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PERMEABLE REACTIVE BARRIER: A TECHNOLOGY FOR GROUNDWATER REMEDIATION - A MINI REVIEW

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ABSTRACT: The pollution of groundwater by organic or inorganic pollutants, originating from either soil leaching or anthropogenic activities, is one of the major environmental issues. Remediation of this water source is of highest priority because many countries use it for drinking purpose. Pump-and-treat method is represented for many decades the major technique to treat groundwater infected with organic/inorganic pollutants. In last two decades, this technique becomes to be in lack with the sense of modern concepts of sustainability and renewable energy. Permeable reactive barriers (PRBs) technology was introduced as an alternative method for traditional pump-and-treat systems to remediate contaminated groundwater that was achieving these concepts. Within this issue, this technology has been proven to be a successful and most efficient promising method used by many researchers and in several projects due to its direct and simple techniques to remediate groundwater. A rapid progress from bench scale to field scale implementation in the PRB technique is recognized through the last few years. In addition, this technique was modeled theoretically for characterizing the migration of contaminants spatially and temporally through the barrier and, consequently, these models can be used for estimating the longevity of this barrier. An overview of this technique and the promising horizons for scientific research that integrates this method with sustainability and green technology practices are presented in the present study.

KEYWORDS: Groundwater (GW), Adsorption, Contamination, Permeable reactive Barrier (PRB), COSMOL.

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1.INTRODUCTION

Permeable reactive barrier (PRB) technique is mostly perceived as a physical method for remediating contaminated groundwater, due to its design and mechanism of pollutant removal. Nevertheless, researchers [1; 2] reported that biological reaction is one of the several mechanisms (degradation, precipitation and sorption) of pollutant removal in PRB technique. Although alternative terms such as biological PRB, passive bioreactive barrier, bio-enhanced PRB have been proposed to accommodate the bioremediation or biotechnology aspect of the technique, the role of microorganisms have been reported to be mostly enhancement rather than an independent biotechnology [3]. In general, PRB is an in situ technique used for remediating groundwater polluted with different types of pollutants including heavy metals and chlorinated compounds (Table 1).

Table 1: Some pollutants removed by permeable reactive barriers (PRBs) technique

Reactive material	Nature of pollutant	Initial concentration	Mechanism of pollutant removal	% Removal	References
Clay	Cs-137	10^5 Bq/m^3	Sorption	--------	4
Oxygen reactive compound and clinoptilolite	NH_4–N	5–11 mg/L	Ion exchange and biological nitrification	>99	5
Natural pyrite (FeS_2)	Cr(VI)	10–100 mg/L	Sorption	27–100	6
Zero-valent iron coupled with polyhydroxybutyrate	1, 2-dichloroethane	10 mg/L	Biological degradation	20–80	7
Mixture of zero-valent iron, Zeolite and activated carbon	Landfill leachate	------------	-----------	55–94	8
Bio-barrier (*Arthrobacter viscosus*)	Polyaromatic hydrocarbons	100 µM	Biodegradation	>80	9
Bio-barrier (*Trametes versicolor*, white-rot fungi)	Orange G dye	150 mg/L	Biodegradation	97	10
Organic substrates and zero-valent iron (ZVI)	Heavy Metals (Al, Zn and Cu)	15, 20 and 1.2 mg/L	Precipitation	>95	11
Granular oxygen-capturing materials (ZVI powder,	Nitrate and nitrite	40 mg/L	Biodegradation	>94	12

sodium citrate and inorganic salts) and granular activated carbon					
Bioaugumented Bio-barrier (*Mycobaterium* sp. and *Pseudomonas* sp. immobilized bead) PRB	Benzene, toluene, ethylbenze and xylene (BTEX)	100 mg/L	Biodegradation	84–97	13
Granular iron	Chlorinated volatile organic compounds (VOC)	-------------	Degradation	--------------	14

In this technique, a permanent or semi-permanent reactive barrier (medium) mostly made up of a zero-valent iron [15; 8] is submerged in the trajectory of polluted groundwater. As polluted water flows through the barrier under its natural gradient, pollutants become trapped and undergo series of reactions resulting in clean water in the flow through [1; 2]. Ideally, the barriers are usually reactive enough to trap pollutants, permeable to allow the flow of water but not pollutants, passive with little energy input, inexpensive, readily available and accessible [4] (Figure 1).

