

ISSN: 2230-9926

## **RESEARCH ARTICLE**

Available online at http://www.journalijdr.com



International Journal of Development Research Vol. 15, Issue, 04, pp. 68197-68200, April, 2025 https://doi.org/10.37118/ijdr.29330.04.2025



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# ANALYSIS OF CLOGGING FAILURE MODES IN PERMEABLE REACTIVE BARRIERS (PRBS) DURING GROUNDWATER REMEDIATION OF LANDFILL CONTAMINATION

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### **ARTICLE INFO**

*Article History:* Received 11<sup>th</sup> January, 2025 Received in revised form 19<sup>th</sup> February, 2025 Accepted 17<sup>th</sup> March, 2025

Accepted 17<sup>th</sup> March, 2025 Published online 30<sup>th</sup> April, 2025

*Key Words:* Permeablereactive barriers, Contaminated groundwater, Landfill leachate, Clogging.

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### ABSTRACT

Groundwater is a crucial component of water resources, yet the situation of groundwater pollution in China is severe, with municipal solid waste landfills being one of the primary sources of contamination. Permeable reactive barriers (PRBs), a passive in-situ remediation system, offer numerous advantages in treating groundwater contaminated by landfills but are prone to clogging issues. This study examines the clogging problem in PRBs during the remediation of contaminated groundwater from both exogenous and endogenous particulate perspectives, exploring the clogging patterns of PRBs. It reveals that the sources and particle sizes of solids/flocs in contaminated groundwater are diverse, with minimal retention by the site's permeable layer. These particles are adsorbed and intercepted by the PRB, while the interaction between contaminated groundwater and PRB fill materials generates new solids/flocs, which deposit or adhere within the pores of the reactive materials, reducing the permeability of the PRB. PRB clogging failure is often the result of the combined effects of exogenous and endogenous particulate to the pore structure of the PRB materials.

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*Citation: ZHAO Ziyi. 2025.* "Analysis of Clogging failure modes in Permeable reactive Barriers (PRBS) during Groundwater Remediation of Landfill contamination". *International Journal of Development Research*, 15, (04), 68197-68200.

# **INTRODUCTION**

Groundwater, stored in the pores, fractures, and cavities of rocks beneath the earth's surface, is a vital component of water resources (YOUNGER, 2009). Currently, groundwater pollution in China is becoming increasingly severe, with some regions facing particularly acute issues. Among these, municipal solid waste landfills have been identified as a major source of groundwater contamination (HAN, 2016). Pollutants in groundwater primarily include heavy metals (e.g., manganese, iron, aluminum), organic contaminants (e.g., volatile organic compounds, polycyclic aromatic hydrocarbons, pesticide and fertilizer residues), inorganic salts (e.g., nitrates, nitrites, sulfates, phosphates, and fluorides), and pathogens (HAN, 2016; Huang et al., 2014 and KURWADKAR, 2017). Contaminated groundwater poses significant risks to human health, making the remediation of groundwater polluted by landfills an urgent priority. Currently, groundwater remediation technologies are broadly categorized into in-situ and ex-situ methods (Huang, 2021). In-situ remediation, with its cost-effectiveness and minimal environmental disruption, has gradually become the primary approach for groundwater pollution control (WANG, 2022). Permeable reactive barriers (PRBs) are a passive in-situ remediation system. They involve excavating a narrow trench along the flow path of contaminated groundwater and installing a permeable barrier filled with reactive materials to intercept and treat the contaminant plume (Shu, 2002). PRBs require no external power, occupy no surface space, have low costs, allow for material replacement and repair, and can effectively remove a wide

range of pollutants. Once installed, they incur almost no operational costs and have minimal ecological impact, making them widely applicable (Shu, 2002). However, PRBs are prone to clogging during operation (OBIRI-NYARKO, 2014), especially when treating groundwater contaminated by landfills. This is because landfillcontaminated groundwater is complex in composition, containing substantial solid materials (Han, 2015). Additionally, the interaction between PRB fill materials (e.g., zero-valent iron, zeolite, activated carbon) and contaminated groundwater can generate new solid substances (BUDANIA and DANGAYACH, 2013). While most current research focuses on optimizing fill material ratios or developing new materials to mitigate PRB clogging (ZHOU, 2014; Chen, 2023; LI, 2024; PAN, 2024 and YUAN, 2024), studies on the mechanisms of PRB clogging, particularly from the perspectives of exogenous and endogenous particulates, are scarce. This study investigates the clogging issues in PRBs during the remediation of contaminated groundwater by examining both exogenous and endogenous particulate clogging. It also explores PRB clogging patterns based on the characteristics of landfill groundwater contamination and geological conditions. This research holds significant implications for decision-making in PRB remediation of landfill-contaminated groundwater.

