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Review

The latest research trends in the removal of cesium from radioactive wastewater: A review based on data-driven and visual analysis

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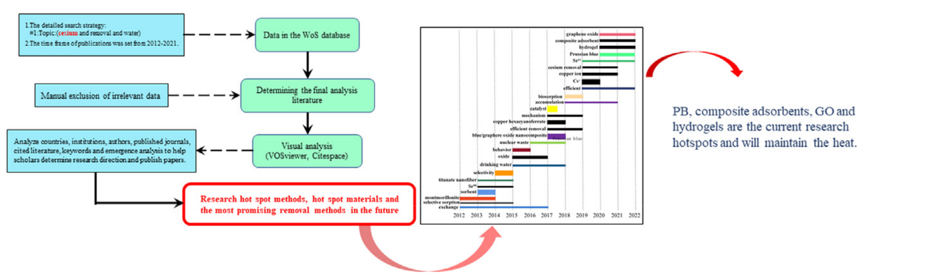
HIGHLIGHTS

- Summarize the published articles.
- Provide reference value for Cs removal.
- Point out the hot spots of Cs removal research.

GRAPHICAL ABSTRACT



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ABSTRACT

The widespread adoption of nuclear energy has increased the amount of radioactive cesium (Cs) that is discharged into waste streams, which can have environmental risks. In this paper, we provide a comprehensive summary of current advances in aqueous Cs removal by employing a bibliometric analysis. We collected 1580 articles related to aqueous Cs treatment that were published on the Web of Science database between 2012 and 2022. By applying bibliometric analysis combined with network analysis, we revealed the research distribution, knowledge base, research hotspots, and cutting-edge technologies in the field of aqueous Cs removal. Our findings indicate that China, Japan, and South Korea are the most productive countries with respect to Cs removal research. In addition, both historic events and environmental threats might have contributed to research in Asian countries having a higher focus on Cs removal as well as strong international cooperation between Asian countries. A detailed keyword analysis reveals the main knowledge base for aqueous Cs removal and highlights the potential of the adsorption-based method for treating Cs contamination. Furthermore, the results reveal that exploration of functional materials is a popular research topic in the field of Cs removal. Since 2012, novel materials, including Prussian blue, graphene oxide, hydrogel and nanocomposites, have been widely investigated because of their high capacity for Cs removal. On the basis of the detailed information, we report the latest research trends on aqueous Cs removal, and propose future research directions and describe the challenges related to effective Cs treatment. This scientometric review provides insights into current research hotspots and cutting-edge trends in addition to contributing to the development of this crucial research field.

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Contents

1. Introduction

Rapid economic development has created an ever-increasing demand for energy worldwide. As a form of renewable energy, nuclear energy has been considered a lot for applications in various fields because of its low carbon emissions, high efficiency, and low cost (Hwang et al., 2020; Lovering et al., 2016). Although the exploitation and development of nuclear energy have been promoted globally, diverse environmental concerns, such as the generation of nuclear waste and radionuclide contamination, have emerged because of the use of nuclear power (Chen et al., 2019; Manolopoulou et al., 2011). The release of radionuclides (e.g., ^{137}Cs, ^{90}Sr, ^{235}U, ^{131}I, and ^{57}Co) into the surrounding environment poses a severe threat to public health; thus, safe disposal of nuclear waste is a crucial key for the sustainable development of nuclear energy (Liu et al., 2021; Rethinasabapathy et al., 2021; Rethinasabapathy et al., 2019; Tokunaga et al., 2018; Wang et al., 2019; Yuan et al., 2018).

Among the radionuclides that are frequently encountered in aqueous radioactive waste streams, ^{137}Cs is a high-level waste constituent posing considerable environmental and health risks. Nuclear wastewater from the Fukushima nuclear accident in Japan contains radioactive cesium (^{137}Cs) up to 200 Bq/mL, which is much higher than the safety standard value set in Japan and around the world. The large threat posed by ^{137}Cs is related to its long half-life (ca. 30 years), high solubility in water, and radioactivity as a gamma emitter. The poor degradation and facile bioaccumulation of ^{137}Cs may cause somatic and genetic problems for living organisms, as evidenced by the Chernobyl and Fukushima accidents in 1986 and 2011, respectively (Audi et al., 2003; Borai et al., 2009; Li et al., 2020). These two events led to the release of immense amounts of harmful radionuclides into waste streams, which have seriously damaged the surrounding environment and affected public health (Kim et al., 2018; Sylvester et al., 2013; Vetter, 2020; Yin et al., 2017). In addition to soluble ^{137}Cs contaminants, some radioactive ^{137}Cs-rich particles can be inhaled or ingested and thus pose a severe threat to human health (Suetake et al., 2019). The inhaled particles become deposited in the alveolar space, whereas the ingested particles are completely absorbed from the intestine and distributed to the soft tissues throughout the body along the circulatory system (Sangvanich et al., 2010a). Cesium (Cs) has been reported to be the leading cause of thyroid cancer (Kirillov et al., 2003). Because of the high solubility and radioactivity of ^{137}Cs, removing soluble ^{137}Cs contaminants from waste streams is of a major global research interest.

A number of review articles on Cs removal have been published in the past decades, highlighting the need for a systematic overview that summarizes the overall research direction and identifies the research hot spots and cutting-edge trends of Cs removal (Chen et al., 2020; Lalhmunsiama et al., 2018; Lehto et al., 2019; Rauwel and Rauwel, 2019; Wang and Zhuang, 2019). Bibliometric analysis is a systematic method for analyzing large volumes of research and identifying publication patterns through mathematical and statistical techniques (Donthu et al., 2021). This method can be used to evaluate the latest scientific activities from three perspectives: quality, quantity, and networks. Qualitative analysis mainly involves the quality and impact of research by, for example, identifying the current topics in a research area and research hotspots, whereas quantitative analysis involves determining the level of research activity by using quantitative indicators, such as the total research output in a given field and the number of publications by a researcher. Network analysis involves applying relationship indicators to investigate the connections and interrelations between different keywords, institutions, and countries. Bibliometric analysis enables effective identification and analysis of the contributions from diverse authors, journals, institutions, and countries to elucidate the development of a particular research field. This method also yields a clear statistical map of the research field and enables researchers to extrapolate future research directions (Durieux and Gevenois, 2010; Ellegaard, 2018). Data from bibliometric analysis can guide researchers and institutions in making evidence-based decisions regarding research priorities as well as in adopting novel research strategies; this knowledge also enables policy makers and research funders to evaluate the prospects of research projects.

In this paper, we reveal the current research hotspots pertaining to aqueous Cs removal, and point out the key concerns in Cs treatment processes, and provide future directions for developing and optimizing new techniques for effective Cs removal from contaminated water.

2. Data and methods

2.1. Data collection

Literature retrieval was performed using the Web of Science (WoS) database. To understand the recent developments in Cs removal technologies and predict research trends, we considered articles published between 2012 and 2022. We used “cesium,” “removal,” and “water,” as search terms and included only English publications. After detailed screening, 1580 records were obtained. The results were then downloaded in “full record and reference” form, and the following information was extracted from each article: title, abstract, author, author keyword, state, organization, journal, and references. Fig. 1 presents the literature retrieval and selection process.

2.2. Data analysis

VOSviewer was used to analyze the retrieved papers (van Eck and Waltman, 2010). This scientific knowledge visualization tool principally employs co-occurrence clustering. Relevant bibliographic information is provided through cooperation analysis, keyword co-occurrence analysis, coupling analysis, and other functions. We applied a three-step process to compile bibliographic information:

Step 1: The similarity matrix is calculated using the normalized co-occurrence matrix, and the similarity measure is used to represent the association strength. The similarity between two elements (s_{ij}) (e.g., keywords, countries, and authors) is therefore defined as follows:

$$S_{ij} = \frac{c_{ij}}{w_i w_j} \tag{1}$$

where c_{ij} is the number of co-occurrences between elements i and j, with w_i and w_j representing the total number of occurrences or co-occurrences of elements i and j, respectively.

