A bibliometric analysis of permeable reactive barrier enhanced electrokinetic treatment for sustainable polluted soil remediation

M A Budihardjo¹, R P Safitri¹, B S Ramadan¹, A J Effendi², S Hidayat², Y V Paramitadevi³, B Ratnawati³, A Karmilia¹

¹Environmental Engineering Department, Faculty of Engineering, Universitas Diponegoro, Indonesia
²Environmental Engineering Department, Faculty of Civil and Environmental Engineering, Institut Teknologi Bandung, Indonesia
³Environmental Management and Engineering Program, College of Vocational Studies of Bogor Agricultural University, Kumbang Street no 14, Bogor, West Java 16151, Indonesia

bimastyaji@live.undip.ac.id

Abstract. Research on soil remediation continues to develop, one of which is electrokinetic remediation combined with a permeable reactive barrier as a medium to prevent the migration of metals removed from the anode and cathode spaces. Thus, it is hoped that there is no need for reprocessing the residue resulting from electrokinetic remediation. This study aims to conduct a bibliographical analysis related to electrokinetic remediation coupled by permeable reactive barriers for heavy metal contaminated soil and to examine the effect of using various types of reactive barrier materials and their placement on the pollutants removal in the soil. Based on the results of bibliographic analysis, 26 relevant scientific articles were obtained, and the most publications in 2020 with 27% additional article publications are found. China and Environmental Science and Pollutant Research are the countries and journals that contribute the most to publications related to EK-PRB on heavy metal polluted soils.

1. Introduction

Rapid industrial development has a positive impact on human life and has a negative impact with losses that can even be greater and have a broad impact on the lives of other living things. These negative impacts are often caused by industrial process. If the waste is not managed correctly, it can cause water, soil, air pollution, and environmental damage and harm the human health. The nature of heavy metals is difficult to degrade and can accumulate in the body of living things, causing disease. Heavy metals or metalloids such as Pb, Hg, As, Cu, Zn, Cr, Cd, Sr, Fe, Mn, Sn, Ni, Cs, and U are pollutants that pollute the soil the most because of their mobility and solubility [1]. Although heavy metals pollute only a tiny part of the soil, it has a very high risk for surrounding environment [1], because it can enter the food chain and accumulate long term in the body. The accumulation of heavy metals is because they cannot be degraded both biologically and chemically [2].
There are various technologies for remediation of soil contaminated with heavy metals, including bioremediation, phytoremediation, permeable reactive barrier, electrokinetic separation, pump and treat system, and soil cover and coating [3]. Electrokinetic remediation is technology that can be a solution in removing metal content in polluted soil because it can be used on various types of soil, both fine-grained soil [2], mud, and ash from combustion. Remediation using electrokinetic is a process that involves an electric field on polluted soil produced by applying a direct current (DC) to the electrode [4]. Therefore, this study aims to conduct a bibliographical analysis of the publication of electrokinetic remediation with the permeable reactive barrier to find gaps from previous studies and to assess the effect of the type and location of reactive barrier placement on the removal of metal content in polluted soil.

2. Methodology

2.1. Systematic review

The bibliographic analysis consists of collecting and analyzing step to the information related to scientific articles [5]. In conducting the bibliographic analysis, PRISMA method was used to identify and analyze publications related to electrokinetic remediation in metal contaminated soils combined with permeable reactive barrier technology. The PRISMA method is an analytical method consisting of the stages of identification, screening, eligibility, and inclusion [6]. The article database was obtained from Scopus, which was accessed on June 30, 2021. Several keywords and limitations were used for elimination at the identification stage. The keyword "electrokinetic remediation" (1446 documents), followed by the keywords "metal" (1223 documents), "soil" (1209 documents), and "permeable reactive barrier" (214 documents). In addition, the limitation of the publication year from 2012 to 2021 (183 documents); types of article documents, review journals, and conference journals (180 documents); and writing in English (169 documents). At the screening stage, elimination is carried out based on the title and abstract. From the results of the elimination obtained 69 documents that are relevant to the research objectives. Furthermore, elimination is carried out at the eligibility stage based on the research method, and 40 relevant documents are obtained. In the inclusion stage, 26 selected documents, less relevant to research related to the types of pollutants in the polluted soil set aside, are eliminated.

