

# A bibliometric review of research trends on bioelectrochemical systems

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**Bioelectrochemical systems (BES) have received widespread attention for their capability of simultaneous wastewater treatment and electricity or fuels/chemicals production. The present study was performed to evaluate the global scientific output of BES-related research based on the Science Citation Index-Expanded database from 1991 to 2014. The publication outputs, journals, subject categories, countries and institutes were analysed to identify the world research on BES. In addition, ‘word cluster analysis’ was applied to give an insight into the research trends on BES. The results indicate that the annual publications on BES increased steadily, especially after 2004. China, with 7 institutes included among the 15 most productive institutes, is the largest contributor on BES research during the past 24 years. Microbial fuel cell is the most frequently studied reactor, and ‘electricity generation’ and ‘wastewater treatment’ are two dominant applications of BES. The use of two nanostructured materials – carbon nanotubes and graphene – is increasingly studied in the BES field.**

**Keywords:** Bibliometric analysis, bioelectrochemical systems, electricity generation, wastewater.

BIOELECTROCHEMICAL systems (BES) are unique systems that convert chemical energy into electricity or fuels/chemicals through the catalytic activities of microorganisms or enzymes. Based on the biocatalysts and their application mode, BES can be classified into enzymatic fuel cell (EFC), microbial fuel cell (MFC), microbial electrolysis cell (MEC), microbial desalination cell (MDC) and microbial solar cell (MSC). The historical origin of BES research can be dated back to 1911, when Potter<sup>1</sup> first discovered that certain bacteria can transfer electrons extracellularly while decomposing organic compounds. However, no significant discovery was made in the next few decades. In 1980s, when the wide application of electron transfer mediators had considerably improved the power outputs, BES got more attention from researchers<sup>2</sup>. The mediators used in BES are mostly toxic, expensive and easy to be washed away, which has heavily

hindered their industrial utilization. Therefore, it was only after 2002 that researches on BES achieved remarkable improvements due to the discovery in 1999 by Kim *et al.*<sup>3,4</sup> of a mediator-less MFC using an Fe(III)-reducing bacterium – *Shewanella putrefaciens*. In recent years, many studies have been carried out on the electron transfer mechanisms<sup>5–7</sup> and methods to enhance the performance of BES<sup>8,9</sup>. Moreover, several review articles have been published on the methodology<sup>10</sup>, substrates<sup>11</sup>, recent development<sup>12–14</sup>, challenges<sup>15</sup> and future applications<sup>16</sup> of BES reactors. However, a comprehensive statistical review of the global BES-related research has not been reported to date.

Bibliometrics, first introduced by Pritchard<sup>17</sup>, is an effective method that uses quantitative analysis and statistics to describe the research trend of a specific field. The bibliometric method has been broadly applied to analyse the scientific production and research trends in numerous fields, such as nitrate removal<sup>18</sup>, solid waste<sup>19</sup>, desalination<sup>20</sup> and materials science<sup>21</sup>. In the present study, a bibliometric analysis has been performed on BES-related research from 1991 to 2014. Basic publication items, including publication outputs, subject categories, journals, countries and institutes were identified to describe the status of BES research. Moreover, ‘word cluster analysis’ was applied to evaluate the research trend during 1991–2014. These results could provide a basis for better understanding of the development of the global BES research.

## Data sources and methods

Documents used in this study were obtained from the on-line version of SCI-Expanded (SCIE), Thomson Reuters’ *Web of Science* database. In the 2014 edition of the *Journal Citation Reports (JCR)*, 8618 journals with citation references across 176 scientific disciplines have been listed. Due to the absence of abstract and keywords in articles before 1991, only articles published in and after 1991 have been discussed in this study. Articles related to BES were queried based on the topic (‘bioelectrochemical system\*’, or ‘bio-electrochemical system\*’, ‘bioelectrochemical reactor\*’ or ‘bio-electrochemical reactor\*’, ‘microbial fuel cell\*’, ‘microbial desalination

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cell\*', 'microbial electro dialysis cell\*', 'microbial electrochemical cell\*', 'microbial thermoelectric cell\*', 'microbial biofuel cell\*', 'microbial photoelectrochemical cell\*', 'microbial electrolysis' or 'enzymatic biofuel cell\*', 'enzymatic fuel cell\*', 'microbial solar cell\*', 'microbial electrochemical snorkel' or 'microbial electrosynthesis'). Articles originating from England, Scotland, Wales and Northern Ireland were reclassified as from the United Kingdom (UK), and publications from Hong Kong were combined with those from mainland China. The reported Impact Factor (IF) of each journal was obtained according to *JCR* 2014.