Step 2: The similarity matrix obtained in the previous step is then computed using VOSviewer. In this step, we determine the minimum weighted sum of the squared Euclidean distances between all pairs of elements, and the calculations are listed as follows:

$$\min\ V(x_1, \ldots x_n) = \sum_{i<j} s_{ij} \|x_i - x_j\|^2 \tag{2}$$

$$\text{subject to } \frac{2}{n(n-1)} \sum_{i<j} \|x_i - x_j\| = 1 \tag{3}$$

where $V(x_i) = (x_{i1}, x_{i2})$ is the location of element i in a two-dimensional map, $||\bullet||$ is the Euclidean norm, and n is the number of elements to be mapped.

Step 3: Eq. (2) is transformed through translation, rotation, and reflection to verify the results.

Each node or link map generated through VOSviewer represents a bibliometric network of objects, with a focus, generally, on one type of object. The objects can be articles, countries, research institutions, authors, or keywords. VOSviewer links these items from the WoS database files to the network through bibliographic linking, citation, co-authorship, co-occurrence, or co-citation (van Eck and Waltman, 2017). The strength of the linkage is expressed by a quantitative indicator, with a higher value suggesting a greater strength. These visualization maps can be used to analyze future directions in a research field.

3. Bibliographic analysis of publication data

3.1. Yearly quantitative distribution of literature

Fig. 2 presents the annual number of articles and six major WoS categories in the publications of 2012–2022. The number of papers increase from 58 in 2012 to 141 in 2022, with a peak number of publications observed in 2020 (242 articles). The steady annual growth in publications on Cs removal highlights the increased public attention and research interest in treating radioactive contaminants. Our analysis of the publication sources also revealed that more than half (1060) of the papers were published in the fields of ecological and environmental sciences.

3.2. Quantitative analysis of producing countries and major research institutions

Our analysis indicated that 78 countries contributed to research on Cs removal technologies. **Table S1** lists the 10 most productive countries with respect to Cs removal research; publications from these countries represented 76.34 % of the total number worldwide. Among these countries, China ranked first in publication output, with 495 published articles (31.33 %) and 10,995 citations (27.67 %). The second most productive country was Japan, with 288 published articles (18.23 %) and 7440 citations (18.72 %). South Korea ranked third, with 224 published articles

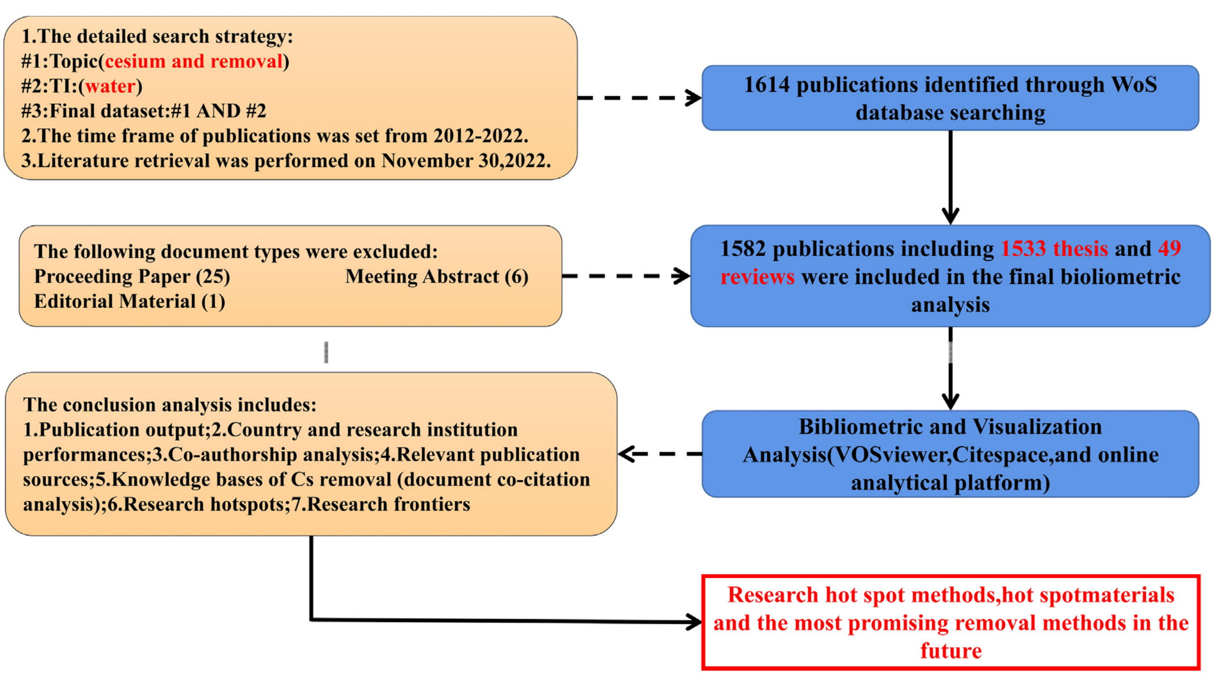


Fig. 1. Literature retrieval flowchart.