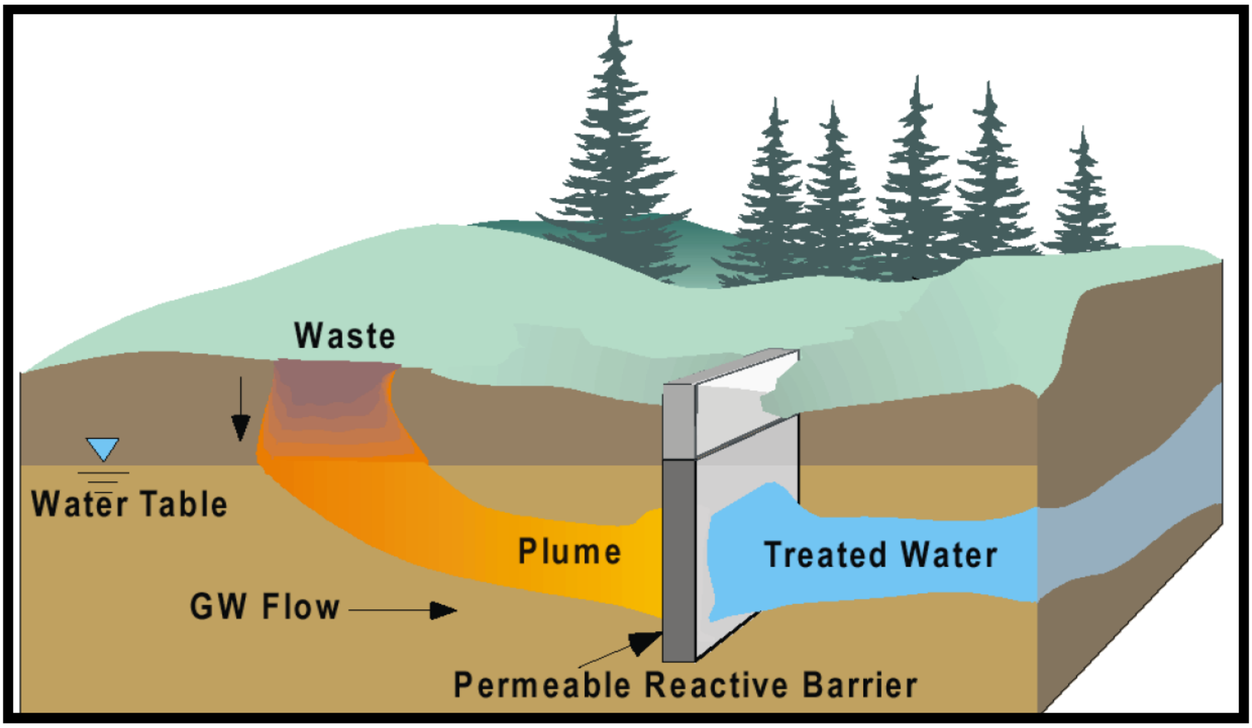

Figure 1: Conceptual Schematic of an In Situ Permeable Reactive Barrier

The effectiveness of this technique depends mostly on the type of media used, which is influenced by pollutant type, biogeochemical and hydrogeological conditions, environmental and health influence, mechanical stability, and cost [2; 6].

The main advantages of PRB application are:

1. PRB's are particularly well suited to locations where the contaminants exist as a soluble or mobile phase in the subsurface groundwater aquifer and can change oxidation state upon contact with the reactive treatment media.
2. PRB's are particularly well suited to contaminant plumes that are heterogeneous in composition and concentration (US EPA, 2000).
3. On-going operation and maintenance functions are generally limited to compliance monitoring and where necessary, replenishment or replacement of reactive treatment media. Therefore, operating costs are typically significantly lower than traditional pump and treat systems.
4. PRB operation does not alienate land use above the PRB. Therefore active site restoration and development can occur above a PRB without affecting its performance and in some cases can improve performance by reducing surface water infiltration into the contaminant plume.

The main disadvantages of PRB application are:

5. PRB's are not well suited to treatment of insoluble/immobile contaminants (i.e. some DNAPLs).
6. PRB's installed in groundwater aquifers with low hydraulic conductivities or groundwater velocities are likely to require a long residence times to treat the contaminants-of-concern. This may result in a significant increase in PRB installation costs and offer no cost advantage over conventional pump and treat systems.
7. Precipitates that may form in a PRB can reduce permeability and long-term effectiveness.
8. Thorough and careful site characterization is required pre-design to allow for consideration of potential changes to groundwater flow or contaminant migration, as post-installation improvements to PRB performance can be difficult and costly.

