of rest [30]. This study exemplifies the importance of porous carbon-based materials and polysaccharides in Zinc-iodine ESSs because these materials provide a large surface area for electrode-electrolyte interaction, facilitating ion transport, and improving the stability of the electrode material. To understand the effect of cathode doping on Zinc-iodine ESSs, Yang et al. [5], used active carbon fiber cloth N-doped porous carbon to enhance the cathode performance to attain impressive electrochemical performance. Cathode modifications have recently received attention as a more effective technique to improve Zinc-iodine battery performance over time.

4.1.3 Anodes

A key step in developing new and better materials for energy trappings is the connection of cathodes and anodes. Anodes are best known for their improved efficiency and capacity for energy collecting, as well as being reasonably priced [92]. Interest in potential active mediated anode materials with low toxicity, high safety, and abundant supply for zinc-iodine batteries has peaked [77]. As a result, various zinc anode materials have been investigated to increase internal resistance and restrict the reversibility of zinc-iodine batteries. [93]. For instance, Wei et al.,[94] reported a zinc-iodine battery with a cycling capacity of 400 h at 5 $mAcm^{-2}$ after 1000 cycles in a 1M $ZnSO_4$ aqueous electrolyte. To address the problem of self-discharge, sluggish kinetics, low practical energy density, and dendrite formation associated with zinc-iodine batteries, a novel three-dimensional functionalized graphene anode that facilitates iodide redox reactions and immobilizes dissolved polyiodides was designed [88]. The composite anode (zinc coated with reduced graphene oxide) suppressed the detrimental shuttling effect, enhanced the performance, stabilized the anode during the repeated zinc stripping/plating, and thus prevented dendrite formation. The rGO-Zn electrode exhibited a dendrite-free state after 100 discharge/charge cycles at a high current density of 10 A g^{-1}, (see SEM image in Fig. 13 a). Based on this, Chen et al. [75] studied the effect of Zn anode coupled with W_2N/N-C on iodine batteries and reported minimal polarization throughout the charge/discharge process, as well as increased reversibility and cycling stability for 2000 cycles. The increased electrochemical performance was attributed to the confinement effect of W_2N/N-C for the reversible conversion of iodine and polyiodide, hence improving redox reversibility and active material usage. Similarly, the effect of vermiculite nanosheets (VS) suspension electrolyte on the cycle stability and self-discharge behavior using Zn anode to provide a protective layer for Zn-I_2 coin cells were studied [95] and an improved battery performance was achieved with the Zn-anode material by realizing an ultralong lifespan (>6000 cycles), high-capacity retention (84.6%), and high reversibility (Coulombic efficiency: 99.65%), as well as significant reduction in corrosion. Also, a multifunctional MOF (Zn-BTC) was used as Zn-anode material, and the MOF membrane suppressed the shuttling of I_3^- and restrained related parasitic side-reactions on the Zn anode as illustrated in Fig.13b [55].

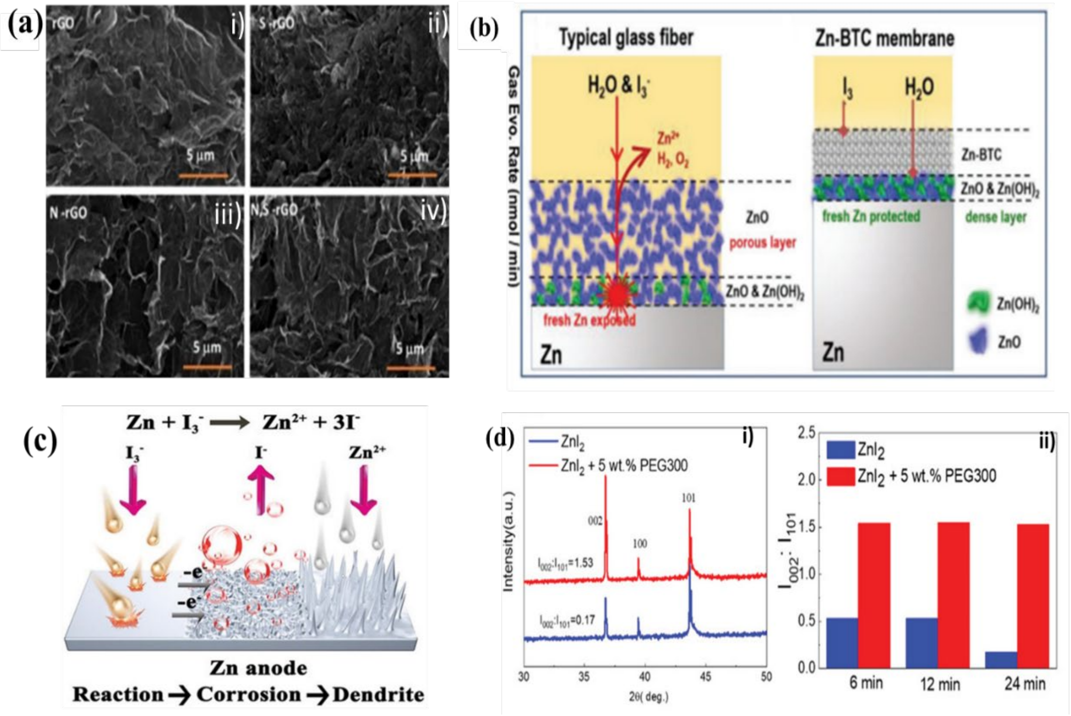

Fig.13. (a) Dendrite free SEM images of rGO electrode materials. Reprinted with permission from Elsevier [88], (b) Schematic illustration of component evolution of Zn anode after the corrosion of iodine species and H_2O with GF separator. Reprinted with permission from Wiley and Sons [55], (c) Schematic diagram for the mechanism of polyiodide corroding Zn anode [96], (d) Electrochemical crystal growth structure and orientation of electro-reduced Zn in 2 M ZnI2 and ZnI_2 + 5 wt% PEG300 aqueous electrolytes. (i) XRD analysis of Zn deposits after 24 min growth under galvanostatic conditions at a rate of 4 mA cm^{-2}. ii) The I_{002}:I_{101} peak ratio deduced from XRD measurements for Zn deposit after 6, 12, and 24 min growth, again at a fixed rate of 4 mA cm^{-2}. Reprinted with permission from Wileyand Sons [97].