Contributions of different institutes and countries were estimated by the affiliation of at least one author of the articles. Collaboration pattern was determined by the addresses of the authors, where the term 'single country/institute' was assigned if all the authors' addresses were from the same country/institute and the term 'international/inter-institutional collaboration' was assigned if the articles were co-authored by researchers from different countries/institutes.

All articles related to research on BES during the past 24 years (1991–2014) were assessed from the following aspects: publication outputs, subject categories and journals, countries and institutes. To obtain the research trend information, 'word cluster analysis' which combined the words in title, author keywords and abstracts was applied.

## Results and discussion

### Publication output

Though the basic principle of BES was discovered about 100 years ago, it is only in recent years that the system has attracted widespread interest and achieved remarkable improvements. During 1991–2003, BES-related publications increased at a relatively low rate, from only one article published in 1991 to 15 articles in 2003, with a total of 53 articles published in SCIE database (Figure 1). Research on BES experienced tremendous growth over the past 11 years (2004–2014). The number of publications increased to 680 in 2014, which was 45.3 times that in 2003. A total of 3131 BES-related journal articles were published during 1991–2014 in SCIE database, and these articles were used for further analysis.

### Distribution of subject categories and journals

According to the classification of subject categories in the *JCR*, the 3131 articles are distributed in 50 subject categories. The subject categories containing over 500 BES-related articles are biotechnology and applied microbiology (1070), energy and fuels (955), chemistry (845), electrochemistry (805), engineering (767), and environmental sciences and ecology (563). The articles

belonging to these top six categories cover 91.9% of the total articles published during the past 24 years. Figure 2 shows the annual publication outputs of the top 6 subject categories. It can be observed that since 2005, BES-related studies in these six subject categories have experienced a rapid increase. This indicates that BES is an emerging multidisciplinary research area.

The 3131 BES-related articles were published in 386 journals in the SCIE database. Table 1 lists the top 10 most productive journals, with the respective IF, *h*-index and average number of citations per BES related article. A total of 1421 articles on BES were published in the 10 journals, accounting for 45.4% of the total articles published during the past 24 years. Among them, *Biore source Technology* published the most articles (428), comprising 13.7% of the total, followed by *Environmental Science and Technology* (5.9%), *Journal of Power Sources* (5.6%), *International Journal of Hydrogen Energy* (5.3%) and *Biosensors and Bioelectronics* (4.4%).

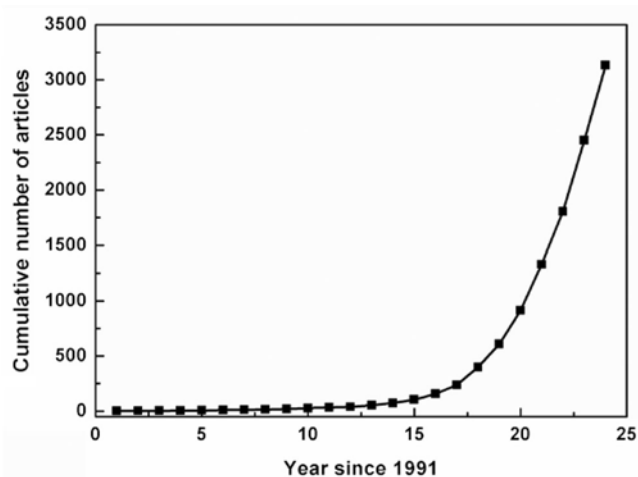


Figure 1. Cumulative number of articles by year from 1991 to 2014.