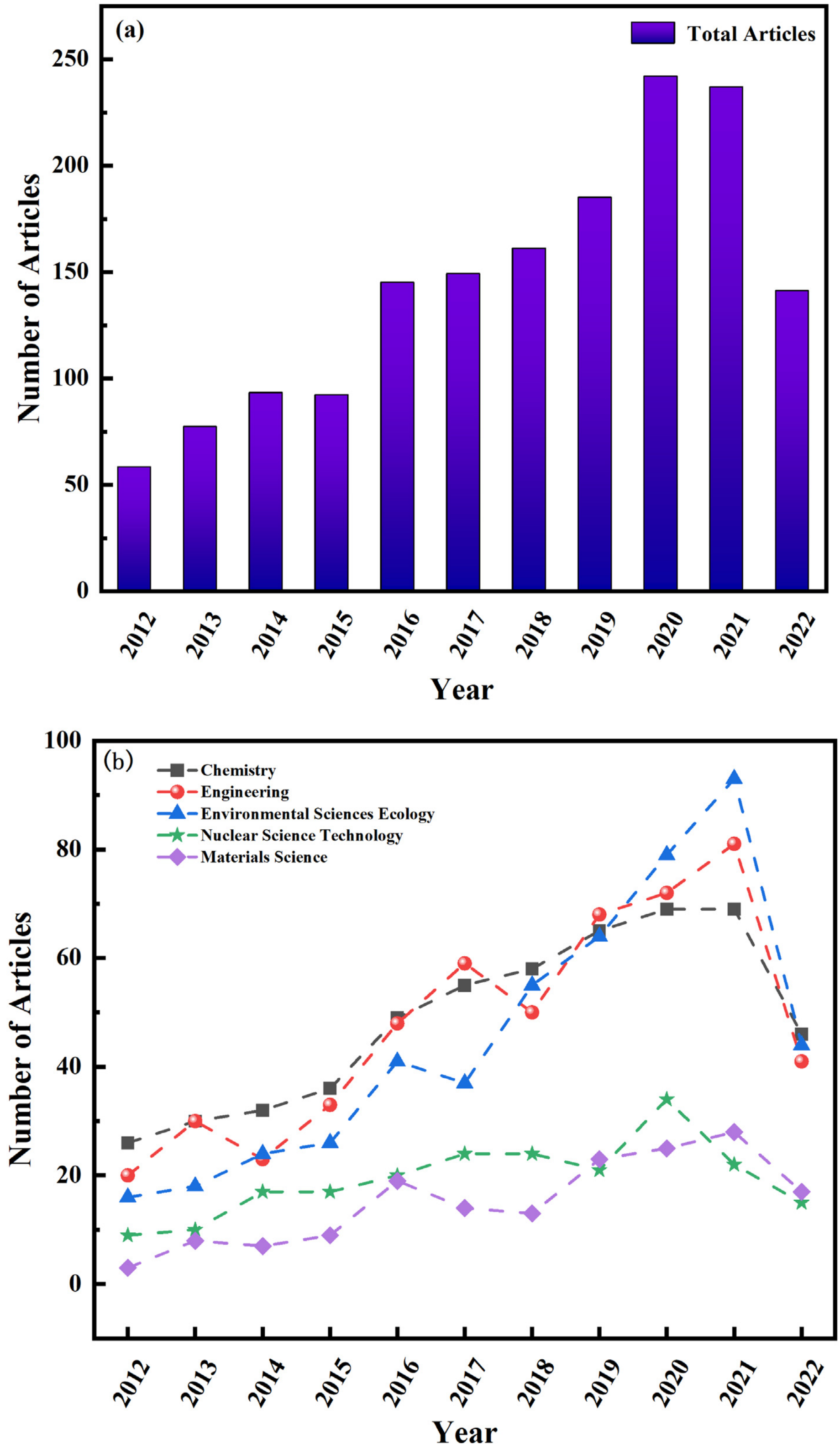


Fig. 2. Annual scientific production: (a) total and (b) WoS top categories.

(12.11 %) and 3705 citations (9.44 %). The publications and citations of China, Japan, and South Korea accounted for more than half of the global amount, whereas the countries ranked 4th to 10th were responsible for only 31.20 % of the published articles and 22.08 % of the citations in the world. To analyze the research collaboration between countries, we used VOSviewer to determine the country distribution of Cs removal publications (Fig. 3). We analyzed the research connections between the 35 countries that had published at least eight relevant articles. In Fig. 3, the influence of a country in the research field is reflected by the size of the node, and the degree of cooperation between countries is indicated by the thickness of the link. Our results revealed that the four most productive countries were China, Japan, South Korea, and Egypt; the linking results

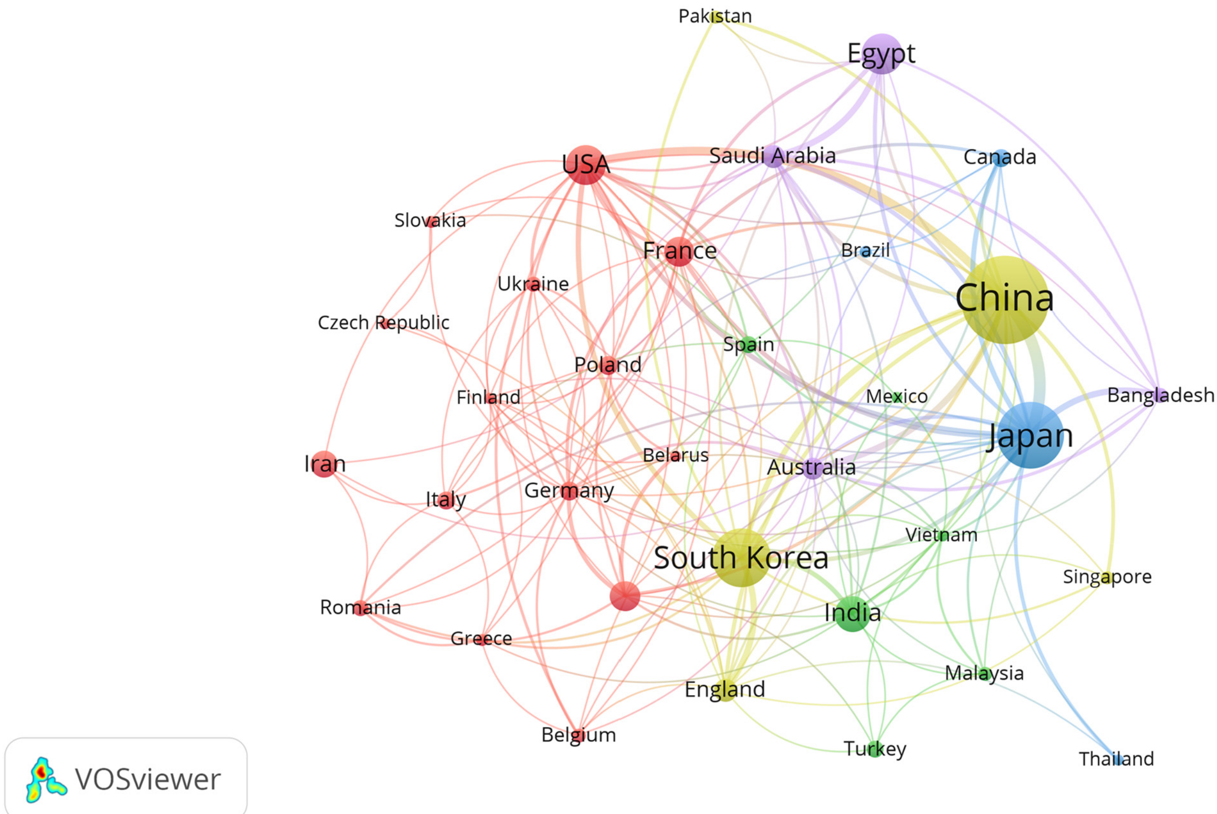


Fig. 3. Collaboration network between countries for Cs removal research.

revealed that Asian countries cooperated more frequently than did countries in other regions. These findings suggested that Asian countries have been more focused than countries in other continents on researching radioactive containment technologies; this may be attributed to historical reasons and current environmental threats.

To explore the institutional contributions of and collaborations between research institutions on Cs removal research, we extracted the information of 1580 institutions from the retrieved articles. **Table S2** lists the 10 most productive institutions with respect to Cs removal research; the Chinese Academy of Sciences was the most productive institution, producing highly cited publications related to Cs removal. Although the Japan Atomic Energy Research Institute (JAERI) published only 31 articles over the target period, these publications were cited 1768 times, leading the JAERI to rank second among the research institutions. This high number of citations for papers published by JAERI indicates that the research from the institution is insightful and has practical applications for treating Cs-contaminated water. The strong research focus on radioactive waste treatment in Japan may be related to the Fukushima nuclear power plant accident.

To obtain a clearer perspective on the international cooperation on the research topic, we conducted network analysis of global research institutions working on Cs removal. Each node in Fig. 4 represents an institution, and each line connecting the nodes represents a partnership between two research institutions. To ensure only the most relevant institutions were represented in the figure, we included only those with at least 10 relevant publications; 45 research institutions met this requirement. Only minor variations were observed in the node sizes in Fig. 4, indicating the research institutions working on aqueous Cs removal were uniformly distributed. According to our results, the Chinese Academy Sciences was ranked highest, with 11 links and a link strength of 47; the University of Tsukuba was ranked second, with 10 links and a link strength of 24; and the National Institute of Advanced Industrial Science and Technology was ranked third, with 8 links and a link strength of 33. Two of the three institutions mentioned above are Japanese, highlighting the active participation of this country's institutions in international cooperation.

3.3. Co-authorship analysis of main research groups

Co-authorship analysis provides information on author productivity in a given field, which can serve as the basis for further analysis of partnerships and collaborations. We analyzed the authorship contributions of the 1580 papers reviewed in this study. Our results indicated that 5047 authors published only one article, 602 published two articles, and 627 published three or more articles. To visually represent the author contributions and productivity, we used VOSviewer to visualize the distribution and network of publishing researchers (Fig. 5). In this analysis, we set the minimum number of coauthors and cited authors to 5, and 178 prominent researchers reached this threshold. Three researchers, namely Park Chan Woo, Lee Kune-Woo, and Yang Hee-Man, were highly productive of Cs removal research, and they collaborated extensively with other researchers (Fig. 5).