To address the problem of corrosion and dendritic development on the anode of zinc-iodine batteries, Zhang et al., [96] developed a sulfonate-rich ion-exchange layer (SC-PSS) to control the transport and reaction chemistry of polyiodide and Zn_2^+ at the zinc/electrolyte interface. The produced ZIBs lasted over 6000 cycles with exceptional capacity retention (90.2%) and reversibility (99.89%), and the inhibition mechanism is illustrated in Fig. 13c [96]. In a similar study, the zinc anode reversibility increased by 3000 cycling stability with a low reversible deposition between 30 and 40 mV of rate capabilities [98]. Equally, experiments on anode materials made of zinc flakes for cutting-edge zinc-iodine batteries produced a capacity of 234.1 mAh g^{-1} at 1 A g^{-1} [26]. To control the electro-crystallization of zinc in creating a smooth and compact zinc electrodeposition, a zinc anode with a high areal capacity of 10 mAh cm^{-2}, 120 stable cycles, and a high CE of 99.5 has been reported [99] (see Fig. 13d). The results of the analyzed studies unmistakably demonstrate that the basic material and other derivatives of zinc materials are promising anodes for zinc-iodine batteries. However, due to iodine's insulating properties and the shuttle effect of polyiodide, aqueous zinc-iodine batteries suffer from limited capacity and poor stability. To address this issue, Yang et al. [55] used a metal-organic framework (MOF) as an ionic sieve membrane to suppress I_3^- shuttling and restrain parasitic side-reactions on the Zn anode, thereby regulating the electrolyte solvation structure and improving both the iodine cathode and the Zn anode simultaneously. The $Zn-I_2$ batteries have a very long lifetime (>6000 cycles), good capacity retention (84.6%), and strong reversibility (Coulombic efficiency: 99.65%). Also, a nitrogen-doped porous carbon material is synthesized by a simple two-step pyrolysis process by mixing urea and glucose to form iodine nitrogen-doped porous carbon composites (I_2-NPC) by thermal diffusion [100]. Similar to the previous findings, a high specific capacity of 345.3 mAh g^{-1} at 0.2 C was achieved with outstanding cycling stability (capacity retention of 80.9% after 10, 000 cycles at 10 C), high operating potential of 1.37 V, and an extremely low voltage gap of 52 mV of $Zn-I_2$ due to the high specific surface area (526.6 m2 g^{-1}) of I_2-NPC, indicating a significant number of active sites.

3.1.4 Electrolyte system for zinc-iodine batteries

Rechargeable iodine batteries have emerged as promising alternatives due to their positives such as low cost, large abundance, good electrochemical reversibility, and high theoretical capacity. However, these systems have encountered various problems, including the shuttle effect, poor kinetics, low thermal stability, and high iodine species solubility. Numerous approaches have been suggested to deal with these problems, including the use of electrolyte additives, carbon fiber, carbon microtubes, and graphene, all of which have safe designs and increased performance [60]. Since the first $Zn-I_2$ prototype was discovered in the 1980s, zinc iodine ($Zn-I_2$) batteries have received a lot of attention among aqueous zinc-based EES due to their relatively high redox potential (I^-/I_3^-, 0.53 V vs. SHE) and significant theoretical capacity (211 mAh g^{-1}) of iodine with reversible reaction kinetics [74]. Various electrolyte solutions for zinc-iodine batteries have been the subject of numerous studies. For instance, Lee et al.[101] investigated the concentration effect of I_2 in the electrolyte as a crucial factor in determining performance regarding dye-sensitized solar cells (DSSC). Using 1M $ZnSO_4$ as the electrolyte, Yu et al. [60] evaluated the electrochemical performance of an I_2-NPC chemically built electrode. The results of the analysis show high cathode and anode peaks with enhanced performance. It is noteworthy to note that, in addition to activating reagents, the use of inert additives during the chemical activation process possesses the potential to change the microstructures and enhance the electrochemical performance of $Zn-I_2$ ESS [74]. Based on this knowledge, nitrogen-doped porous MOFs are produced with high carbon and nitrogen contents as precursors. The metal-nitrogen linkage caused the parent metal's lattice to expand and the metal d-band to contract, which improves the surface chemical characteristics of MOFs [75]. To enhance battery performance using additives, such as organic solvent, electrode-electrolyte interface impedance, and the redesign of electrolyte structure, additions tend to increase the solubility and stability of electrolytes while reducing side reactions. The metal-organic framework zinc hex-acyanoferrate(III) (ZnHCF) was employed as the electrode material to create a cation reservoir for a Zn-I_2 battery [93]. The battery's electrochemical performance was good, exhibiting dendrite-free behavior during cycling and a 400-hour average Coulombic efficiency of 99.6%. By deep cycling 5 mA cm^{-2}, the Zn@ZnHCF electrode's cycle capacity was revealed and shows that the Zn-I_2 complete cells' high-rate cycle capacity remained stable for 1000 cycles. The microstructure of the resultant carbon was significantly impacted by the addition of additives like $CaCl_2$ and CsCl-NaCl eutectic salt during the synthesis procedure with $ZnSO_4$ as an electrolyte. The electrodes are made by combining super P, polytetrafluoroethylene (PTFE) binder, and as-obtained porous carbon/I_2 composites (CDs/I_2, CSs/I_2, CNSSs/I_2, and CNSs/I_2). According to the study, the interaction properties including heteroatomic content, surface area, and microstructures affect how well iodine cathodes function electrochemically [84].