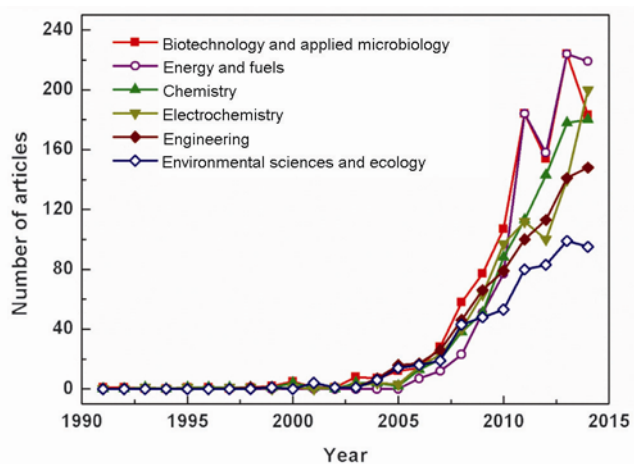


Figure 2. Growth trends of BES-related articles of the top 6 categories.

**Table 1.** Top 10 most productive journals on BES research

Journal	TP (%)	IF	<i>h</i> -index	TC/TP
<i>Bioresource Technology</i>	428 (13.7)	4.494	38	15.6
<i>Environmental Science and Technology</i>	184 (5.9)	5.330	62	75.4
<i>Journal of Power Sources</i>	175 (5.6)	6.217	33	21.8
<i>International Journal of Hydrogen Energy</i>	165 (5.3)	3.313	25	14.9
<i>Biosensors and Bioelectronics</i>	139 (4.4)	6.409	37	31.9
<i>Electrochimica Acta</i>	70 (2.2)	4.504	19	17.4
<i>Electrochemistry Communications</i>	68 (2.2)	4.847	28	38.8
<i>Energy and Environmental Science</i>	66 (2.1)	20.523	29	36.4
<i>Applied Microbiology and Biotechnology</i>	66 (2.1)	3.337	28	36.0
<i>Water Research</i>	60 (1.9)	5.528	27	46.9

TP, Total number of publications (the percentage of articles in total publications is given within brackets); IF, Impact factor, based on *JCR* 2014; TC, Total number of citations.

**Table 2.** Top 15 most productive countries/territories in BES research

Country	TP	R (%)	R (FP)	R (RP)	R (SP)	R (CP)
China	924	1 (29.54)	1 (816)	1 (799)	1 (653)	2 (271)
USA	922	2 (29.48)	2 (743)	2 (749)	2 (609)	1 (313)
South Korea	267	3 (8.54)	3 (204)	3 (209)	3 (160)	3 (107)
India	176	4 (5.63)	4 (158)	4 (157)	4 (139)	12 (37)
UK	175	5 (5.59)	5 (133)	5 (127)	6 (92)	4 (83)
Japan	152	6 (4.86)	6 (124)	6 (125)	5 (111)	9 (41)
Germany	94	7 (3.01)	10 (64)	10 (65)	11 (44)	7 (50)
France	90	8 (2.88)	7 (76)	7 (76)	7 (61)	17 (29)
Australia	90	8 (2.88)	14 (50)	14 (49)	13 (35)	5 (55)
Canada	87	10 (2.78)	10 (64)	11 (64)	9 (53)	14 (34)
The Netherlands	84	11 (2.69)	8 (68)	9 (66)	8 (54)	16 (30)
Spain	83	12 (2.65)	12 (60)	12 (59)	10 (48)	13 (35)
Taiwan	83	12 (2.65)	8 (68)	8 (72)	11 (44)	10 (39)
Belgium	75	14 (2.40)	15 (42)	15 (48)	17 (22)	6 (53)
Singapore	71	15 (2.27)	13 (52)	13 (56)	16 (25)	8 (46)

R, Rank (the percentage of articles in total publications is given within brackets); FP, First author publications; RP, Reprint author publications; SP, Single-national publications; CP, International collaborative articles.

According to Bradford's law of scattering<sup>20</sup>, the above-mentioned 5 journals, publishing about 34.8% of the total articles, are the core journals in BES field. *Energy and Environmental Science*, which has the highest IF (20.523) among the top 10 journals, has published 66 articles on BES. *Environmental Science and Technology*, with an IF of 5.330, has the highest *h*-index (62) and also the highest average citations per article (TC/TP, 75.4) in the BES field.