3.4. Quantitative analysis of main source journals

The 1580 articles we retrieved were distributed among 391 journals, with 235 journals (60.1 %) publishing only one article, 51 journals (13.0 %) publishing two, and 105 journals (26.9 %) publishing three or more. We ranked the 391 journals according to the number of citations their publications received; the top 15 journals are listed in Table 1. *Chemical Engineering Journal* published 102 papers on the removal of Cs in water, and papers in this journal had the most citations (4660), indicating this journal is important in this field. Among the papers published in *Chemical Engineering Journal*, the Radioactive cesium removal from nuclear wastewater by novel inorganic and conjugate adsorbents by Awual et al. (2014a) and conjugate adsorbents was the most frequently cited paper with 294 citations. The article compares inorganic adsorbents of mesoporous silica and diphenyl ether-18 crown ether immobilized mesoporous silica as

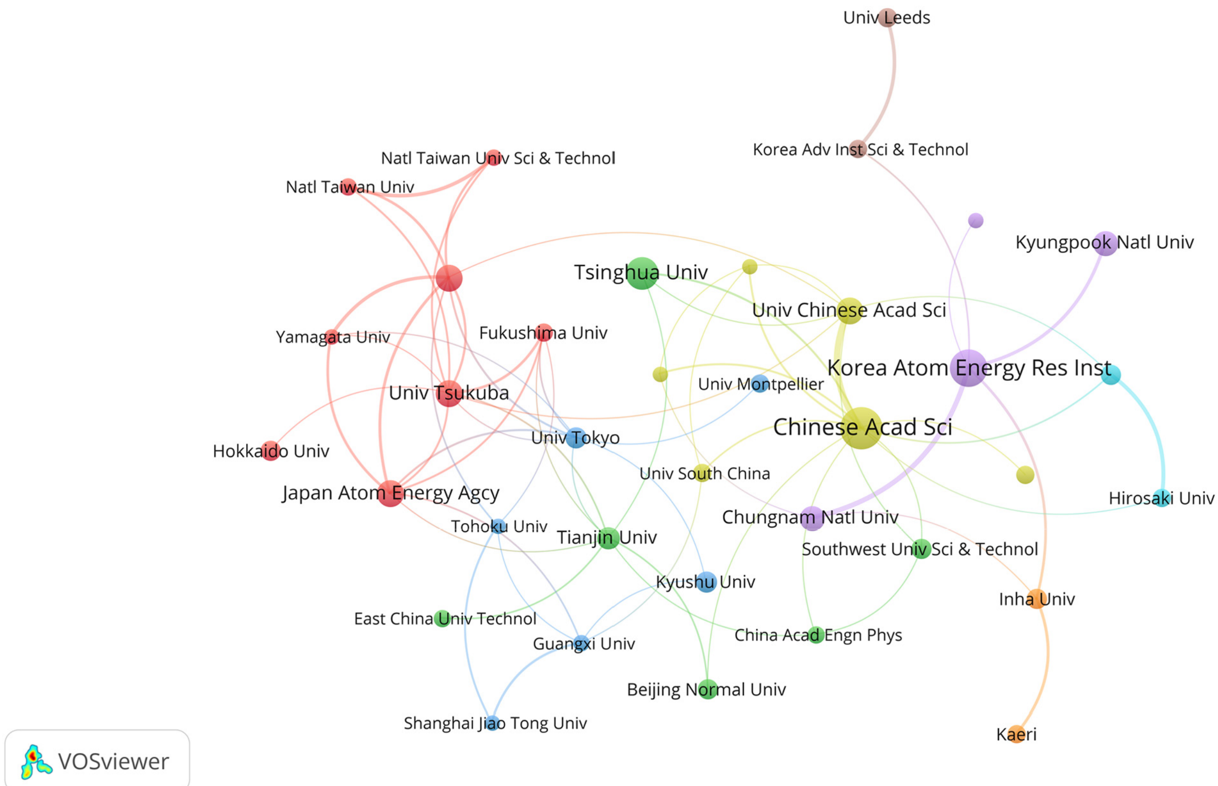


Fig. 4. Collaboration network between research institutions on Cs removal research.

coexisting adsorbents and finds that both adsorbents can remove Cs^+ from water under neutral conditions with a maximum adsorption capacity of 27.40 and 50.23 $mg{\cdot}g^{-1}$, independent of other coexisting ions. The publications with the second to fifth most citations were the *Journal of Hazardous Materials* (3784), *Journal of Radioanalytical and Nuclear Chemistry* (1389), *Environmental Science & Technology* (939), and *Chemosphere* (908). The most cited article published by *Journal of Hazardous Materials* is selective cesium removal from radioactive liquid waste by crown ether immobilized

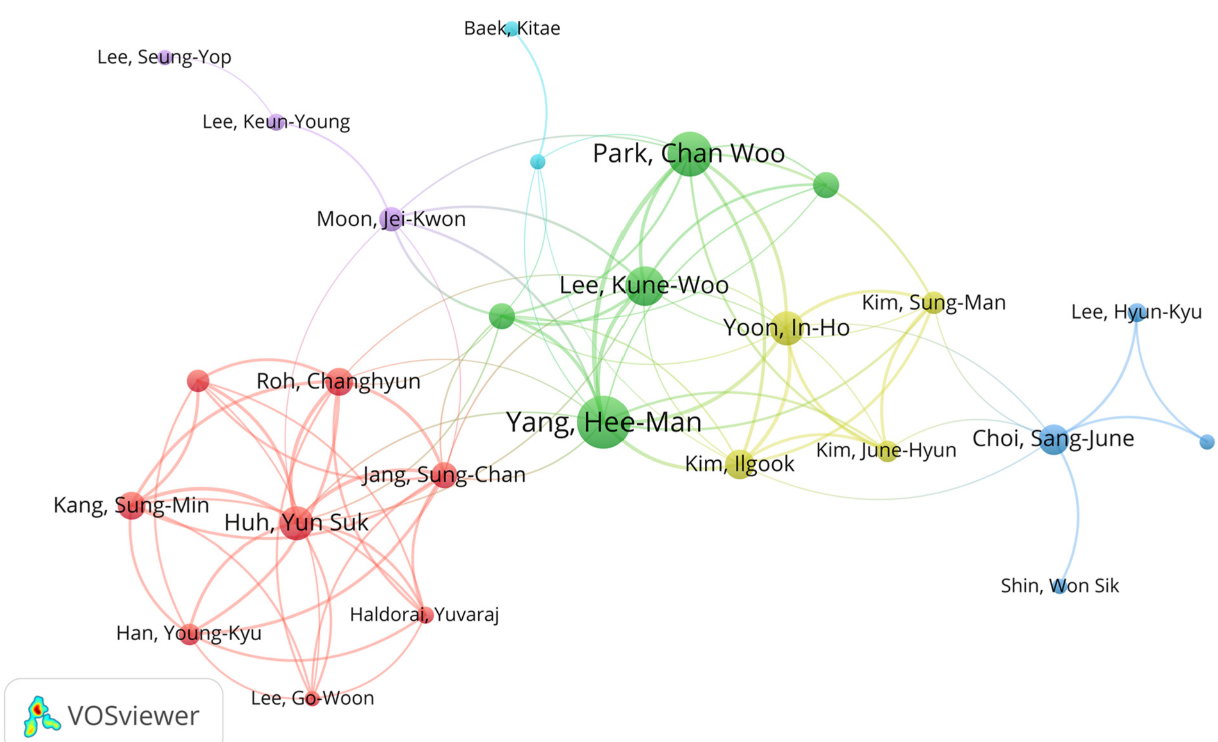


Fig. 5. Collaboration network between productive authors in the Cs removal research field.