BODY OF PAPER

Configurations and techniques of PRB

PRBs can be achieved as replaceable, semipermanent, or permanent units. Continuous wall or curtain is the basic configuration of barriers that stands up and transversely faces the direction of the contaminant front. The advantages of this configuration are they: rely on conventional methods of installation, are easy to conceptualize, creating fewer disturbances to the natural groundwater flow pattern, and can be constructed using relatively simple design methods. Furthermore, their effectiveness has been documented in the literature [16; 17; 18; 19]. Starr and Cherry [20] introduced the term ''funnel and gate'' which its concept was first mentioned by McMurtry and Elton [21] and sometime used interchangeably with PRBs; however, funnel and gate configuration consisted of impermeable walls that directed groundwater to the reactive middle gate or panel. The election between these two configurations is based on the characteristics of the reactive medium and site. Expensive reactive materials use funnel and gate configuration to restrict the relatively high construction costs, when compared to continuous barriers [1]. Furthermore, the adoption of funnel and gate configuration promotes the use of double or multi-reactive barriers for multi-action, improving the efficiency of treatment for more than one type of contaminants [22](Figure 2).

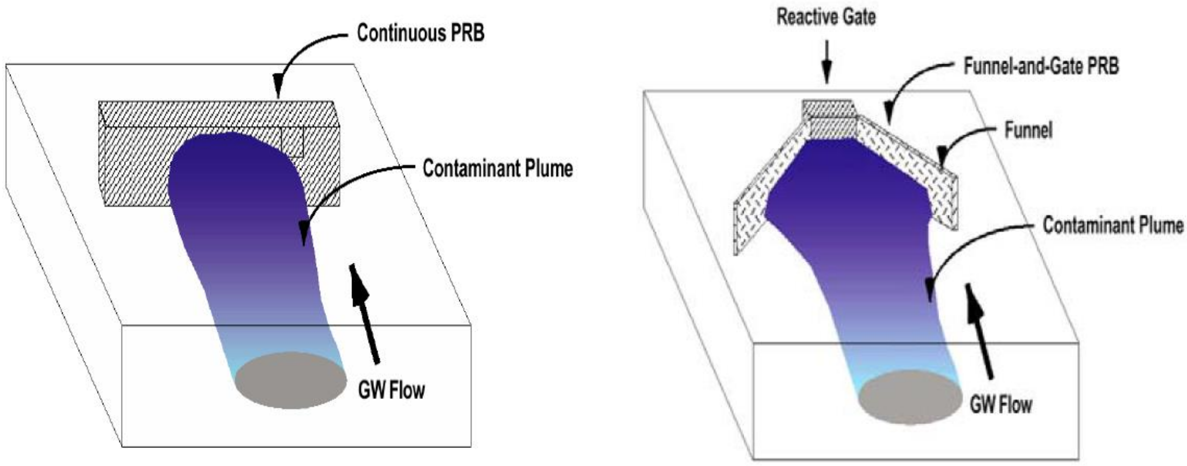

Figure 2: (left) Plume capture by a continuous PRB trenched system. The plume moves unimpeded through the reactive zone; (right) Plume capture by a funnel-and gate system. Sheet piling funnels direct the plume through the reactive gate