#### Publication distribution of countries and institutes

The contributions of countries/territories to BES-related research output were evaluated by the location of the affiliations of at least one author of the articles. Among the 3131 articles, 3 had no author address information. The remaining 3128 articles were published by 58 countries/territories. Among the 3128 articles, 2424 (77.5%) are single-country publications and the others (704,

22.5%) are internationally collaborative publications. Table 2 lists the top 15 countries/territories ranked by the total publications. It is evident that China and USA are the largest contributors, publishing 924 and 922 articles respectively, which is far more than those of other countries. China started BES research rather late. The first article published in SCIE database with a Chinese author appeared in 2006. However, the output of China increased at a rather high rate, and surpassed USA in 2012. China ranks first in the number of total publications, first author publications, reprint author publications and single-country publications, and second in international collaborative publications. Except for Germany, Australia, Belgium and Singapore, all the other 11 most productive countries/territories have more single-country publications than collaborative publications, and India has the highest percentage of independent articles (79%), followed by Japan (73%).

The 2128 articles on BES were resubmitted by 1311 different institutes, among which 699 (53.3%) institutes

**Table 3.** Top 15 most productive institutes in BES research

Institute	TP	R (%)	R (FP)	R (RP)	R (SP)	R (CP)
Penn State Univ., USA	222	1 (7.1)	1 (159)	1 (164)	1 (110)	2 (112)
Chinese Acad. Sci., China	158	2 (5.1)	3 (85)	3 (86)	2 (37)	1 (121)
Harbin Inst. Technol., China	135	3 (4.3)	2 (93)	2 (88)	5 (31)	3 (104)
Univ. Massachusetts, USA	61	4 (2.0)	4 (51)	5 (50)	3 (33)	20 (28)
Guangdong Inst. Ecoenvironm. and Soil Sci., China	60	5 (1.9)	6 (47)	6 (49)	22 (12)	5 (48)
Univ. Tokyo, Japan	59	6 (1.9)	14 (33)	20 (29)	65 (5)	4 (54)
Univ. Queensland, Australia	59	6 (1.9)	14 (33)	14 (33)	12 (17)	7 (42)
Wageningen Univ., The Netherlands	56	8 (1.8)	7 (45)	7 (43)	34 (9)	6 (47)
Univ. Sci. & Technol. China, China	56	8 (1.8)	4 (51)	4 (51)	7 (26)	17 (30)
Univ Ghent, Belgium	55	10 (1.8)	14 (33)	11 (36)	18 (15)	8 (40)
S. China Univ. Technol., China	53	11 (1.7)	8 (43)	7 (43)	8 (25)	20 (28)
Dalian Univ. Technol., China	52	12 (1.7)	10 (38)	12 (34)	14 (16)	10 (36)
Tsinghua Univ., China	51	13 (1.6)	9 (40)	9 (39)	14 (16)	11 (35)
Univ Wisconsin, USA	49	14 (1.6)	12 (35)	14 (33)	18 (15)	12 (34)
Arizona State Univ., USA	47	15 (1.5)	11 (37)	10 (37)	4 (32)	44 (15)

R, Rank (the percentage of articles in total publications is given within brackets); FP, First author publications; RP, Reprint author publications; SP, Single-national publications; CP, International collaborative articles.

published only one article and 196 (15%) published two articles. Table 3 provides a list of the top 15 most productive institutes during 1991–2014. Pennsylvania State University ranks first in total publications, first author publications, reprint author publications and single-institutional publications and second in inter-institutional publications, with a total of 222 articles, followed by the Chinese Academy of Science (158) and Harbin Institute of Technology (35). A bias might appear because the Chinese Academy of Science has branches in many cities, and a different ranking would be generated if articles are divided among the branches. It is noteworthy that 7 of the 15 institutes are from China, which is in accord with the rapid increase in publications from China.

Among the top 15 most productive institutes, only the University of Massachusetts and Arizona State University, both from USA, have more single-institutional publications than collaborative publications. Inter-institutional collaboration is more common in other institutes, especially for University of Tokyo, 91.5% of whose publications are collaborative articles, and only 5 single-institutional articles. The Wageningen University, the Netherlands also prefers inter-institutional collaboration, with only 9 single-institutional articles out of 56 total publications.