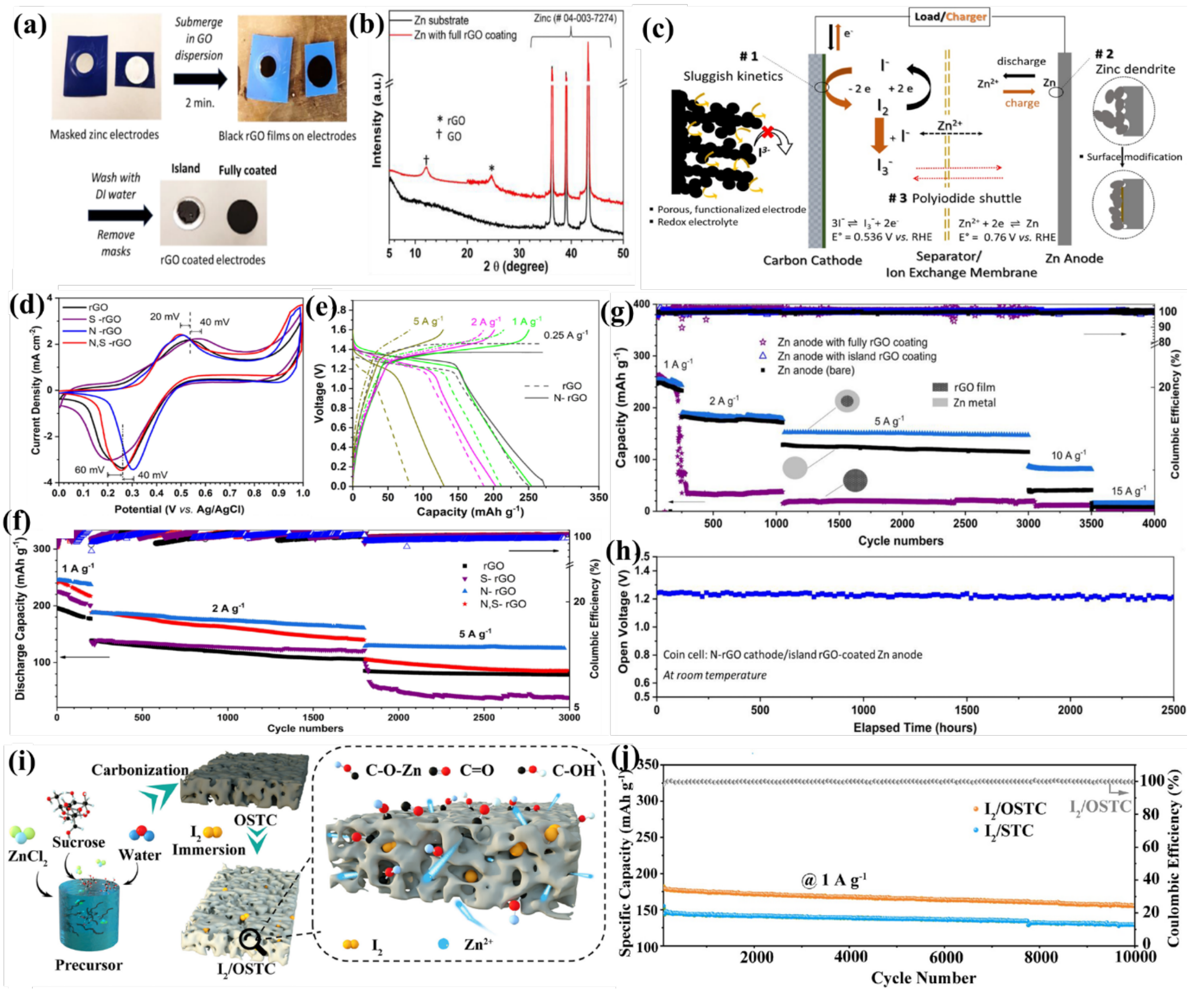

Fig 14. (a) Fabrication of rGO-coated Zn compound anodes, (b) XRD of a fully rGO-coated and a bare Zn anode, (c) Schematic illustration of problems of conventional Zn-I_2 battery and proposed solutions, (d) CVs of various (functionalized) graphene electrodes at 5 mV/s in a voltage window of 0–1 V vs. Ag/AgCl, (e) Voltage profiles of rGO and N-rGO cells at various current densities, (f) Discharge capacity at various current densities over 3000 cycles, (g) Performance of devices with various Zn anodes, (h) Open circuit voltage of a heavily-used coin cell over time after 2000 galvanostatic dis-/charge cycles at 5 A g^{-1}. Reprinted with permission from Elsevier [88]. (i) Preparation of I_2/OSTC and working mechanism as a cathode in zinc-iodine battery, (j) Cycling performance of the I_2/OSTC and I_2/STC cathodes over 10,000 cycles. Reprinted with permission from Elsevier [23].

Furthermore, the issue of iodine species dissolution into the aqueous electrolyte was addressed by creating a porous oxidized salt-templated carbon (OSTC) with abundant carbonyl groups [23]. This was achieved using $ZnCl_2$ and H_2O as a gasification agent/oxidant and a salt-template host to tightly confine iodine. The result was an exceptional rate performance for the as-assembled Zn-I_2 battery, as well as remarkable cycling stability (high capacitance retention of 85.04% after 10,000 cycles). To reduce water-induced parasitic reactions and improve capacity retention and cycle stability of Zn plating/stripping, an artificial solid electrolyte interphase (ASEI) consisting of carboxymethyl cellulose (CMC) was applied to a zinc sheet and a zinc-graphite composite anode. CMC suppresses corrosion by lowering water activity on the anode's surface and controls the flux and local concentration of Zn ions at the surface, allowing for uniform Zn dissolution/deposition. A CMC-ASEI fabricated Zn-iodine battery exhibited good cycling stability (2,000) at 5 mA cm^{-2} [102]. The importance of polymer additives like PDMA, PDPA, PVDM, P4VP, and P2VP for boosting the electrolyte performance of zinc-iodine batteries has been discussed extensively by Zeng et al. [62]. For instance, P2VP blocks the Zn^{2+} transport while making the electrolyte sensitive and preventing deterioration. In a study by Chai et al. [91], a Nafion cation exchange membrane was used as a key additive in effectively blocking I^- and I_3^- anions while also maintaining high levels of the two anions in the cathode area. This prevents a decline in specific capacity and improves the cycle stability of electrode materials. Zou et al. [70] report the addition of acetonitrile (ACN) to control Zn^{2+}, H_2O, and Cl^- coordination in the electrolyte to prevent the harmful hydrolysis of the battery species. Table 6 provides lists of recent works in electrode materials, electrolyte composition for enhanced electrochemical performance.

Table 6. Electrodes, electrolytes, and electrochemical performance of zinc-iodine batteries

Type of Electrode material	Type and composition of electrolytes	Electrochemical performance (capacity retention and cycles)	Stability/Solubility	Application	Ref.
I_2@W_2N/N-C	M $ZnSO_4$	Specific capacity of 200.9 mA h g^{-1} after 2000 cycles, capacity retention of 85%	170.3 mA h g^{-1} at 5C	Zinc-iodine batteries	[87]
CF-NSs	C/I_2 materials, Acetylene black, and polyvinylidene fluoride in *N*-methyl-2-pyrrolidone (NMP)	Specific capacity of 132.6 mAh g^{-1} and capacity retention of 97.2 % after 20,000 cycles	5 A g^{-1} cycle stability with excellent reversibility	Zinc-iodine based SCHD	[74]
PANI@CF	1 M H_2SO_4	Specific capacity of 235.9 mAh g^{-1} at 0.35 A g^{-1} (~1.7 C), long cycle life (75.7% capacity retention after 10,000 cycles) at 3.5 A g^{-1} (~17 C)	High stability	Zn//I battery	[77]
Zn@ZnHCF	1 M $ZnSO_4$	Favorable with dendrite-free behavior during cycling and an	Improved with the introduction	Zn-I_2 battery	[93]