Table 1
Ranking of publications in the field of Cs removal from water research by citation frequency, 2012–2022.

Rank	Journal	Country	Impact factor (2021)	No. of citations	No. of articles	Citation per publication
1	Chemical Engineering Journal	Switzerland	16.744	4660	102	46
2	Journal of Hazardous Materials	Netherlands	14.224	3784	108	35
3	Journal of Radioanalytical and Nuclear Chemistry	Netherlands	1.754	1389	130	11
4	Environmental Science & Technology	USA	11.357	939	15	63
5	Chemosphere	England	8.943	908	54	17
6	Journal of Environmental Radioactivity	England	2.655	801	46	17
7	Scientific Reports	Germany	4.997	770	26	30
8	RSC Advances	England	4.036	737	27	27
9	Separation and Purification Technology	Netherlands	9.136	652	30	22
10	Science of the Total Environment	Netherlands	10.753	614	32	19
11	Water Research	England	13.400	552	13	42
12	Journal of Environmental Chemical Engineering	England	7.968	517	32	16
13	Bioresource Technology	England	11.889	467	8	58
14	Chemistry of Materials	USA	10.508	449	6	75
15	Journal of Colloid and Interface Science	San Diego	9.965	437	9	49

new class conjugate adsorbent, with 266 citations. The preparation of the adsorbent is based on the direct immobilization of diphenyl ether-24-crown ether (DB24C8) on mesoporous silica, which can adapt to neutral conditions of pH and can reach an adsorption capacity of 77.70 $mg \cdot g^{-1}$, which can be recycled several times (Awual et al., 2014b). Among the articles published in *Journal of Radioanalytical and Nuclear Chemistry*, adsorption of uranium, cesium and strontium onto coconut shell activated carbon has the highest citation frequency with 103 citations (Caccin et al., 2012). The article published in *Environmental Science & Technology* with highly efficient enrichment of radionuclides on graphene oxide supported polyaniline has 519 citations. Sun et al. synthesized graphene oxide-supported polyaniline (PANI@GO) composites by chemical oxidation with an adsorption capacity of 1.39 $mmol \cdot g^{-1}$ at pH = 3.0 (Sun et al., 2013). Among the journals published in *Chemosphere*, highly efficient U(VI) capture by amidoxime/carbon nitride composites: Evidence of EXAFS and modeling has the highest citation frequency of 96 (Hu et al., 2021). These findings may assist researchers in identifying the most prominent publications in this research field.

4. Knowledge bases, research hotspots and frontiers

4.1. Knowledge bases of Cs removal (document co-citation analysis)

The retrieved papers to construct the knowledge base of Cs removal were analyzed. To evaluate the contribution of publications in this research field, we adopted cocitation analysis, which involves considering the frequency at which two files are simultaneously referenced by another file. **Fig. S1** presents the density mapping of the files reviewed in this study. Each node represents a cited document, and the number of citations determines the node color. A bright yellow color suggests the paper has more citations. Only 45 of the 43,329 publications met the requirement due to the minimum citation value of 50. The 15 most cited papers are listed in Table 2; most of these publications focused on composite materials for aqueous Cs removal. In Cs removal, Cs contaminants are removed through adsorption, and the composite materials are modified with common adsorbents, such as silica, clay, and biochar.

Table 2
Fifteen most cited references in the Cs removal research field in 2012–2022.

No.	Title	Authors	Source	Co-citations
1	Selective capture of cesium and thallium from natural waters and simulated wastes with copper ferrocyanide functionalized mesoporous silica	Sangvanich et al. (2010b)	Journal of Hazardous Materials	157
2	Radioactive cesium removal from nuclear wastewater by novel inorganic and conjugate adsorbents	Awual et al. (2014a)	Chemical Engineering Journal	136
3	Removal of cobalt, strontium and cesium from radioactive laundry wastewater by ammonium molybdophosphate–polyacrylonitrile (AMP–PAN)	Park et al. (2010)	Chemical Engineering Journal	133
4	Efficient removal of cesium from low-level radioactive liquid waste using natural and impregnated zeolite minerals	Borai et al. (2009)	Journal of Hazardous Materials	112
5	Evaluation of zeolite A for the sorptive removal of Cs^+ and Sr^{2+} ions from aqueous solutions using batch and fixed bed column operations	El-Kamash (2008)	Journal of Hazardous Materials	108
6	Selective cesium removal from radioactive liquid waste by crown ether immobilized new class conjugate adsorbent	Ho and McKay (1999)	Journal of Hazardous Materials	104
7	Pseudo-second order model for sorption processes	Awual et al. (2014b)	Process Biochemistry	102
8	Adsorption of cesium on copper hexacyanoferrate–PAN composite ion exchanger from aqueous solution	Nilchi et al. (2011)	Chemical Engineering Journal	99
9	Prussian blue caged in alginate/calcium beads as adsorbents for removal of cesium ions from contaminated water	Vipin et al. (2013)	Journal of Hazardous Materials	92
10	In situ controllable synthesis of magnetic Prussian blue/graphene oxide nanocomposites for removal of radioactive cesium in water	Yang et al. (2014b)	Journal of Materials Chemistry	91
11	Adsorption of cesium from aqueous solution using agricultural residue – Walnut shell: Equilibrium, kinetic and thermodynamic modeling studies	Ding et al. (2013)	Water Research	89
12	Adsorption removal of cesium from drinking waters: A mini review on use of biosorbents and other adsorbents	Liu et al. (2014)	Bioresource Technology	87
13	Extraction of radioactive cesium using innovative functionalized porous materials	Delchet et al. (2012)	Rsc Advances	80
14	Magnetic Prussian blue/graphene oxide nanocomposites caged in calcium alginate microbeads for elimination of cesium ions from water and soil	Yang et al. (2014a)	Chemical Engineering Journal	77
15	Proton-exchange mechanism of specific Cs^+ adsorption via lattice defect sites of Prussian blue filled with coordination and crystallization water molecules	Ishizaki et al. (2013)	Dalton Transactions	75

A high number of citations generally suggests that a paper has high academic value and professional influence. The total link strength of an article is the citation strength of the article. The most cited paper in this field is "Selective capture of cesium and thallium from natural waters and simulated wastes with copper ferrocyanide functionalized mesoporous silica" by Sangvanich et al. (2010b) in the *Journal of Hazardous Materials*. The authors of the paper synthesized copper ferrocyanide functionalized mesoporous silica and explored its application in removing Cs and thallium from both natural water and simulated wastewater. During the target period, the paper was cited 169 times with 44 links; the total link strength was 964, which was the highest of all the papers included in this review. This high number of citations highlights the importance of this paper in the field of aqueous Cs removal. Another highly cited article is "Radioactive cesium removal from nuclear wastewater by novel inorganic and conjugate adsorbents" (Awual et al., 2014a), which was published in the *Chemical Engineering Journal*. During the target period, the article had 153 cocitations, 44 links, and a total link strength of 763. The authors of the paper explored preparation methods for conjugated adsorbents and investigated their stabilities and selectivities. The low-cost conjugate adsorbent proposed in their paper exhibited substantial potential for treating the wastewater that was leaking from the Fukushima nuclear power plant and provided insights relevant to future adsorbent design. The article "Removal of cobalt strontium and cesium from radioactive laundry wastewater by ammonium hydroxide-acrylonitrile (AMP-PAN)" and published in the *Journal of Chemical Engineering* was the third most cited article; the authors analyzed the physical adsorption of a material through weak van der Waals forces to achieve ion exchange and removal. The paper had 143 citations, 44 links, and a total link strength of 580 (Park et al., 2010).