Day et al. [23] presented a special type of funnel and gate, which uses a buried vessel to contain the reactive materials in removable/replaceable ‘‘cassettes.’’ The cassette system permitted the regular removal and replacement of the reactive material and/or maintenance of the system without excavating and removing the vessel. Furthermore, Elder [24] achieved groundwater flow through reactive barrier set vertically upward inside caisson formation to more shallow level that assisted in obtaining more uniform flow and easy monitoring of the flow. This configuration is known as caisson PRB [24; 25]. USDOE [26] modified an existing funnel and gate PRB to improve its operation by bringing the effluent through vertical well and in a siphoned manner to zero valent iron (ZVI) treatment vessel, so that there is no remediation in the path till the effluent reaches the vessel, which draws out the treated water finally to the drain field. This configuration was termed as trench permeable reactive barrier (TPRB). However, the trench was meant for groundwater transfer only and there was no treatment effort through it [27]. A similar technique used passive groundwater capture and treatment by reactor cells in the remediation process [1]. A Geo Siphone^{TM} /GeoFlow technology introduced the same technique by utilizing the natural hydraulic gradient between two locations to enhance water flow carrier with plume. This flow was directed toward the cell containing the reactive material, which intercepted with contaminant front to complete the treatment process [27]. Hudak [28] suggested longitudinal reactive barriers instead of transverse barriers with respect to groundwater flow direction and modeled this configuration. The author concluded that longitudinal trenches are suitable choice for narrow contaminant plumes moved with flow in low velocities. As an advancement in this technology, development of undredged reactive barriers that can be suitable for remediation of deep groundwater contaminant plume or confined aquifers was introduced. Istok et al. [29] and Fruchter et al. [30] established an in situ treatment technique to create subsurface permeable reactive zone inside the deep or confined aquifer by injecting treatment reagent using injection non-discharge wells. This system was called ''in situ redox manipulation (ISRM)'' and was developed and applied at the Hanford disposal pond site in the Washington State to achieve the required treatment. Non-excavation techniques such as deep soil mixing [17], hydraulic fracturing, or in situ redox manipulation (ISRM) [31] were used for installation of PRBs at greater depths. Another similar most recent technique is to create virtual PRBs (vPRBs) using quasi-passive in situ groundwater circulation well system, GCW. An in situ vPRB is located within the groundwater contamination plume in combination with overlapped circulation cells; this generates effective hydraulic control within the aquifer through large-diameter spherical capture zone. The polluted water is captured by this system and treated within the aquifer in the well. Several studies have focused on this type of remediation to improve the role of PRB in treating the wider range of pollutants such as dense non-aqueous phase liquids (DNAPLs) with less expensive and more efficient technique in comparison with pump-and-treat system [32]. Furthermore, electrokinetic (EK) concepts have been recently integrated to the PRB technology to improve its functions in groundwater remediation. This process is able to remove contaminants from low-permeable media; thus, the EK process can be used to overcome clogging problems in the PRB due to precipitation reactions [33]. In addition, Chung and Lee [34] investigated the potential application of atomizing slag used as reactive material for PRB in combination with EK for removal of inorganic or organic pollutants from the contaminated groundwater. The results indicated that the applied configuration increased the removal rate of cadmium due to electromigration mechanism. In other work, Cang et al. [35] used a coupled EK with PRB of ZVI for treating Cr-contaminated soil and concluded that this technique is feasible in the clean-up process due to the rationing of the precipitate portions that occur between anode and cathode reservoirs on the one side and PRB porous volume on the other side. Recently, with worldwide spreading of nanotechnology, many researchers have started to utilize nanotechnology concepts to treat passively contaminated groundwater. Rajan [36] summarized the use of nanomaterials such as nZVI and carbon nanotubes (CNT) in groundwater remediation for drinking and reuse. The use of nanotechnology can be considered a faster and more cost-effective solution for in situ remediation [37]. Nanomaterials have been evaluated for use in nanoremediation such as nanoscale zeolites, nanoscale ZVI particles, carbon nanotubes, metal oxides, noble metals and titanium dioxide, nanoclays, magnetic nanoparticles, and nanomembrane. In comparison with other remediation methods, this approach provides an overall reduction in the contaminant levels; however, it is still under research with limited field applications [36;37]. Arau´jo et al. [38] reviewed the researches of using both metallic iron and nanomaterials within permeable reactive barriers to reduce of nitrate concentration in drinking water which has been worldwide prevalence over the last two decades. In general, they support the concept that truly in short term, the utilization of nZVI materials with permeable reactive barriers is a good performance technology for denitrification, but the long-term impact of the use of this materials in this remediation process, both on the environment and on the human health, is far to be conveniently known. They recommended that further research work is needed on this issue to decide that nanosized iron-based permeable reactive barriers for the removal of nitrate from drinking water can be truly considered an eco-efficient technology.

Reactive materials

Reactive media used in permeable barriers should be compatible with the subsurface environment. That is, the media should cause no adverse chemical reactions or by products when reacting with constituents in the contaminant plume and should not act as a possible source of contaminants itself. This requires that the material be well understood and characterized. To keep PRB costs to a minimum, the material should persist over long periods of time, i.e., it should not be readily soluble or depleted in reactivity, and the material should be readily available at a low to moderate cost. This material should minimize constraints on groundwater flow by not having excessively small particle size, and it should not consist of a wide range of particle sizes that might result in blocked inter-granular spaces. Worker safety, with regard to handling the material, should also be considered [39; 40]. Granular activated carbon (GAC), zeolite, ZVI, red mud, fly ash, peat, activated sludge, tree leaves, recycled concrete, shredded cast iron, steel fibers from tires, blast furnace slag, steel slag dust, basalt dust, paper ash, plant shell and weed, bone char, non-living biomass, maize cob, phosphatic compounds, waste foundry sand, etc., are examples for materials that can be used in the PRB for containment of the pollutants [41].

Decontamination mechanisms

The common target contaminants in the groundwater are sorted into two main groups [1]:

1. Organic compounds that include methane, ethane, propane, aromatics compounds, etc.
2. Inorganic compounds that include zinc, cadmium, copper, nickel, chromium, manganese, anion compounds, etc.