		average coulombic efficiency of ~99.6% for 400 h	of a cation reservoir		
PANI	Zinc halide solution	Capacity of 2 mAh cm^{-2} at 6 mA cm^{-2}, and an excellent capacity retention of 99.9% after 1000 cycles	Excellent	Zinc-iodine batteries	[15]
PNC-1000-I_2	1 M $ZnSO_4$	4 A g^{-1} and prolonged lifespan after 10,000 cycles	Excellent rate performance	Zn/PNC- I_2 Battery	[78]
Zn/ZnGn	CMC	Capacity density of 5 mAh cm^{-2} after 2000 cycles	Improved stability resulting in enhanced capacity retention and cycle stability	Zinc-based batteries	[102]
CNFs	1M Zn $(CF_3 SO_3)_2$ and M $LiCF_3SO_3$	Specific capacity of 174.4 mAh g^{-1} at 1 C and good cycling stability over 3000 cycles	Improved thermal and electrochemical stability	Zinc-iodine batteries	[9]
Zn metal anode	1M ZnSO4 in 35% w/w block copolymer	High capacity retention (>90%) after 100 cycles at different current densities (10-200 mA g^{-1})	High stability	Zn–I_2 Battery	[103]
Polyiodide doped conductive polymer (PANI) cathode	Aqueous electrolyte (2 M ZnSO4)	Capacity of 160 mAh/g and remarkable cycling stability with capacity retention of 79% after 700 cycles at 1.5 A/g	Excellent	Zinc-iodine batteries	[83]
Zn metal anode	Vanadium sulfide (VS) in a 1 M $ZnSO_4$ aqueous solution	230.6 mA g^{-1} after 13000 cycles and a capacity retention of 91.0%, CE of 100.8% 40000 cycles with a negligible capacity decay at 20 C	Ultra-long lifespan	Zinc-iodine battery	[95]
PB-based materials	1 M ZSPI	Capacity retention of 94%, power density of 5538 W kg^{-1} 1500 charge–discharge cycles	Excellent electrochemical stability	Zn-I_2 batteries	[104]
Reduced Graphene oxide	Water-In-Salt (WIS) in concentration	100% capacity retention after 2000 cycles at 50 mA cm^{-2}	High performance	Zn/rGO I_2	[105]

	of $ZnCl_2$ and KI				
SC-PPS@Zn	M $ZnSO_4$ and 1M KI	Cycling capacity of 6000 cycles, capacity retention of 90.2% and columbic efficiency of 99.89%	Extension of battery life	Zinc-iodine batteries	[96]
ZnI_2/N-C	1 M $ZnSO_4$	Energy density of 220 Wh kg^{-1} after 250 cycles	Stable	Stimulus Responsive Zinc-Iodine Aqueous Batteries	[62]
3DC@I_2	$ZnSO_4$	Test time of 3000 hours at a capacity of 1 mAh cm^{-2}, current density of 5 mA cm^{-2}	Excellent performance	Zn-I_2 batteries	[98]
PTA@ZIF-8	2 M $ZnSO_4$	Discharge capacity of 186.5 mAh g^{-1} and a stable cycling performance over 100 cycles	Excellent performance	Zn-I_2 battery	[75]
PCM-NP	$ZnSO_4$	Remarkable stability during 1,200 cycles with a high capacity retention rate (90.7%)	Good stability	Zn-I_2 battery	[90]
Carbon nanostraws (HCNS)	Iodine and a cation exchange membrane	High capacity of 234.1 mAh g^{-1} at 1A g^{-1}	Excellent performance	Zinc-iodine batteries (ZIBs)	[91]
MnO_2	1M $MnSO_4$	Areal capacity of 10 mAh cm^{-2}, as well as a long-term cycling stability of over 950 s and a retention of 94%	Improved reversibility of the MnO_2/Mn_{2+}	MnO_2-Zn battery	[106]
Iodine/carbon cloth	$ZnSO_4$	High capacity (335 mA h g^{-1} at 1 mA cm^{-2}) and long-term cyclic stability (91% capacity retention after 200 cycles	Remarkable flexibility and mechanical robustness	Zn-iodine battery	[59]
Double-layered cathode	ZnI_2	Efficiency (up to 95.6%) and voltage efficiency (up to 91.3%)	Excellent performance	Zinc-iodine batteries (ZIBs)	[107]
Metal-organic gel (MOG-I)	Zinc sulfate, potassium iodide, and sulfuric acid	Delivers high performance and long-term stability with a capacity of 184.9 mA h g^{-1} and a superior capacity retention of	Excellent performance	Aqueous zinc-iodine batteries (AZIBs)	[12]

		95.8% even after 1500 cycles at 1 C rate			
Active carbon fiber cloth (I_2/ACF)	ZnSO4	Capacity of 174.4 mAh g^{-1} at 1C, stable cyclic life over 3000 cycles with ~90% capacity retention	Excellent performance	Zinc-iodine battery	[56]
PAC carbon	$ZnCl_2$ and LiCl	Capacity of 594 mAh g^{-1}, energy density of 750 Wh kg^{-1} and an ultra-long cycle performance of 6000 cycles	Excellent performance	Zinc-iodine battery	[70]
PANI	$ZnCl_2$/H_2O-40GBL	81.2% capacity retention after 10000 cycles at 2 A g^{-1}	Enhanced Zn reversibility and dendrite growth in aqueous electrolytes	Zn-PANI iodine batteries	[108]
Carbon fiber paper (CFP)	Zn-I Water-In-Salt (WIS) on the redox couples of I_2/[ZnIx(OH_2) 4-x]$_2$-x	Specific capacity of 211 mAhg$_{-1,}$ charge capacity of 3.7 mAhcm^{-2} after 300 cycles	Improved energy density and safety application	Dual plating zinc-iodine battery (DPB)	[72]
Prussian blue analogues (PBAs)	2 M ZnSO4	Highest discharge capacity of 236.8 mAhg^{-1}, current density of 0.1 Ag^{-1}, 81.2% capacity after 400 cycles	Excellent cycling performance	Aqueous zinc-iodine (I2//Zn) battery	[109]
Nanopores of conductive accelerated carbonation cured (ACC) fibers	Mild aqueous zinc sulfate	High specific capacity (255 mAh·g^{-1} at 0.5 C) and long-term cyclability (> 1,500 cycles) at ≥ 99% Coulombic efficiency at a high current rate (5 C), capacity retention of 87%	Superior aqueous battery performance	Aqueous rechargeable Zn-I2 battery (ZIB	[110]
Perylenetetracarboxylic diimide (PTCDI),	Potassium chloride/iodine (KCl/I_2)	Discharge capacity retention of about 70% after 900 cycles at 80 mA and 25 ± 1 °C	Improved battery performance and stability	Iodine rechargeable batteries	[111]
Zn	Alginate-based hydrogel	A high capacity retention of 83.1 mAh g^{-1} (72% remained) even after 100 h resting and achieved	Stable battery performance	Zinc-iodine battery	[112]

	an obviously higher reversible capacity (~120 mAh g^{-1})	

3.1.5 Zinc-iodine energy storage system topologies

Grid energy storage demands high energy density, low cost, and long-term cycle stability storage batteries [74]. One solution is to increase the use of earth-abundant charge carriers, which are more ecologically friendly and more compatible with aqueous electrolytes than other metals (such as Li, Na, K, and Mg). High selectivity and permeability membrane design and fabrication have gained popularity due to their natural inspiration. These complex membranes are commonly used in a variety of separation processes. In recent years, their applications have expanded in the area of energy storage, which is essential for widespread use in intermittent and unstable renewable sources [113].