# **MATERIALS AND METHODS**

Landfill Leachate and Groundwater Mixing Experiment: To eliminate the interference of solid suspended particles naturally

present in leachate and groundwater on the determination of whether their interaction generates flocs or precipitates, a refrigerated centrifuge was used to centrifuge both leachate and groundwater separately. The leachate and groundwater were centrifuged at 8,000 rpm for 40 minutes. Then, 250 mL of the supernatant from each was mixed (v/v = 1:1) and left to stand for 48 hours. A portion of the mixed water was collected for further use.

Interaction between PRB Fill Materials and Contaminated Water: Landfill leachate and groundwater were mixed at a 1:1 ratio (v/v = 1:1). Zero-valent iron (ZVI) powder (particle size: 100  $\mu$ m), zeolite (particle size: 1–2 mm), and activated carbon (particle size: 1–2 mm) were separately placed into 500 mL beakers, each filled to a volume of 250 mL. Contaminated groundwater was then poured into each beaker until the liquid level reached 500 mL. After standing for 48 hours, a portion of the water was collected for further use.

*Particle Size Analysis of Solid Particles:* The particle sizes of solids/flocs in landfill leachate, groundwater, and the mixed liquid were measured using laser particle size analysis.

**Pore and Throat Analysis:** The connected pores of the extracted core samples were equivalently replaced using a classical sphere-rod model. In this model, large pores were replaced by spheres of equivalent radius, and the connecting throats between pores were replaced by rods of equivalent radius. The sphere-rod model allowed for statistical analysis of the connectivity and size distribution of pores and throats in the core samples.

# RESULTS

Analysis of Exogenous Particulate Clogging Mechanisms: Exogenous particulates refer to particulate matter carried into the PRB by contaminated groundwater. These primarily include solids/flocs from landfill leachate (WU, 2014 and GUO, 2006), solids/flocs from groundwater (ZHAI, 2019 and BAUMANN, 2006), and solids/flocs generated by the interaction between landfill leachate and groundwater (ZHAI, 2019 and FATTA, 1999). A schematic diagram of exogenous clogging is shown in Figure 1. The main factors contributing to exogenous particulate clogging in PRBs, besides the size of solids/flocs carried by contaminated groundwater, include the development of pores and fractures in the permeable layer of the landfill site (CARLIER, 2019 and ZHAN, 2023). As groundwater flows through the permeable layer, the layer partially retains the solids/flocs.



Figure 1. External-source particle blockage illustration

*Analysis of Exogenous Particulate Clogging:* Taking a municipal solid waste landfill in Sichuan as an example, the particle sizes of solids in groundwater and leachate were analyzed using a laser particle size analyzer. The particle size distribution of solids/flocs in groundwater is shown in Figure 2a, while that in leachate is shown in Figure 2b. The solids/flocs in the groundwater of this site are primarily distributed between 140 nm and 310 nm, with few exceeding 310 nm. In contrast, the particle size distribution of solids/flocs in the leachate is concentrated around 27 nm and 210 nm, with a very small fraction exceeding 500 nm. To analyze the

solids/flocs generated by the interaction between landfill leachate and groundwater in the transport pathway, the original solids/flocs in leachate and groundwater were removed using refrigerated centrifugation. The centrifuged groundwater and landfill leachate were then mixed, and the particle size distribution of the resulting solids/flocs was measured after a reaction period. The results, shown in Figure 3, indicate that the interaction between groundwater and leachate produced solids/flocs ranging from 15 nm to 10 µm, with the majority concentrated between 1 µm and 3 µm. Similar findings were reported by Zhai et al. (ZHAI, 2019), who studied upstream groundwater (uncontaminated), groundwater near the landfill (heavily contaminated), and downstream groundwater. These results collectively demonstrate that the solids/flocs in contaminated groundwater are caused by multiple factors. Additionally, when these solids/flocs are carried by water flow through the PRB, they are removed due to interception and adsorption mechanisms.