4.2. Research hotspots

Article keywords generally represent the research hotspots in a research field. To reduce the number of misclassifications in the present study and obtain a more precise visualization, we set the minimum value of the keyword event to 10. We performed a network analysis of keywords related to the removal of Cs using VOSviewer; only one keyword with a similar meaning was taken; for example, we considered singular and plural forms, abbreviated and full forms, and British and American spellings. In this study, 69 of the 3592 retrieved keywords met the requirement; we subsequently conducted a cluster analysis of the keywords (Fig. 6). The keywords were classified into five clusters (presented in yellow, blue, red, purple, and green); Table 3 lists the keywords for each cluster and the frequency of their co-occurrences. Different colors represent the relevance of keywords, the less the keywords extend out of the line, representing the less relevance to other keywords, the same color keyword relevance is strong, different color keyword relevance is weak.

4.2.1. Adsorption mechanism (Cluster 1 in yellow)

The focus of the keywords in Cluster 1 (in yellow) is mechanistic analyses of adsorption in the field of Cs removal, including kinetics, thermodynamics, and selectivity. Adsorption-based techniques have attracted considerable attention because of their straightforward operations and low costs. Kinetic and thermodynamic analyses are vital methods and have been widely used for investigating adsorption behavior. Researchers have explored and applied various models to identify the underlying mechanisms of the adsorption process. For example, the adsorption process can be clarified by considering adsorption isotherms, which be used to determine the relationship between the adsorption capacity and solution concentration at equilibrium reaction (Chen et al., 2013; Lee et al., 2016; Yakout and Hassan, 2014). Moreover, adsorption kinetic models can reveal the mechanisms underlying adsorption mass transfer. Although many cutting-edge techniques (e.g., computational quantum mechanical modeling and synchrotron radiation-based techniques) have demonstrated strong potential for clarifying pollutant adsorption mechanisms, adsorption kinetic and thermodynamic analyses have been the key research topics in the field of Cs removal. The extensive use of traditional mathematical formulas to determine the adsorption mechanisms involved in aqueous Cs

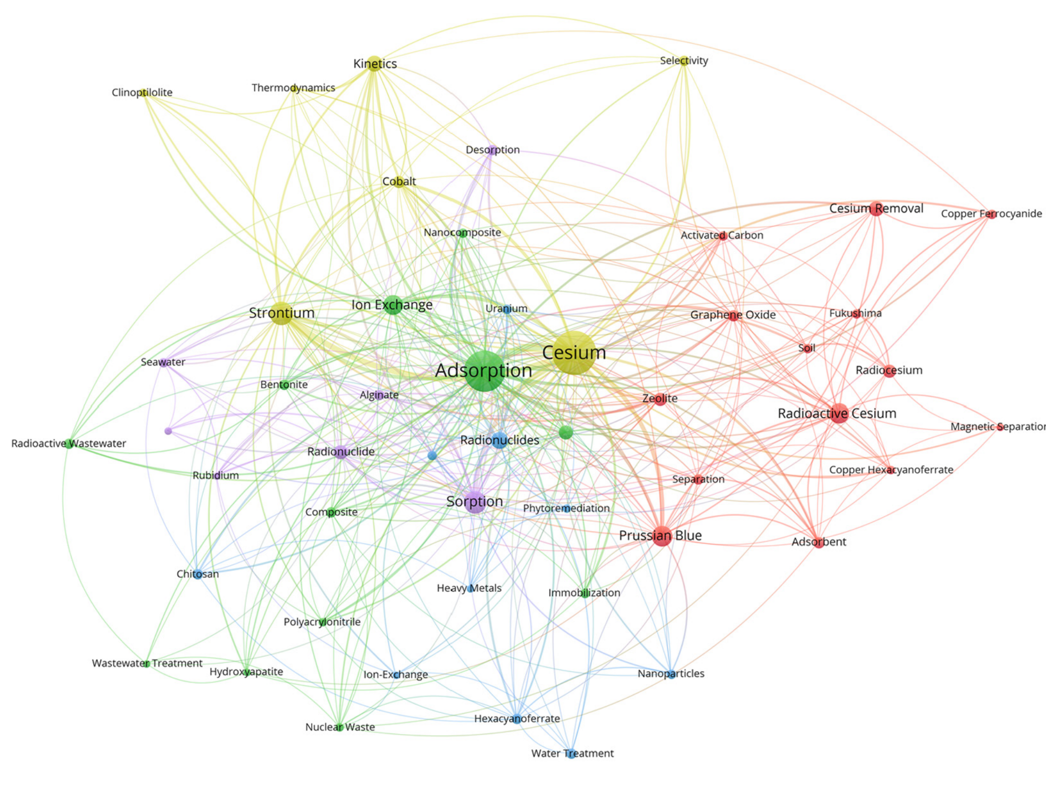


Fig. 6. Co-occurrence network of author keywords in the Cs removal research field.

Table 3
Co-occurrence of author keywords in Cs removal studies.

Cluster 1 (yellow)	Cluster 2 (blue)	Cluster 3 (red)	Cluster 4 (purple)	Cluster 5 (green)
Clinoptilolite (295)	Radionuclides (49)	Prussian Blue (79)	Sorption (80)	Adsorption (289)
Strontium (87)	Chitosan (26)	Radioactive Cesium (62)	Radionuclide (37)	Ion Exchange (62)
Kinetics (47)	Hexacyanoferrate (23)	Cesium Removal (37)	Alginate (29)	Radioactive Waste (34)
Cobalt (28)	Water Treatment (23)	Zeolite (30)	Desorption (26)	Composite (27)
Selectivity (31)	Nanoparticles (19)	Radioactive cesium (26)	Seawater (21)	Radioactive Wastewater (24)
Cesium (19)	Uranium (17)	Adsorbent (24)	Liquid Radioactive Waste (15)	Bentonite (20)
Thermodynamics (15)	Biosorption (16)	Graphene Oxide (23)	Rubidium (11)	Immobilization (17)
	Phytoremediation (12)	Separation (17)		Nanocomposite (14)
	Heavy Metals (11)	Activated Carbon (16)		Nuclear Waste (13)
	Ion-Exchange (10)	Fukushima (16)		Polyacrylonitrile (11)
		Copper Ferrocyanide (15)		Hydroxyapatite (11)
		Soil (13)		Wastewater Treatment (10)
		Copper Hexacyanoferrate (12)		
		Magnetic Separation (10)		

Note: Numbers in parentheses represent the number of times a keyword appears.

removal indicates that kinetics and thermodynamics are vital for understanding the interaction between adsorbents and Cs contaminants.

4.2.2. Treatment methods (Cluster 2 in blue)

The focus of the keywords in Cluster 2 (in blue) is treatment methods in the field of Cs removal, which mainly involve adsorption, ion exchange, biosorption, and phytoremediation. Among these methods, ion exchange was a research hotspot for aqueous Cs removal during the target period. Compared with precipitation and extraction, ion exchange is highly selective, radiation resistant, thermal stable and easily performed in removing aqueous Cs contaminants. Although the ion exchange method has many advantages, but the current general stability of the ion exchanger alone is not strong, how to stabilize the adsorption is the focus of current research. Although ion exchange is limited by the exchange capacity, current research has been conducted to optimize the design of ion exchanger and applied to effective Cs removal. Ion exchange is a key method for removing radioactive contaminants from water, and the selection of a suitable ionic exchange reagent for capturing nuclides is vital in the treatment process. Studies have explored various ion exchangers for removing nuclides from both simulated and real wastewater. Transition metal hexacyanoferrate, one type of ion exchanger, has demonstrated considerable potential for treating radioactive contaminants. Moreover, Lehto et al. (2019) evaluated the performance of common exchangers (zeolite, silicotitanate, hexacyanoferrate, and sodium titanate) for treating waste effluents generated during the cooling of damaged nuclear power plant reactors; their analyses revealed the outstanding efficacy of hexacyanoferrate for treating aqueous Cs contaminants. Other ion exchange agents functionalized with hexacyanoferrates also exhibited superior performance in treating wastewater with nuclide contaminants. For example, both polyacrylonitrile–potassium cobalt hexacyanoferrate and polyacrylonitrile–potassium nickel hexacyanoferrate demonstrated effective Cs adsorption; they can thus serve as efficient and inexpensive adsorbents (Du et al., 2014). The loading of potassium zinc hexacyanoate on the ferrite improves the thermal stability of the ferrite; this composite material has a uniform geometry and a clear spherical structure, and has a high Cs removal efficiency, showing excellent removal effects in the pH = 2–10 range; the maximum adsorption capacity of cesium by the material is 1965 mg·g^{-1} (Sheha, 2012). These findings indicate that ionic exchange remains the dominant method for nuclide removal from water, and ferricyanide-based materials with excellent ion exchange capacities have potential that merits further investigation.