In contrast to conventional zinc-iodine batteries, Zn-I_2 ESS topologies such as flow batteries can provide better scalability and longer cycle life, while hybrid systems can combine the benefits of various battery chemistries to produce more power and higher energy densities. The creation of feasible and effective Zn-I_2 ESS for a variety of applications is anticipated to be significantly impacted by these developments in device architecture design and optimization. Building various zinc-based ESS, such as zinc ion batteries, zinc ion hybrid capacitors, zinc metals, zinc iodine batteries, zinc-air batteries, and so on, has lately sparked considerable attention to fully use the Zn anode's excellent operating safety and theoretical capacity [74]. A different strategy involves investigating various battery topologies, including dual-ion batteries (DIBs) and flow batteries [72]. In flow batteries, a membrane is required for isolating active species and transporting charge-carrier ions [113]. Given that the electrical capacity of a system like a flow battery depends on the quantity of active materials available, scaling up the function is simple by expanding the size of electrolyte tanks [114]. In this regard, Li et al. demonstrated that an M ZnI_2 electrolyte in a Zn/I-RFB solution may provide a discharge energy density of 167 Wh L^{-1}. Weng et al. [114] were able to attain an energy density of 101 Wh L^{-1} posolytenegolyte as catholyte and anolyte in a bromide ion. Composite membranes have emerged as a potential remedy to the issue of selectivity and conductivity due to their distinctly built controlled selective layer and porous support layers. Therefore, composite membranes like metal-organic frameworks (MOF) and covalent organic frameworks (COF) inherent ordered structures have been used in recent years to create specific layers for zinc-iodine ESS [115]. In a study by Wu et al [113], a binder-controlled restrained second-growth method was used to grow UD-66/67 membrane (Fig. 15a) for Zn-I_2 flow battery

(ZIFB), and outstanding electrochemical performance was attained. The MOF layers produces more uniform zinc deposition with better reversibility and dissolution as demonstrated by the schematic illustration (see Fig. 15b) of hydrated cations passing through UD-66 [113]. Similarly, carbon felts and Ti mesh were used as cathodes and a Zn-plated Fe mesh as the anode in a polyiodide battery that had a positive electrode surface but no solvent to prevent structural alteration. A durapore membrane was sandwiched between the electrodes, and the charge-discharge was 20 mA cm^{-2} with electrolytes flowing at 200 mL per minute at 27°C. The additive, propylene carbonate (PC) increased the discharge capacity to 22.7 Ah L^{-1} and the coulombic efficiency to over 90% over 50 cycles [114] (see Fig. 15c).

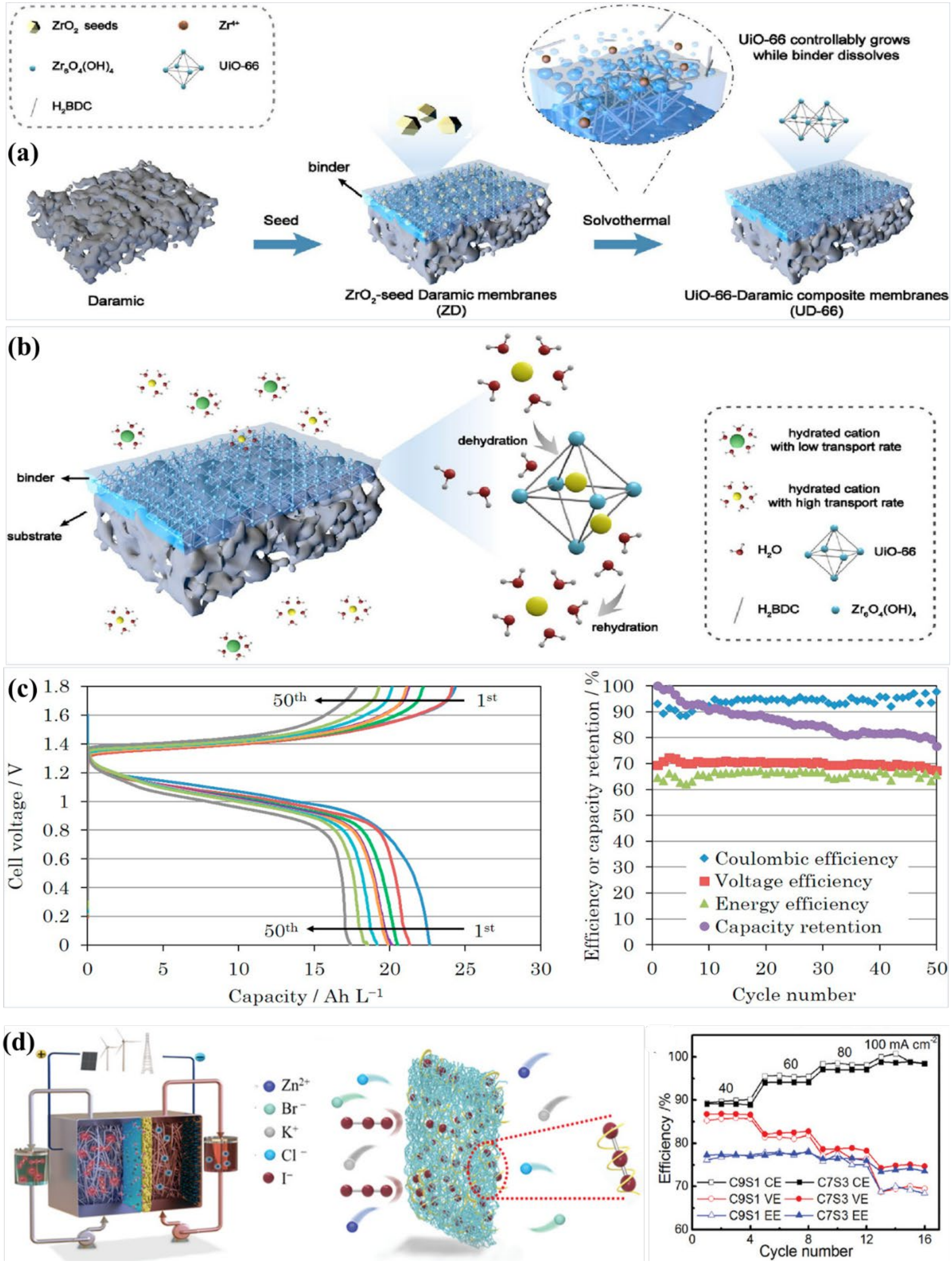

Fig 15. (a) Illustration of binder-controlled restrained second-growth of UiO-66 on duramic (b) An illustration of hydrated cations passing through UD-66. Reprinted with permission from Elsevier [113] (c) Charge-discharge curves of the single electrolyte-type Zn/I-RFB measured at a current density of 20 mA cm^{-2} through 50 cycles. Reprinted with permission from Elsevier [114], (d) Schematic of ZIFB and modus operandi of the chitosan-starch composite membrane as well as rate capability of the ZIFB assembled composite membrane. Reprinted with permission from Wiley and Sons [116].