2.2. Data analysis

The selected documents are then downloaded in RIS format from Scopus. Bibliometric analysis was carried out to find gaps from previous studies. This analysis can be done using VOSviewer 1.6.15, which can display keyword relationships in selected documents in the form of a complex and dynamic map [6]. The downloaded database in RIS format is inputted into VOSviewer and analyzed using co-occurrence analysis type with keyword analysis unit. The selected keywords are then automatically eliminated again to get the most relevant keywords to the objectives to be researched.

3. Result and discussion

3.1. Descriptive analysis

The trend of publications on electrokinetic remediation of metal removal in contaminated soils with permeable reactive barriers fluctuated from 2012 to 2021. Publications on electrokinetic remediation with permeable reactive barriers in 2013 experienced an increase of 12% in 2016 and 2019. The addition of publications was 4% in 2014 and 2018, which was less than the previous year. An 8% increase in publications occurred in 2017 and a 15% increase in publications in 2021. The most prominent number of publications was in 2020, reaching 27% from the previous year and with the number of publications being 7 documents. The number of publications on electrokinetic remediation of metal removal in contaminated soil with permeable reactive barriers can be seen in Figure 1.
China, Iran, Australia, and Taiwan have the most publications on metal removal electrokinetic remediation in contaminated soils with the highest permeable reactive barrier. China has an outstanding contribution to research on remediation of metal-contaminated soils because the rapid progress of modern industry in China causes soil pollution [7], so a method is needed to restore the polluted soil. There are 16 different journals publish research on electrokinetic remediation to remove metal content in soil using a permeable reactive barrier. Journal of Environmental Science and Pollution Research has the highest contribution, which is 19% of the total publication found. Furthermore, other journals with the most contributions include Chemosphere and Chemical Engineering Journal as much as 12% and Journal of Hazardous Materials and Separation and Purification Technology as much as 8%.

**Figure 1.** Number of publications on metal removal electrokinetic remediation in soil contaminated with permeable reactive barrier.

**Figure 2.** Co-occurrence keyword map of publications related to electrokinetic remediation using permeable reactive barrier based on their relationship.
3.2. Bibliographic analysis
The relationship between keywords and the division of keyword clusters is shown in Figure 2. Cluster 1 with the highest number of keyword variations is shown in red. In general, this cluster grouped critical words related to electrokinetic remediation mechanisms. Cluster 2 is shown in green, where grouped keywords are related to the utility of electrokinetic remediation. Cluster 3 is blue, a group of keywords associated with factors affecting electrokinetic remediation.

Based on the year of publication, the keywords "chromium" and "electrohydrodynamics," indicated in purple, are primarily found in older articles, namely the year of publication in 2016 and below. Meanwhile, the keywords "heavy metals," "ecosystem restoration," "soil pollution control," and "cadmium," which are indicated by yellow, are often found in recent articles. This condition shows that electrokinetic remediation on polluted soil is currently being developed and is becoming a concern in the ecosystem and environmental restoration efforts. More fully, the analysis of keyword groups based on the year of publication is shown in Figure 3.

![Figure 3](image)

**Figure 3.** Keyword map of publications related to electrokinetic remediation using permeable reactive barrier based on the year of publication.

3.3. Contaminant type

![Figure 4](image)

**Figure 4.** The number of metals used as targets of electrokinetic remediation with permeable reactive barrier.
Based on the analysis results, the removal of Cr content in contaminated soil using electrokinetic remediation with PRB was the most studied. This situation is because Cr(VI) can dissolve over a wide pH range. In addition, during migration to the anode, the species will change according to the existing pH conditions. Therefore their movement and mobilization need to be investigated [8]. Cr(VI) is carcinogenic which can be a soluble oxyanion [9]. Cr metal pollution occurs due to various human activities, such as industrial activities, littering, metal smelting activities, thus requiring fast and precise handling [9].