### Hot topics

The author keywords of an article can supply details of the article subjects and a statistical analysis of them can offer the research trend information of a particular field. Among the 3131 articles, 2428 with author keywords information were analysed. A total of 4479 keywords were listed by authors on the BES field, among which 3262 (72.8%) were used only once and 544 (12.1%) were used twice. When analysing the frequency of the author

keywords used in the past 24 years on the BES field, the synonymic single words and congeneric phrases were summed. For example, ‘microbial fuel cell’, ‘microbial fuel cells’, ‘microbial fuel cell (MFC)’, ‘microbial fuel cells (MFCs)’, ‘MFC’ and ‘MFCs’ were all counted as ‘microbial fuel cell’. ‘Microbial fuel cell’ (1465) is the most frequently used author keyword, followed by ‘microbial electrolysis cell’ (160), ‘wastewater treatment’ (131), ‘bioelectrochemical system’ (126), ‘biofuel cell’ (111), ‘electricity generation’ (110), ‘biocathode’ (98), ‘biofilm’ (97), ‘power density’ (90) and ‘bioelectricity’ (76). Based on the results of author keywords analysis, studies on BES can be separated into three categories: different types of BES reactors, applications and methods to improve performance.

In order to analyse the historical development of BES research, a new method called ‘word cluster analysis’ was applied in this study. This method has been used to study the research trend on drinking water<sup>22</sup>, proteomics<sup>23</sup>, nitrate removal<sup>18</sup> and atmospheric simulation<sup>24</sup>, and its procedure has been described in detail in the literature<sup>22</sup>. In the present study, article titles, author keywords and abstracts were combined to form the word base.

According to the results of author keywords analysis, MFC and MEC are the two most frequently studied BES reactors; other reactors, including EFC, MDC and MSC are less studied. Figure 3 shows the annual outputs on different types of reactors. It is evident that the annual publications on MFC, MEC and EFC show an increasing trend. MFC is the most frequently studied and also the earliest studied BES reactors. In 1962, Davis and Yarbrough<sup>25</sup> had reported on the preliminary experiments of MFC. The discovery of mediator-less MFC had given a significant boost to MFC research, which could be illustrated by the fact that the number of articles on MFC has doubled from the year 2007 (66) to 2008 (137). The construction materials and methods, substrates, current

progress and the challenges of MFC have been reviewed in detail<sup>10,11,13,15,26,27</sup>. MEC, which is also known as biocatalysed electrolysis cell or bioelectrochemically assisted microbial reactor<sup>28,29</sup>, is a modified MFC, whose cathode is anaerobic and thus a small external voltage is needed for hydrogen evolution<sup>30</sup>. MEC was independently discovered by two different research teams just a few years ago<sup>28,29</sup>, and publications on MEC have increased in recent years. MDC was first reported by Cao *et al.*<sup>31</sup> in 2009. The annual outputs on MDC have increased slowly in the past few years, and remained stable (16 articles) during 2012–2014. Though MDC can minimize power consumption in the desalination process, there are still some technical problems to be solved before practical application. It is reported that 200 ml of artificial wastewater (acetic acid as carbon source) is needed to desalinate only about 3 ml of salt water. Moreover, production of protons at one electrode and consumption at the other make these chambers more acidic and alkaline<sup>32</sup>. As a result, further studies are required for a better understanding of this technology and to improve its use in commercial applications. The principle and performance of MEC, EFC, MSC and MDC have also been reviewed in detail<sup>12,14,16,36</sup>.

BES is a new technology with great promise and varied applications. Figure 4 shows the annual publications on different applications of BES. It is no surprise that electricity generation is the main application of BES, as the fundamental purpose of BES is to recover electricity from organic matter to obtain electrical energy or produce other fuels/chemicals. Using MFC to recover energy from wastewater was first presented in 1991 (ref. 34). In 2004, Liu *et al.*<sup>35</sup> found that MFC can produce electricity from domestic wastewater and accomplish biological wastewater treatment simultaneously. This finding marked the establishment of a relationship between electricity generation and wastewater treatment. After that, simultaneous electricity generation and wastewater treatment in MFCs

was studied using many kinds of wastewater, including domestic wastewater<sup>36–38</sup>, swine wastewater<sup>39,40</sup>, food processing wastewater<sup>41</sup>, brewery wastewater<sup>42</sup>, etc. Moreover, wastewater treatment can also be achieved in other BES reactors, such as MECs<sup>43,44</sup> and MDC<sup>45</sup>.