The performance of a ZIFB with a supplemental ammonium iodide electrolyte, a zinc chloride anolyte, and an iodide evolution process at the cathode was examined [6], and tremendous cycling stability (1,100 cycles) at a high current density of 80 mA cm^{-2} with 82% increase in energy efficiency. In another study, a flow battery's trifunctional electrolyte was examined, and it was found that zinc iodide dispersed in NH_4Br electrolytes acted as the active material. This provided an enhanced capacity of 35 A h L^{-1} with an energy efficiency of 85% and was consistently cycled for more than 100 cycles without short circuits [117]. Similarly, Liu et al. [118] utilized a bottom-up approach in creating a graphene quantum dot (GQD) coated graphite felt (GQD-GF) electrode for a Zn-I_2 redox battery. The static Zn-I_2 redox battery built using GQD-GF can function at large currents while exhibiting outstanding energy density, Coulombic efficiency, and cycle stability. In a similar way, a Zn-I_2 hybrid supercapacitor (Z-IH) has been designed that incorporates redox iodide ion into the $ZnSO_4$ electrolyte to increase energy density using B, N dual-doped porous carbon microtubes (BN-CMTs) as a cathode. This design facilitates the faradaic reaction on the electrode surface by changing the electronic structure and density state of carbon. The BN-CMT-based Z-IHS had an incredibly high capacity of 416.6 mA h g^{-1}, a high energy density (472.6 W h kg^{-1}) at a power density of 1600 W kg^{-1} in the voltage range of 0.2-1.8 V, and good cycling stability with capacity retention of 99.1% over 10 000 cycles at 10 A g^{-1}. [119]. According to Jang et al. [120], iodine coating affects charge-transfer resistance when iodide is electro-oxidized in redox flow batteries. To overcome the current restrictions of the low utilization and shuttle effect of iodine species for Zn iodine RFBs, Xie and co-workers [31] designed a low-cost self-healing porous polyolefin membrane using stable electrolytes (KI, $ZnBr_2$, and KCl) that fills the pores of the membrane with a solution containing I_3^- that can react with zinc dendrite and consume it, allowing the battery to self-recover from micro-short-circuits caused by overcharging. At 80 mA cm^2, ZIFB had a high energy efficiency (82%) and an output of 700 W after 300 continuous cycles. Analogous to Xie's design, Gao et al. [116] highlight the construction and operation of ZIFB with two electrolyte tanks with a chitosan-starch composite membrane between the anode and cathode compartments. At a current density of 80 mA cm^{-2}, the membrane significantly increases selectivity, yielding a Coulombic efficiency of 98.6% and an energy efficiency of 77.4% (see Fig. 15d). The membrane is a great choice for RFBs because it also offers adjustable hydrophilicity/hydrophobicity and reduced cost. Similarly, an all-aqueous hybrid alkaline Zn-I_2 flow battery, exhibiting a 330.5 W h L^{-1} energy density has been reported [58]. To increase the battery's voltage, the authors suggest the adjustment of the electrolyte's pH using a triiodide/iodide. Their design demonstrated a high coulombic efficiency with an energy density of 70% over 200 hours [58]. Yang et al. [121] stabilized iodide ions in Zn-I_2 flow batteries using PVP with a coulombic efficiency of 99%. Table 7 presents various cell configurations, merits, and demerits as well the capacitance of the various zinc-iodine ESS.

Table 7: Cell configurations, unique features, and capacitance of zinc-iodine ESS

Novel cell configuration	Characteristics	Advantages	Disadvantages	Capacitance	Ref.
Hybrid battery system	-Iodine loading of 2.0 mg/cm^2 on each electrode. -Whatman filter paper as separator. -Good electrode-electrolyte interface. -Sharp pattern of high reversibility.	-Generates additional capacity through reversible adsorption-desorption process. -It is energy-efficient. -They involve low maintenance cost	-Relatively expensive. -They are complicated to install. -Battery life is shorter. -Process control is difficult.	A capacity retention of 97.2 %	[74]
Hybrid battery supercapacitor (Z-IHS)	-Hollow structures with irregular rectangular openings. -Favourable ion diffusion during charge/ discharge process	-Excellent behavioural properties -Has a fast charge/discharge potency. -It involves reduced cost.	-It has a low energy density. -There is limitation with the full energy spectrum. -It has a high self-discharge potency. -There is the need for voltage balancing.	Capacitance retention of 99.1% over 10 000 cycles	[119]
Flow battery	-It has a porous polyolefin membrane. -Formation of the black solid on the positive graphite felt electrode -Battery achieves significant polarization after 35 cycles	-Excellent performance -Flexible layout -It has long cycle life.	-It has a low cycle energy efficiency. -Dendrite formation leading to short circuiting	High capacity retention and coulombic efficiency around 95%	[114] [122] [116] [31] [53]
Ammonium Chloride Zinc-Iodine Redox Flow Battery (AC-ZIFB)	-Decoupled electrolyte design system. -Chlorine ions inhibiting I_2 precipitation. -Presence of polyethylene (PE) membrane	-Excellent energy potential. -It has an independently scalable performance. -It has a long life span with an unlimited cycle number. -Electrolytes are not flammable	-It has a low energy density. -Undesired electrolyte transition through membranes is likely. -Temperature operations are lower permissible	Capacity retention was nearly 100%	[6]

Zinc-iodine single flow battery (ZISFB)	-Carbon felt material with high porosity (>90%), filled with high concentration electrolyte. -High specific capacity and a super high energy density. -graphite felt electrode and porous polyolefin as a separator	-Super high energy density and long cycling life. -It has an independently scalable performance. -It has a long life span with an unlimited cycle number. -Electrolytes are not flammable	-It has a low energy density. -Undesired electrolyte transition through membranes is likely. -Temperature operations are lower permissible	An areal capacity running for more than 500 cycles	[59]
Symmetric cell	-A home-made cell with a three-electrode system. -A cation-exchange membrane (Nafion 211) -Two symmetric cells for the redox reaction at a constant capacity of 0.715 A·h	-High current and power density. -It has design flexibility. -It has an enhanced cycle life.	-Exhibits even lower energy density. -It has limited energy retention -It has a limited voltage range.	Constant capacity of 0.715 A·h	[120]

3.1.6 Separator membrane for Zn-I_2 batteries

The separator in a zinc-iodine battery plays a crucial role in preventing direct contact between the anode and cathode while allowing the passage of ions. In most button batteries, glass fiber (GF) is the most utilized separator [123]. Nonetheless, due to the dreadful shuttle effect of polyiodides, it is frequently ineffective in Zn-I_2 ESSs [124]. Microporous polyethylene (PE) or polypropylene (PP) membranes are the most utilized separator materials in zinc-iodine batteries [125]. These membranes are suited for battery applications due to their high ion conductivity, chemical stability, and mechanical strength. They effectively inhibit zinc particle migration from the anode to the cathode, which can create short circuits [126]. A study by Zhang et al. [127] investigated the performance of different separators in Zn-I_2 batteries. They found that microporous PE membranes exhibited superior electrochemical performance compared to other separator materials, such as glass fiber and cellulose acetate. The PE membranes showed higher energy efficiency, lower self-discharge rates, and improved cycling stability. Another research by Li et al. [128] focused on improving the performance of Zn-I_2 batteries through the modification of separator materials. They developed a composite separator by coating a layer of polydopamine (PDA) on a microporous PE membrane. The PDA layer enhanced the wettability of the separator, resulting in better contact between the electrolyte and electrode materials. This modification led to improved battery performance, including higher energy density and longer cycle life.