3.4. PRB material
Various kinds of materials have been developed as reactive barriers in electrokinetic remediation, both in composite materials and materials from waste recycling. In general, zero-valent iron is the most widely used material, either with a combination of other materials, zero-valent iron with activated carbon [11], zero-valent iron with quartz sand [12], zero-valent iron with zeolite [13] or without combination [4,14]. Based on the research of Saeedi et al. (2013) for the removal of Cr in kaolinite, the use of nano zero-valent iron as PRB was the most optimum when combined with 0.1 M EDTA as catholyte [4]. It was found that the reduction of Cr reached 63.75%, and the reduction reached 18.9%. The same study was also conducted by Zheng et al.(2020) using zero-valent iron grain to remediation soil contaminated with Cr [14]. This study used citric acid as an electrolyte and obtained a higher Cr removal of 82.86%. The combination of zero-valent iron with zeolite can remove Cr metal up to 97% and reduce Cr (VI) to Cr (III) by 98% [13]. Besides being effective in the removal of Cr, zero-valent iron is also effective in the removal of other heavy metals in contaminated soil, such as Xiao et al. (2020), who conducted a study on uranium (U) removal in soil samples obtained in the area near uranium tailings mining by combining zero-valent iron with activated carbon as a reactive barrier [11]. Pre-treatment of uranium-contaminated soil samples using a mixture of citric acid and FeCl$_2$ resulted in a U removal efficiency of 80.58 ± 0.99% and could even reduce U(VI) to U(IV).

| References          | Target Contaminant | Reactive Barrier                                                                 |
|---------------------|--------------------|----------------------------------------------------------------------------------|
| Cappai et al., 2012 | Cr, As             | Red mud                                                                          |
| Yu et al., 2019     | Cr                 | Cetyl trimethyl ammonium bromide-zeolite (CTMAB-Z) and Fe(0) mixture             |
| Wang et al., 2019   | Cr                 | Polypyrrole-linen fabric (PPy-LF)                                                |
| Zheng et al., 2020  | Cr                 | Zero valent grain                                                                |
| Xu et al., 2016     | Cr                 | Hydrocalumite (CaAl-LDH)                                                         |
| Xu et al., 2017     | Cr                 | Hydrocalumite (CaAl-LDH)                                                         |
| Xu et al., 2019     | As and Cr          | Hydrocalumite (CaAl-LDH)                                                         |
| Saeedi et al., 2013 | Cr                 | Nano zero-valent iron                                                           |
| Suzuki et al., 2014 | Cr                 | Fe$_3$O$_4$                                                                      |
| Wang et al., 2021   | Cr                 | Hyperbranched polyethyleneimine (HPEI)-enriched polyacrylonitrile (PAN) electrospun nanofiber membrane (ENFM) |
| Nasiri et al., 2020 | Cr                 | Fe$_3$O$_4$                                                                      |
| Fu et al., 2013     | Cr                 | Fe$^0$ and zeolite mixture                                                        |
| Hu, 2013            | Cd                 | Nanometer powder Fe, granular activated carbon, and sand mixture                  |
| Xu et al., 2020     | Cd                 | Porous graphene oxide (PGRO) carbon nanomaterial                                  |
| Zhou et al., 2020   | Cd                 | Zeolite and ZVI mixture                                                          |
| Ding et al., 2017   | Cd                 | Humin                                                                            |
| References          | Target Contaminant | Reactive Barrier                                      |
|---------------------|-------------------|------------------------------------------------------|
| He et al., 2021     | Cd and Zn         | SA/PVA/MgO/ATP hydrogel                              |
| Ghobadi et al., 2021| Cd, Cu, Pb, and Zn| Organic compost                                      |
| Ghobadi et al., 2021*| Cu               | Compost                                             |
| Ghobadi et al., 2020| Cu               | Biochar                                             |
| Ribeiro et al., 2018| Pb               | Eggshell inorganic fraction powder (EGGIF)          |
| Xiao et al., 2020   | U                | ZVI and AC mixture                                   |
| Yao et al., 2020    | As               | Activated carbon                                    |
| Zanjani et al., 2012| Ni               | Activated carbon                                    |
| Zhou et al., 2016   | F                | Activated alumina and calcium chloride mixture       |
| Zhu et al., 2016    | F                | Bamboo charcoal                                      |