As shown in Figure 4, the number of articles on denitrification and biosensors is much less than the other three applications and increases at a relatively slow rate. Complete cathodic denitrification to N<sub>2</sub> gas with a chemical anode was established in 1993. However, H<sub>2</sub> gas produced from electrolysis of water was used as the intermediate reducing agent and a high-energy input was required to electrolyze water. Gregory *et al.*<sup>46</sup> found that a pure culture of *Geobacter metallireducens* was able to reduce nitrate to nitrite with the electrode as the sole electron donor. In this process, hydrogen formation was not needed, but denitrification was not completed. It was in 2007 that a MFC with both a bioanode and biocathode performing simultaneous organics removal, power production and complete denitrification without relying on H<sub>2</sub>-formation or external energy input was first established<sup>47</sup>. In recent years, BES for nitrate removal has been utilized in wastewater treatment<sup>48</sup> and ground water *in situ* remediation<sup>49</sup>. MFC can be applied as a microbial biosensor for analysis of target chemicals, and continuous on-line process monitoring and control. Research on MFC as a biosensor started rather early. In 1977, Karube *et al.*<sup>50</sup> developed a MFC-based biochemical oxygen demand (BOD) biosensor with *Clostridium butyricum* immobilized on the electrode. Despite its early start, MFC-based biosensor developed slowly. There were only 7 articles published on MFC-based biosensors during 1991–2002. After 2002, more articles were published in this field, which might be due to the development of mediator-less MFC. MFC-based BOD biosensors have long-term stability, fine reproducibility and accuracy compared with other types of BOD sensors, and have

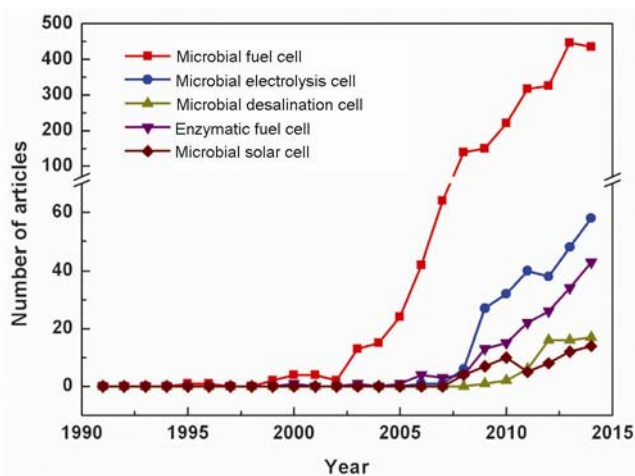


Figure 3. Research trends of different types of BES reactors.

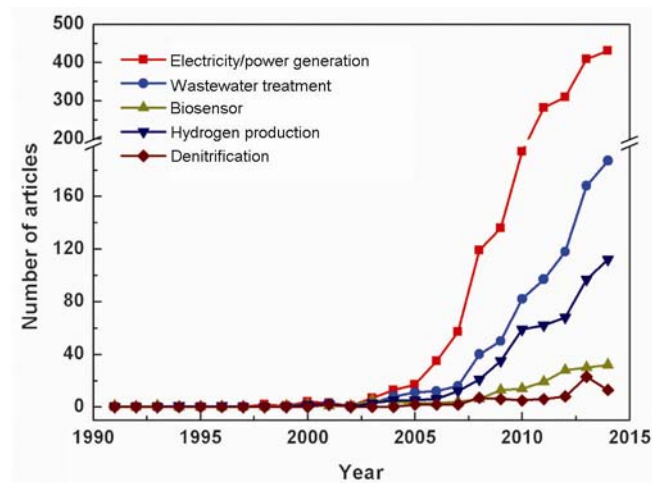


Figure 4. Research trends of different BES applications.