Nafion 117 membrane was also employed as a separator in zinc-iodine batteries [129]. It was shown that immobilized sulfonic acid groups on the Nafion membrane can restrict I_3^- ions in the catholyte, considerably reducing the shuttle effect. notwithstanding, the "only plugging but not draining" technique could impede ion transport and result in delayed reaction kinetics. As a result, the most important consideration for selecting an excellent separator entails choosing a material that could facilitate the transport of Zn ions, promote the reversible deposition/stripping of zinc, accelerate reaction kinetics, and inhibit the shuttle of I_3^- and I_5^- ions via space restriction or charge interaction. Nevertheless, achieving synergistic effects on Zn anode and iodine with straightforward and low-cost membrane architecture is difficult [130]. It has been demonstrated that low-cost porous polyolefin membranes can effectively eliminate the accumulation of zinc dendrites with a self-healing effect after micro-short-circuiting as a result of overcharging is feasible [31].

3.2 Challenges and opportunities in Zn-I_2 energy storage systems

The fabrication of Zn-I_2 batteries presents several challenges that must be overcome to improve their efficiency and commercial viability. One major challenge is the low solubility of iodine in the electrolyte, which can lead to poor performance and low energy density [34]. This issue can be addressed through the development of novel electrolytes with improved solubility and stability. Another challenge is the corrosion of the zinc anode, which can lead to reduced battery performance and safety concerns [131]. To mitigate this issue, various coatings and additives have been investigated to improve the stability and durability of the anode materials.

Furthermore, the design of the cathode material is critical for the overall performance of the battery. Iodine has a relatively low conductivity, which can limit the rate of charge and discharge. Additionally, the formation of solid iodine can lead to cathode passivation and capacity loss [8]. To overcome these challenges, various strategies have been proposed, including the use of carbon-based materials as cathodes and the incorporation of conductive additives.

Also, the scalability and cost-effectiveness of the fabrication process remain important considerations for commercialization [132]. While various methods have been developed for the synthesis of electrode materials, electrolytes, and device architectures, further optimization is needed to reduce the cost and improve the efficiency of large-scale manufacturing processes.

Currently, the cost of producing Zn-I_2 batteries is relatively high compared to other types of batteries, mainly due to the cost of iodine and the complex design of the battery.

The issue of limited cycle life is a major challenge with Zn-I_2 ESS [31]. While zinc-iodine batteries have a high energy density and good power output, they also have a relatively limited cycle life. Over time, the performance of the battery can degrade due to the accumulation of unwanted side reactions. More so, the electrolyte used in zinc-iodine batteries has a high viscosity, which can cause flow problems and reduce the battery's overall efficiency. Besides that, iodine is a hazardous material that requires careful handling and disposal. If the battery is damaged or punctured, it can release toxic chemicals posing serious risks to human health and the environment.

In this age of wearable electronics, specific challenges associated with the application of Zn-I_2 batteries for this market niche is the issue of size, weight, and limited cycling life - wearable technology requires small, lightweight with long cycling batteries that can be integrated seamlessly into the device, however, Zn-I_2 batteries are relatively large, heavy and have limited cycling life compared to other types of batteries, which makes them less suitable for use in wearable technology [63, 133]. There is, therefore, the need for continuous research in this area to tap into the advantages of Zn-I_2 batteries for wearable technology. The subject of flexibility is another major setback of Zn-I_2 batteries. Many wearable devices require batteries that can be flexed or bent without losing their functionality. In this regard, some significant progress has been made, however, there is a need for more focused research to overcome this limitation. More so, wearable devices require batteries that can deliver high power output in a short time. Zn-I_2 batteries have a relatively low power density compared to other types of batteries when miniaturized, which makes them less suitable for high-power applications. Similarly, wearable devices require batteries with a long cycle life that can be recharged quickly and efficiently. Zn-I_2 batteries have a relatively short cycle life compared to other types of batteries, which makes them less suitable for use in wearable technology. Also, the cost of producing miniaturized zinc-iodine batteries is relatively high compared to other types of batteries, which makes them less economically viable for use in wearable technology.

Despite these challenges, research is ongoing to overcome and improve the performance and cost-effectiveness of zinc-iodine batteries. In this regard, it is clear from this review that new materials and designs are being developed to improve the cycle life and efficiency of the battery, while also reducing costs. In addition, advances in technology and production processes are expected to make the scaling up of zinc-iodine batteries more feasible in the future. Regarding the application of Zn-I_2 batteries for wearable technology, research is ongoing to improve the performance and suitability of zinc-iodine batteries for wearable technology. For example, new materials and designs are being developed to improve the flexibility and power density of the battery, while also reducing its size and weight [63]. In addition, advances in manufacturing and production processes are expected to make zinc-iodine batteries more cost-effective in the future. Currently, while Zn-I_2 batteries may not be the best choice for all wearable technology applications, they do show promise for use in certain applications where their unique properties can be leveraged effectively.

Generally, Zn-I_2 batteries show great potential as a viable alternative to traditional lithium-ion batteries, and continued research in this area is crucial for advancing renewable energy technologies. One of the potential applications of zinc-iodine batteries is in electric vehicles. These batteries have a high energy density, which means they can store a large amount of energy in a relatively small space. This makes them well-suited for use in electric vehicles, where energy density is a key factor in determining the vehicle's range and performance. Another potential for Zn-I_2 battery application is in renewable energy storage. As renewable energy sources such as solar and wind power become more prevalent, the need for efficient and cost-effective energy storage systems increases. Zn-I_2 batteries could provide a solution to this problem, as they are relatively inexpensive and have a long cycle life.

Zn-I_2 batteries also have a huge commercial application in portable electronics such as smartphones, tablets, and laptops. Their high energy density and low cost make them an attractive option for powering these devices, especially as the demand for portable electronics continues to grow.

Briefly, Zn-I_2 batteries have the potential to be a versatile and cost-effective energy storage solution for a range of applications, including electric vehicles, renewable energy storage, and portable electronics. However, it is important to emphasize that the fabrication of zinc-iodine batteries is a complex and multi-disciplinary endeavor. Addressing these challenges is crucial for the development of Zn-I_2 batteries as a viable alternative to conventional energy storage systems.