However, Fe$_3$O$_4$ has a better ability than Fe0 in recovering Cr to Cr(III) [17]. Nasiri et al. (2020) used Fe3O4 as a reactive barrier and combined it with a chelating agent [19]. The results showed that the combination with the chelating agent EDTA had a more significant effect on Cr removal when compared to the combination using the chelating agent citric acid. In addition, activated carbon and hydrocalumite (CaAl-LDH) are also widely used as reactive barriers in metal-contaminated soil’s electrokinetic remediation. Activated carbon can remove Ni from the soil without reverse osmosis at a voltage gradient of 1.25 V/cm [29]. However, in removing Cd from contaminated soil, according to Ghobadi et al. (2020), biochar can capture more Cd than activated carbon [27]. Meanwhile, research on reactive barrier hydrocalumite (CaAl-LDH) has been extensively studied by Xu et al. (2016) by comparing the efficiency of Cr metal removal using hydrocalumite (CaAl-LDH) as a reactive barrier with different variations of soil moisture [15]. The removal efficiency of Cr can reach 96.6% for Cr(IV) and 67.3% for total Cr at 40% soil moisture. Then Xu et al. (2017) using the same reactive barrier combining electrokinetic remediation with different voltage gradients [16]. The highest Cr(VI) and total Cr removal were obtained by applying a voltage gradient of 1 V/cm. Xu et al. (2019) investigated the pre-treatment using sodium citrate in electrokinetic remediation with hydrocalumite (CaAl-LDH) having a higher effect on As removal than Cr, while the use of EDTA-2Na pre-treatment had a higher effect on Cr removal than As [10]. As the research of Xu et al. (2017), in Cappai et al. (2012), red mud as a reactive barrier is more effective in removing Cr than As [8]. Thus it can be concluded that As metal is more difficult to clean than Cr metal. Therefore, it is necessary to develop research on the efficient reactive barrier in As removal in soil.

Reactive barriers can come from composite materials, such as polypyrrole-linen fabric (PPy-LF) and modified activated alumina. PPy-LF with a molar ratio of SDBS:CTAB = 1:2 has a Cr removal rate of 93.1%. Cr(VI) on the surface of PPy-LF will be reduced to Cr(III), some of which will settle on the surface of PPy-LF, and the rest will enter the soil [7]. Modifying activated alumina with the addition of calcium chloride as a reactive barrier can increase the defluorination ability of activated alumina [30] so that it is better at removing fluoride content in contaminated soil. Reactive barriers can also come from organic matter, for example, peat soil [23], compost, biochar [26], and bamboo charcoal in removing fluoride [31].

3.4. **PRB placement**

Several important reactions occur during electrokinetic remediation, including adsorption-desorption, precipitation-dissolution, crystallization, and corrosion of the electrodes at the anode [32]. Proper placement of reactive barriers can prevent the negative impact of the reactions during the electrokinetic remediation process. Replace reactive barriers in the electrokinetic remediation process also needs to consider the type of pollutant to be removed. Heavy metal cations will migrate towards the cathode [1], so the location of the reactive barrier should be placed close to the cathode. Meanwhile, heavy metal
anions will migrate towards the anode [2]. Therefore a reactive barrier should be placed near the anode. The placement of the right reactive barrier effectively reduces and even eliminates pollutant pollution in the anode and cathode chambers, thereby reducing the processing of liquid waste in the anode and cathode chambers.

Nasiri et al. (2020) conducted a study on the effect of PRB position on Cr removal [19]. The placement of PRB in the middle has a higher Cr removal efficiency than the PRB position on the anode and cathode sides. However, PRB located on the anode and cathode side caused the removal in all parts of the soil to be more uniform than PRB located in the middle. A similar study was conducted by Xiao et al. (2020) in removing uranium on polluted soil [11]. The most optimum PRB placement is placed on three sides, namely at the anode, cathode, and middle. Different things are shown by Zanjani et al. (2012) research to remove Ni, where the most optimum PRB location is on the cathode side [29].

4. Conclusions

Bibliographic analysis was carried out using the PRISMA method and supported by VOSviewer software to map and tabulate information more concisely and efficiently. Based on the results of the elimination of scientific articles, 26 relevant scientific articles were obtained. Scientific articles on electrokinetic remediation with PRB on the heaviest metal contaminated soil in 2020 with 27% addition. China, Iran, and Australia are the countries that contributed the most to the research. Environmental Science and Pollutant Research is the journal with the most publications related to EK-PRB for heavy metal contaminated soil. The most studied elimination target is Cr. The author most actively collaborating on research related to EK-PRB in heavy metal polluted soil is D. Li. The mapping results using VOSviewer show three cluster groups, namely the electrokinetic remediation mechanism cluster, the use of electrokinetic remediation, and factors that affect electrokinetic remediation. It was found that in determining the type of PRB material and the position of PRB in the electrokinetic remediation process, it is necessary to consider the type of pollutant to be cleaned so that it can produce significant removal efficiency by minimizing the negative impact of physical and chemical reactions that occur during the electrokinetic remediation process.