4.2.3. Adsorbent design and optimization (Cluster 3 in red)

The focus of the keywords in Cluster 3 (in red) is the exploration of novel and effective adsorbents for Cs removal. The widespread exploration of advanced functional materials for nuclide pollution control is driven by increasingly severe environmental problems. Researchers have extensively explored and designed various adsorbents for Cs removal; our clustering analysis revealed that the keywords *Prussian blue* (PB) and *zeolite* had 79 and 30 occurrences, respectively, during the target period. PB has been widely investigated for Cs removal because of its unique structure and high selectivity. As coordination network materials, the adsorption behaviors of PB and PB analogue (PBA) are similar to that of zeolite (Attallah et al., 2018; Ding et al., 2013; Jang and Lee, 2016; Jia et al., 2017). The presence of some defects in the cubic lattice of PB and PBA results in them having open structures (Li et al., 2019), and these vacancies enable PB and PBA to adsorb and capture target ions or molecules in the lattice. PB nanoparticles with various hybrid structures have exhibited distinct adsorption performances. Table 4 summarizes the adsorption properties of some magnetic PB nanoparticles with different hybrid structures.

Although PB has demonstrated substantial potential for treating aqueous Cs contamination, the practical application of PB is limited by its low mechanical strength. Various methods have been explored for adsorbent design and optimization. For example, Hu et al. (2012) loaded PB into a diatomite cavity using in situ synthesis. Highly dispersed carbon nanotubes were then used to form a network of carbon nanotubes covering the diatomite to seal the PB particles. The combination of PB with other supports can increase the mechanical strength and separation performance of the adsorbent. PB can be loaded on the surface of granular activated carbon, and covalent organic polymer was then grafted onto the surface of the material to improve its stability as well as to reduce the desorption of PB (Seo and Hwang, 2021). Subsequently, the PB was fixed on a fiber filter, and the fixed strength was enhanced by adding acrylic acid. Because of this

Table 4
Adsorption properties of magnetic PB nanoparticles with different hybrid structures.

Adsorbent	Synthesis	Adsorption capacity/(mg·g^{-1})	Removal efficiency/%	Equilibrium time	Reference
Nanoclusters	Single precursor	45.87	>99.7	6 h	Yang et al. (2016)
Nanocomposites with graphene oxide	Anchoring the magnetic PB onto the graphene surface	55.56	>90.0	12 h	Yang et al. (2014b)
Nanoparticles	Single precursor	96.00	NA	24 h	Thammawong et al. (2013)
Nanocomposites	Co-precipitation	280.82	NA	24 h	Jang and Lee (2016)
Nanoparticles with PEG	Hydrothermal	274.70	64.8	1 h	Qian et al. (2017)
Microparticles	Hydrothermal	16.30	97.0	10 min	Wang et al. (2020)

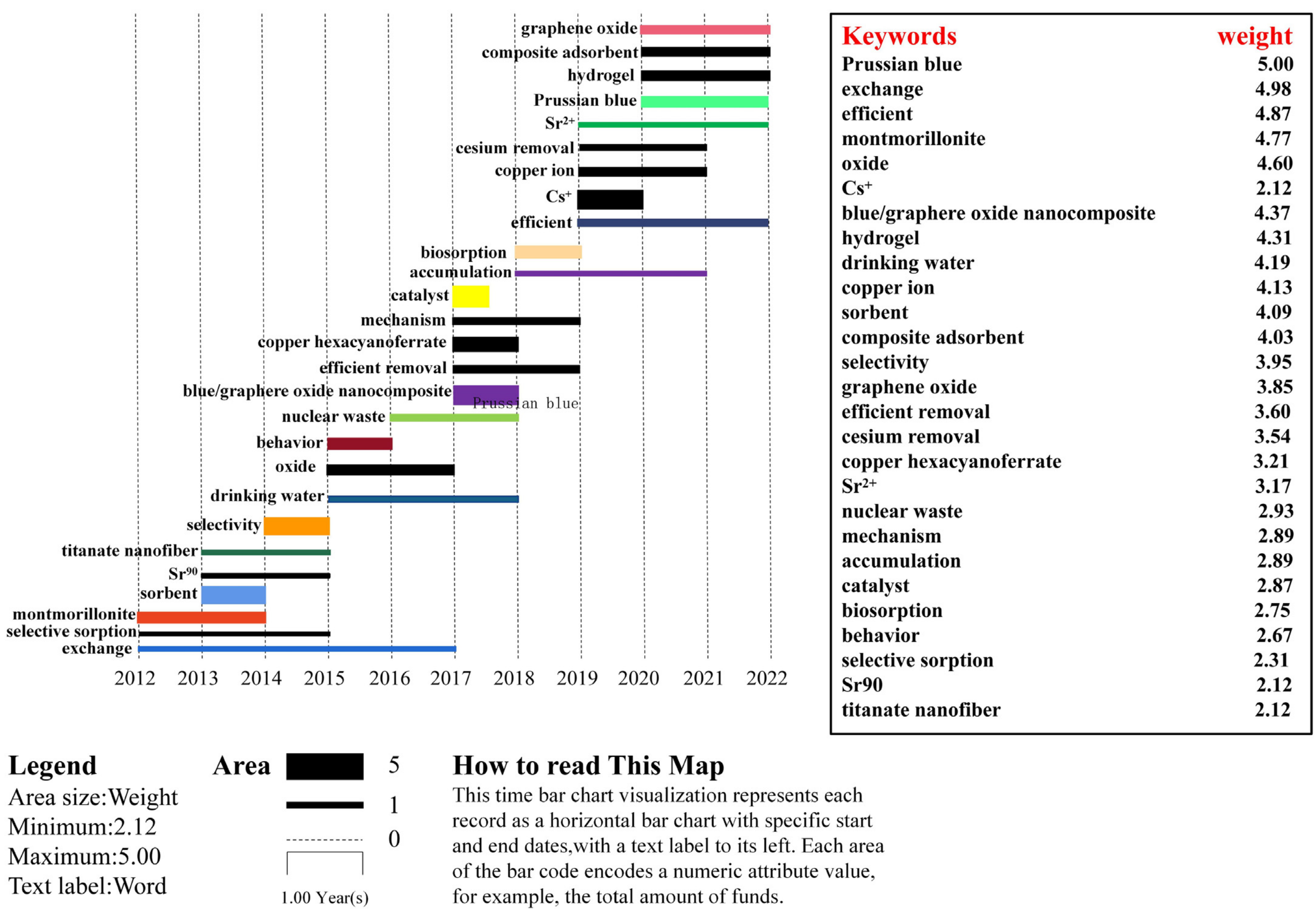
material's high immobilization strength, it can reduce exfoliation in the washing process and has a high reuse rate (Kim et al., 2019). These findings indicate the optimization of adsorbents is a key topic in research on Cs removal, and the exploration of functional materials was a main research hotspot during the target period.