Development trends in Zn-I_2 *Materials and Electrolytes research*

To unlock the full potential of Zn-I batteries, the development of suitable materials and electrolytes is of paramount importance. The key trends in the development of zinc-iodine materials and electrolytes and their significance in advancing efficient and sustainable energy storage solutions are discussed briefly.

A crucial aspect of Zn-I_2 battery development lies in the improvement of materials. Further research into advanced electrode architectures is needed to enhance the electrochemical performance of zinc-iodine batteries [9]. By employing novel designs that suppress dendritic growth or utilize nanostructured morphologies, the issues related to dendrite formation and volume expansion during cycling can be adequately addressed. In addition, alloying zinc with other metals and incorporating additives has shown promise in improving electrode stability and cycling performance [134]. Elements like bismuth and tin, as well as conductive polymers, have shown great potential to enhance the overall battery performance.

The development of iodine electrodes is another critical area of focus. Iodine reduction reactions tend to be slow, leading to poor kinetics and reduced energy efficiency. To overcome this challenge, more focused research on the exploration of different catalysts such as new carbon-based materials, metal nanoparticles, and conducting polymers is necessary [135]. These catalysts accelerate iodine reduction kinetics, ultimately enhancing the battery's overall performance. Furthermore, the design and synthesis of iodine-based materials with tailored properties are being explored to optimize solubility, stability, and reversible redox reactions, thereby improving energy storage capabilities.

The electrolyte design plays a vital role in facilitating ion transport and maintaining electrochemical stability in Zn-I batteries. In the case of aqueous electrolytes, optimizing the pH and ionic strength has been a key focus [136]. By adjusting these parameters, it can influence the solubility, conductivity, and electrochemical reactions of zinc and iodine species. pH-buffering systems, as well as the composition and concentration of salts, are actively investigated to improve the performance and stability of aqueous electrolytes [137]. Furthermore, the introduction of additives and complexing agents has shown promise in enhancing the solubility and stability of zinc and iodine species and suppressing side reactions. Organic additives such as polyethylene glycol and surfactants are being explored for their impact on the properties of the electrolyte. Non-aqueous electrolytes, on the other hand, offer advantages such as wider electrochemical stability windows and higher ionic conductivity. More research is needed in exploring various organic solvents and ionic liquids as potential alternatives to aqueous electrolytes. These alternative electrolyte media can improve the energy density, stability, and safety of Zn-I batteries. Additionally, efforts are being made to develop ion transport enhancers that can improve ionic conductivity and suppress reactivity at the electrode/electrolyte interfaces. Strategies may involve the use of lithium salts, solid-state electrolytes, or surface passivation techniques to achieve these objectives. For instance, gel electrolytes have emerged as a promising alternative for Zn-I_2 batteries, offering advantages such as improved safety, enhanced stability, and simplified battery design. These gel electrolytes, typically composed of a polymer matrix and an ionic liquid or salt, which exhibit unique properties that contribute to the development of efficient Zn-I_2 ESS [138]. Research into gel electrolytes for Zn-I_2 batteries is still in the infant stages, hence, more focused research is needed in this area.

The development of zinc-iodine materials and electrolytes is a fast-paced field of study driven by the search for efficient and sustainable energy storage solutions. Material advancements in zinc and iodine electrodes are centered on addressing problems such as dendrite growth, volume expansion, and sluggish kinetics. Electrolyte development, whether in aqueous or non-aqueous systems, strives to maximize solubility, stability, and ion transport properties by optimizing pH and ionic strength, using additives, and exploring alternate electrolyte media. These trends all contribute to the ongoing efforts to develop efficient and long-lasting zinc-iodine battery technology for future energy storage applications.

Limitations of the study

Despite the rigorous methodology and the transparency in the analysis of this write-up, the authors acknowledge that there could be other limitations. Time lag is the first limitation identified because per the time frame of this review, there is a clear cut-off date for the inclusion of studies. Newer research and advancements in zinc-iodine batteries beyond the cut-off date may not be captured, potentially limiting the review's currency and applicability to the most recent developments in the field. The review's findings heavily depend on the availability and accessibility of relevant scientific literature from only one database, thus Scopus; there might be limitations in accessing certain publications due to language barriers, or unpublished research. Even in Scopus, only English papers were selected whereas the authors acknowledge that there are dedicated databases in other languages than English. Therefore, the geographical alliances and research output by states, individuals, and organizations in this article may not be definitive due to reasons already mentioned such as the limited database (only Scopus) used, publications in other languages than English, medium of publication, and the time lag.

4. Conclusion

In recent years, the need for energy storage solutions has increased as the demand for renewable energy sources has grown. Zinc-iodine batteries have emerged as a promising alternative to traditional lithium-ion batteries, offering high energy density, low cost, and improved sustainability. This *sciento-qualitative* review has analyzed the research trends and recent advances in zinc-iodine energy storage systems using 161 publications obtained from the Scopus database. The data reveals a growing interest in the development of zinc-iodine batteries, with increasing numbers of research publications, and collaborations especially in Asia with China having the highest number of publications and spearheading most of the collaborations. This work provides important insight into institutions, states/regions that are spearheading research in zinc-iodine ESS. This will help foster collaboration and promote knowledge sharing in a bid to expand the frontiers of knowledge and hasten the pace of research for the commercialization of zinc-iodine ESS in the near future.

Concerning frontier research in Zn-I_2 ESS, significant strides have been made in the areas of electrode material advancements, electrolyte tuning, and hybrid device designs, which have improved system performance and longevity. Zn-I_2 ESS has great potential for various applications, including electric vehicles, renewable energy storage, and portable electronics. The high energy density of Zn-I_2 ESS makes them particularly suitable for electric vehicles, where longer range and lower weight are critical factors. In renewable energy storage, zinc-iodine batteries can help to smooth out fluctuations in energy supply and demand, improving the reliability and stability of the grid. Despite the progress made, there are still some challenges to be addressed in scaling up zinc-iodine batteries for commercial use. These include improving their durability, reducing self-discharge rates, and addressing issues related to the corrosive nature of the electrolyte. However, research and development in this area are advancing rapidly, and there is growing optimism that these challenges can be overcome. The advances made in recent years provide a strong foundation for further research and development, with the potential for Zn-I_2 batteries to play a key role in the transition to a more sustainable and renewable energy future.

Supplementary Materials: The following supporting information can be downloaded at: https://www..............,Table S1. List of publication sources with at least 2 articles in the relevant field; Table S2. List of the top 27 most frequently occurring keywords in the relevant field of study; Table S3. Full list of the top contributing authors in the relevant field of study; Table S4. Table S5: Full List of World Collaboration Map of Country Involvements “From” and “To”

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GRAPHIC ABSTRACT