Figure 2. Particle size distribution of Solid/flocculent matter in water (a) Solid/flocculent matter in leachate (b)



Figure 3. Particle size distribution of solids/flocs after leachategroundwater interaction

Analysis of Fracture and Pore Structure in the Permeable Layer: The distribution patterns of pores and throats in the pore structure of the permeable layer at a municipal solid waste landfill in Sichuan are shown in Figure 4, while the aperture distribution of fractures is presented in Table 1. The pore diameters are primarily distributed between 20 µm and 120 µm, and the throat diameters are mainly concentrated between 5 µm and 60 µm. The minimum fracture aperture is 81.8 µm, with an average of 368.5 µm. The particle sizes of solids and flocs in the leachate and groundwater at this site are one order of magnitude smaller than the pore and throat diameters of the permeable layer and two orders of magnitude smaller than the fracture apertures. The particle sizes of solids and flocs generated by the interaction between groundwater and leachate are also significantly smaller than the pore and fracture dimensions. Therefore, the retention of solids/flocs in the contaminated water by the permeable layer of this site is negligible. The solids/flocs in the contaminated water directly interact with the PRB. Larger particles are directly adsorbed and intercepted by the PRB, while smaller and slower-moving particles can settle and remain within the PRB<sup>[22]</sup>. This negatively impacts the permeability of the PRB. Over time, severe clogging can lead to PRB failure. This conclusion is further supported by core samples obtained from geological drilling at the site (Figure 5), which revealed a significant amount of dark black solids/flocs attached to the permeable layer cores.



Figure 4. Distribution patterns of pores (a) and throats (b) in pore structure

#### Table 1. Fracture aperture distribution

Mean (µm)	Minimum (µm)	Maximum (µm)	Median (µm)
368.49	81.84	10549.31	104.13



Figure 5. Permeable Layer Core

*Analysis of Endogenous Particulate Clogging Mechanisms:* Endogenous solids/flocs clogging refers to the generation of new solids/flocs due to chemical, physical, or biological interactions between PRB reactive materials and contaminated groundwater (Thiruvenkatachari, 2008). These solids/flocs deposit or adhere within the pores of the PRB reactive materials (Flury, 2009), thereby reducing the permeability of the PRB and, in severe cases, leading to PRB clogging failure.



Figure 6. Endogenous solid/flocculent clogging schematic

A schematic diagram of endogenous solids/flocs clogging is shown in Figure 6. Unlike exogenous particulate clogging, endogenous clogging does not require migration pathways external to the PRB. Its primary influencing factors include the composition of landfill leachate, groundwater components, the types of PRB fill materials, and the pore structure of the PRB. The composition of leachate and groundwater is complex and variable (ZHAI, 2019; BAUMANN, 2006 and FATTA, 1999), but the types of PRB fill materials are diverse, and the pore structure can be adjusted as needed. Therefore, the selection of PRB fill materials and their particle sizes is crucial for the long-term effective remediation of contaminated water bodies.

Analysis of endogenous particulate matter blockage: Similarly, taking a municipal solid waste landfill in Sichuan as an example, the interaction between groundwater-landfill leachate and typical PRB reactive materials such as ZVI, activated carbon, and zeolite was studied. The particle size of the resulting liquid was analyzed, and the results are shown in Figure 7. The comparison revealed that the interaction of contaminated groundwater with ZVI, activated carbon, and zeolite generated new solids/flocs, whose particle sizes were significantly larger than those of the original solids/flocs in the groundwater and leachate. Among these, the solids/flocs produced by the interaction with ZVI had the largest particle size, followed by those generated by the interaction with zeolite. These solids/flocs were partially adsorbed and deposited as particles between the grains, while others formed films attached to the surface of the PRB reactive materials. This leads to a reduction in the permeability of the PRB and, in severe cases, causes PRB clogging failure.





# DISCUSSION

The solids/flocs in contaminated groundwater originate from diverse sources (solids/flocs in groundwater, solids/flocs in landfill leachate, and solids/flocs generated by the interaction between groundwater and landfill leachate) and exhibit a wide range of particle sizes. However, the pore, throat, and fracture dimensions of the permeable layer at this site are relatively large, resulting in negligible retention of solids/flocs. These particles are instead adsorbed and intercepted by the PRB. Additionally, the interaction between contaminated groundwater and PRB fill materials generates new solids/flocs, the size of which varies depending on the fill material. For example, those produced by interaction with ZVI are notably larger. These solids/flocs deposit or adhere within the pores of the reactive materials, reducing the permeability of the PRB. PRB clogging failure is often not caused solely by either exogenous particulates or endogenous solids/flocs; both typically coexist. Nevertheless, both endogenous and exogenous solids/flocs clogging are closely related to the pore structure of the PRB materials. Therefore, analyzing the pore structure of PRB materials with different forms and sizes is crucial for ensuring the long-term effective operation of PRBs.

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