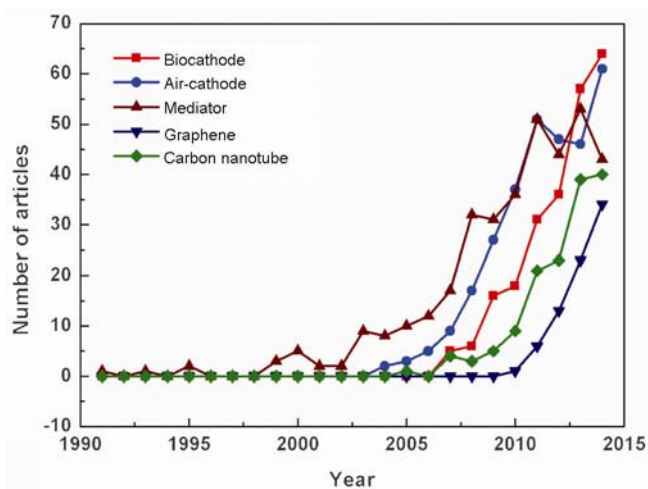
been studied by many researchers<sup>51,52</sup>. MFC-based biosensors can also be used to analyse other parameters, including lactate<sup>53</sup>, volatile fatty acids<sup>54</sup> and toxic substances<sup>55,56</sup>.

The threshold for feasible industrial applications of BES is considered to be  $1 \text{ kW m}^{-3}$  (ref. 57). During the past 24 years, many studies have concentrated on improving the performance of BES to make them commercially available. The effects of factors such as electrode materials, reactor design and configuration and operation conditions on the performance of BES have been described in detail<sup>58,59</sup>. Figure 5 shows several factors that might affect the cost and performance of BES reactors. BES reactors with air-cathode and biocathode can both reduce the installation and operation costs, and are increasingly studied by researchers. The electrode materials have significant impact on BES performance, and many materials, including carbon cloth, carbon felt, fibre brush, and graphite rod, have been used. In recent years, carbon nanotubes (CNT) have exhibited great potential in application in BES reactors, due to their high specific surface area and unique electrical and mechanical properties<sup>60</sup>. CNT can serve as electrode material or be used to modify the anode material, which can attract more exoelectrogens, forming thicker biofilms<sup>61</sup> and facilitate electron transfer from exoelectrogens to the anode by providing a strong interaction between the microbial biofilms<sup>62</sup>. Graphene has attracted tremendous interest due to its desirable properties for electrochemical applications: high conductivity, large specific surface area, applicable electrocatalytic activities and low production costs<sup>63,64</sup>. Liu *et al.*<sup>65</sup> were the first to use graphene for BES. In 2010, they found that graphene-based enzymatic fuel cell exhibited a power density nearly two times greater than that of the single-walled CNT cell. Studies on the application of graphene in BES have increased at a relatively high rate during the past 4 years. However, the number of articles

on mediators in BES has remained stable during the past 4 years. Mediators such as thionine, methyl viologen and humic acid facilitate electron transfer from microbial cells to the electrode. As most of the mediators are expensive and toxic, their application in commercialized MFC is limited. Researchers are focusing on mediator-less BES reactors<sup>66,67</sup> to find safer and cheaper mediators<sup>68</sup>.

## Conclusion

Based on 3131 BES-related publications from the SCIE database, this bibliometric study provided an overview of research in BES technology and identified some significant factors. The keyword analysis revealed that research on BES can be separated into three aspects: different types of BES reactors, applications and methods to improve BES performance. MFC is the most popular BES reactor, and utilization of BES for electricity generation and wastewater treatment is the most studied application. BES-related publications have significantly increased during the past 24 years and are predicted to be growing at a high rate in the future. Articles were published in 386 journals in 50 subject categories. *Bioresource Technology* published the most number of articles in this field, followed by *Environmental Science and Technology* and *Journal of Power Sources*. China, with 7 institutes included in the top 15 productive institutes, is the biggest contributor to BES publications.



**Figure 5.** Research trends of different methods to improve BES performance.

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ACKNOWLEDGEMENT. We thank the National Natural Science Foundation of China for financial support (Grant No. 51278050).

Received 13 July 2015; revised accepted 2 September 2015

doi: 10.18520/v109/i12/2204-2211