Acknowledgments
This research is funded by SAPBN Undip 2021 under scheme of Program Penelitian Kolaborasi Indonesia (PPKI) Tahun 2021 Number 117-07/UN7.6.1/PP/2021

References
[1] Moghadam M J, Moayedi H, Sadeghi M M and Hajiannia A 2016 Environ Geochem Health 38 1217-1227
[2] Wang Y, Li A and Cui C 2021 Chemosphere 265 129071
[3] Camenzuli D, Freidman B L, Statham T M, Mumford K A and Gore D B 2013 Polar Res 32(1) 1751-8369
[4] Saeedi M, Li L Y, and Moradi Gharehtapeh A 2013 Int J Environ Res 7(1) 39-50
[5] Bruni A, Serra F G, Gallo V, Deregibus A, and Castroflorio T 2021 Am J Orthod Dentofacial Orthop 159 (4) e343-e362
[6] Budihardjo M A, Ramadan B S, Putri S A, Wahyuningrum I F S and Muhammad F I 2021 Sustainability 13 6562
[7] Wang Y, Huang L, Wang Z, Wang L, Han Y, Liu X and Ma T 2019 Chem Eng J 373 131-139
[8] Cappai G, De Gioannis G, Muntoni A, Spiga D and Zijlstra J J P 2012 Chemosphere 86 400-408
[9] Yu X, Muhammad F, Yan Y, Yu L, Li H, Huang X, Jiao B, Lu N and Li D 2019 R. Soc Open Sci 6 182138
[10] Xu Y, Li J, Xia W, Sun Y, Qian G and Zhang J 2019 Environ Sci Pollut Res 26 3392-3403
[11] Xiao J, Pang Z, Zhou S, Chu L, Rong L, Liu Y, Li J and Tian L 2020 Sep Purif Technol 244 116667
[12] Yao W, Cai Z, Sun S, Romantschuk M, Sinkkonen A, Sun Y and Wang Q 2020 *J Soil Sediment* **20** 1526-1533
[13] Fu R B, Liu F, Zhang C B and Ma J 2013 *Environ Eng Sci* **30**(1) 17-22
[14] Zheng Y, Yan Y, Yu L, Li H, Jiao B, Shiau Y and Li D 2020 *Environ Sci Pollut Res* **27** 5572-5583
[15] Xu Y, Xu X, Hou H, Zhang J, Zhang D, Qian G 2016 *Environ Sci Pollut Res* **23** 6517-6523
[16] Xu Y, Xia W, Hou H, Zhang J and Qian G 2017 *Environ Sci Pollut Res* **24** 20479-20486
[17] Suzuki T, Kawai K, Moribe M and Niinae M 2014 *J Hazard Mater*
[18] Wang J, Hou L, Yao Z, Jiang Y, Xi B, Ni S and Zhang L 2021 *Chem Eng J* **406** 126822
[19] Nasiri A, Zanjani A J and Darban A K 2020 *Environ Pollut* **266** 115197
[20] Hu H 2013 *Adv Mater Res* **675** 188-191
[21] Xu J C, Ma Q, Chen C, Wu Q T and Long X X 2020 *Chemosphere* **259** 127441
[22] Zhou H, Xu J, Lv S, Liu Z and Liu W 2020 *Sep Purif Technol* **239** 116544
[23] Ding L, Lv W, Yao K, Li L, Wang M and Liu G 2017 *Environ Sci Pollut Res* **24** 3430-3436
[24] He C, Hu A, Wang F, Zhang P, Zhao Z, Zhao Y and Liu X 2021 *Chem Eng J* **407** 126923
[25] Ghobadi R, Altaae A, Zhou J L, McLean P and Ganbat N 2021 *J Hazard Mater* **402** 123891
[26] Ghobadi R, Altaae A, Zhou J L, Karbassiayazdi E and Ganbat N 2021 *Sci Total Environ.* **794** 148668
[27] Ghobadi R, Altaee A, Zhou J L, McLean P and Yadav S 2020 *Chemosphere* **252** 126607
[28] Ribeiro A, Mota A, Soares M, Castro C, Araujo J and Carvalho J 2018 *Key Eng Mater* **777** 256-261
[29] Zanjani A J, Saeedi M and Weng C H 2012 *Environmental* **5**(2) 28-35
[30] Zhou M, Zhu S, Yi Y and Zhang T 2016 *Clean Tech. Environ Policy* **18** 2691-2699
[31] Zhu S, Han D, Zhou M and Liu Y 2016 *Electrochim Acta* **198** 241-248
[32] Wen D, Fu R and Li Q 2021 *J. Hazard Mater* **401** 123345