4.2.4. Desorption and reuse (Cluster 4 in purple)

The focus of the keywords in Cluster 4 (in purple) is the desorption and reuse of adsorbents in the field of Cs removal. In addition to improving adsorption capacity, treating spent adsorbents is a challenge for researchers. Reliable treatment of spent adsorbents is crucial for sustaining the Cs removal process and reducing the amount of secondary pollution. Many researchers have investigated the behavior of spent adsorbents in the desorption–adsorption process to determine their cycling performance. Common desorbents for Cs^+ are solvents such as HCl and NH_4Cl. By placing the adsorbed material in a solution containing HCl or NH_4Cl and following this with homogeneous mixing, the Cs species in the material can be replaced and thus recycled. The desorption rate of transition metal–modified adobe clay can exceed 80 % when HCl is used in the desorption process (Ding et al., 2014). HCl can generally be used as a desorbent, provided that the adsorption of Cs^+ by the material is pH-dependent and the adsorption effect under acidic conditions is poor (Gasser et al., 2018). When the adsorption behavior of Cs^+ is less affected by pH and the material is applicable in a wider pH range, NH_4Cl can be used for desorption; NH_4^+ displaces Cs^+ in the desorption process (Jia et al., 2017). This is evidenced by the composite material ammonium molybdate, which can be desorbed using NH_4Cl with an efficiency higher than 90 % (Ding et al., 2017).

4.2.5. Composite materials (Cluster 5 in green)

The focus of the keywords in Cluster 5 (in green) is the exploration of composite materials for aqueous Cs removal. Composite materials have been widely investigated in the field of Cs removal because of their high adsorption capacity, stability, and straightforward separation process. Our clustering analysis revealed that *composites* was a high-frequency keyword, highlighting the potential of composites as promising adsorbents for Cs removal. To overcome the limitations of the original material (e.g., low adsorption capacity, low selectivity, and difficult separation), researchers have adopted combination strategies to prepare composite materials. For example, Jung et al. (2021) anchored PB nanocrystals on the surfaces of nanoscale porous polyacrylamide spheres to induce high selectivity toward Cs^+ in the composite. Seema (2020) prepared a magnetically self-assembled PB/reduced graphene oxide aerogel; the spent adsorbent could be easily separated using this aerogel and an applied magnetic field. Kim et al. (2021) deposited cyanide potassium copper ferrate on a nanofiber matrix using electrospinning; subsequently, the material exhibited improved adsorption capacity and efficient removal of Cs from seawater. In addition to the optimization of adsorption capacity, selectivity, and separation, the use of composite materials may reduce the toxic effects of nanoadsorbents. In both the scientific and industrial communities, substantial effort has been devoted to identifying optimal loading methods or strategies to increase the controllability of various nanomaterials. Therefore, immobilizing nano adsorbents in a suitable matrix is essential not only to prevent the leaching of the nanoparticles but also to improve their adsorption capacity and separation performance, this topic merits further research. In addition to composite materials, layered materials of pure



Keywords	weight
Prussian blue	5.00
exchange	4.98
efficient	4.87
montmorillonite	4.77
oxide	4.60
Cs^+	2.12
blue/graphere oxide nanocomposite	4.37
hydrogel	4.31
drinking water	4.19
copper ion	4.13
sorbent	4.09
composite adsorbent	4.03
selectivity	3.95
graphene oxide	3.85
efficient removal	3.60
cesium removal	3.54
copper hexacyanoferrate	3.21
Sr^{2+}	3.17
nuclear waste	2.93
mechanism	2.89
accumulation	2.89
catalyst	2.87
biosorption	2.75
behavior	2.67
selective sorption	2.31
Sr90	2.12
titanate nanofiber	2.12

Fig. 7. Burst detection of original keywords in the Cs removal research field in 2012–2022. (Note: Bars of different color indicate burst keywords starting from different years.)

components are also good sorbents for Cs removal. Previous studies have shown the effectiveness of layered ammonium vanadate and MXenes for removing aqueous Cs contaminants (Hwang et al., 2020; Zhang et al., 2022).

4.3. Research frontiers

Burst detection of original author keywords, which can be used to evaluate changes in keyword frequency, is often used to analyze changes in the research directions of a given field. Burst detection analysis enables researchers to obtain the burst weight of each keyword in the research field as well as the burst duration. We used CiteSpace software to extract the 27 keywords with the strongest burst weights in the field of aqueous Cs removal between 2012 and 2022 (Chen, 2006; Chen, 2017). In Fig. 7, each horizontal bar represents a burst keyword, the length of the horizontal bars represents the durations of the burst, and the areas of the horizontal bars represent the burst weights of the keywords. 27 burst keywords were identified during the target period, and their average duration was 2.56 years. The keywords were sorted according to their weights from highest to lowest, and the plots were drawn in chronological order. In 2015, the literature shifted from focusing on mineral sorbents and adsorption selectivity to nuclear waste and composite materials, which continued from 2015 to 2018. Starting from 2018, researchers focused on studying the accumulation effect of Cs contamination and evaluating the effect of biosorption; this shift suggests that the threat of Cs contaminants to human health is increasing and that this area requires urgent attention. From 2020 to 2022, scholars devoted considerable attention to emerging materials for treating Cs contamination. Some advanced functional materials, such as PB, composite adsorbents, hydrogel, and graphene oxide, emerged and attracted substantial attention during the target period. The immobilization of cellulose-based hydrogels was shown to promote the dispersion of KCuHCF nanoparticles, demonstrating the unprecedented adsorption capacity of the composites, with a removal efficiency of >90 % for Cs^+ (Kim et al., 2017). Graphene oxide was synthesized and applied to remove Cs^+ from water with an adsorption capacity of 95.46 $mg \cdot g^{-1}$ at pH = 6, which is a promising material for development (Xing et al., 2020). Our keyword analysis revealed that the increased focus on these emerging materials in the field of Cs removal can be attributed to their superior physiochemical properties. These findings suggest that developing advanced functional materials for effective nuclide removal is vital for driving advances in radioactive wastewater treatment.

5. Conclusions

In this study, the development of methods and techniques for Cs removal was analyzed on the basis of scientific bibliometric statistics. Future directions for research on functional sorbents for treating aqueous Cs contamination were discussed as well. On the basis of key information, such as the sources of research, countries, research institutions, authors' productivity, and keywords, a series of statistical maps was created to reveal the research hotpots and identify the research frontiers in the field of aqueous Cs removal.

(1) The contribution of Asian countries is significant in the selected time period, and the more frequent contact and cooperation between countries provides information for researchers to select potential collaborators.
(2) Adsorption is currently the most researched method in this field, and suitable adsorbent can make the removal efficiency high.
(3) PB is the most researched adsorbent at present, and the synthesis of composite adsorbent using adsorbent and support skeleton can improve the removal efficiency and recovery efficiency and save cost.
(4) Future research trends favor materials such as hydrogels and graphene oxide, which have been extensively studied as auxiliary materials for the design of effective and stable adsorbents for Cs removal.

These bibliometric analytical findings can assist researchers in making evidence-based decisions with respect to their research priorities and may offer researchers and institutions novel research strategies for investigating effective Cs removal. The future focus of research is still on composite materials, and the emergence of new materials like graphene oxide and hydrogels also provides promising alternatives for the development of this field.

CRediT authorship contribution statement

Heyao Liu: Methodology, Investigation, Writing – original draft, Formal analysis. **Lizhi Tong:** Writing – review & editing. **Minhua Su:** Conceptualization, Writing – review & editing, Supervision, Resources, Funding acquisition. **Diyun Chen:** Resources, Funding acquisition. **Gang Song:** Resources. **Ying Zhou:** Writing – review & editing.

Data availability

No data was used for the research described in the article.

Declaration of competing interest

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

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Appendix A. Supplementary data

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