The decontamination mechanisms in the PRB can be classified into three categories [39; 42]:

1. <u>Degradation</u> it is a chemical or biological decomposition of contaminants into harmless compounds. An example of chemical degradation is oxidation of ZVI.
2. <u>Precipitation</u> it is retaining contaminants by immobilization, and their chemical state is not altered. For example, by increasing the pH, some metals are reduced and precipitated in the form of sulfites or hydroxides.

3. Sorption it is retaining contaminants by adsorption or complex formation, and their chemical state is not altered. The most often used media are GAC, zeolites, and others for the removal of inorganic and organic compounds.

Blowes et al. [43] mentioned that the treatment can be grouped into abiotic reduction and immobilization, biologically mediated reduction and immobilization, and adsorption and precipitation reactions. Accordingly, the reactive materials used in the remediation process undergo one of the following reactions [44; 1]:

1. Chemical precipitation of heavy metal compounds
2. Sorption of inorganic or organic compouds
3. Retardation and biodegradation of organic pollutants
4. Abiotic reduction
5. Biotic reduction

In fact, in many cases, remediation of contaminated groundwater can be achieved by two or more of these processes that take place simultaneously [42].

Modeling of contaminant transport through PRB

In PRBs simulation, three aspects must be considered: the hydrogeologic (groundwater flow) aspect in 1D, 2D, or 3D; geochemical (chemical reactions) aspect; and economic aspect (construction and operational costs) to specify the construction and operational costs [45]. A modelling of PRB can be represented physically or mathematically. A mathematical model is a numerical expression of the conceptual model, which can be either an analytical solution involved in solving differential equations, representing the conceptual model, with appropriate initial and boundary conditions, or numerical solution involved in solving a set of algebraic linear equations, representing the conceptual model, instead of the differential equations used in the previous approach [46]. On the physical side, a model should simulate groundwater flow directly by using a scaled reproduction of the real world. Many researchers have combined physical and numerical simulations to obtain the most feasible representative predictions of PRBs behaviour and response. Most of the studies mentioned in the present review had used 1D pilot model of column test to simulate the processes that occur within the PRBs and utilize them to assess and investigate the real behaviour in short and long terms of these barriers either at a laboratory or field scale. Although the column test is generally adequate to simulate remedy processes, some studies based on floor scale test in simulation treatment processes in PRBs have helped to represent the 2D or 3D flow [41]. Nevertheless, with the aid of computers, solving complex problems numerically becomes easier. Many computer simulation codes are available to solve the PRB problems, and the selection of the desired code is based on many considerations such as availability, applicability, and price. The popular groundwater modelling code is MODFLOW [47], and its latest version is Visual MODFLOW^{R} Flex, which provides solution for controlled equation on finite different method. For integral solution, other codes and modules such as MODPATH, RWLK3D, and MT3D, RWLK3D have been used in conjunction with MODFLOW and marketed as software packages such as GMS (groundwater modelling system), Model Cad, Visual MODFLOW, Groundwater Vistas, Horizontal Flow Barrier (HFB), and ZONEBUDGET. Furthermore, other 2D and 3D models are able to simulate contaminant transport with water flow and in PRBs, such as FRAC3DVS [48], FLOWPATH [49], and FEMWATER [50]. At the Royal Institute of Technology in Stockholm, Sweden (2005), graduate students of Germund Dahlquist developed COMSOL Multiphysics code which can be utilized for groundwater contaminant transport and treatment process [51]. This code is based on finite-element numerical method in the solution of coupled partial differential equations (PDEs) with applications including flow and transport in porous media [52]. Di Natale et al. [53] used a commercial 2D model flow, SEEPTM, in combination with FORTRAN code to describe the groundwater flow and Cd(II) transport through GAC barrier. Bakir [54] used COMSOL for predicting the breakthrough curves in comparison with experimental data for removal of metals in the fixed-bed sorption column with seaweed as reactive material. The results signified that COMSOL was an effective tool for generating models accurately and describing metal biosorption onto biomass for single metal systems. Furthermore, Di Nardo et al. [55] developed a 2D numerical model for describing the transport of tetrachloroethylene (PCE) spilled from a solid waste landfill within groundwater and activated carbon PRB. The results showed that the barrier had a good efficiency because the PCE concentration flowing out of the PRB was always lower than the limits provided in the currently enforced Italian legislation. Moreover, Eljamal et al. [56] developed a 1D numerical model for arsenite transport through ZVI barrier taken the chemical reaction into account. The results of the column tests showed that the adsorption rate of As(V) was faster than that of As(III). Orjuela and Gonza´lez [57] proposed a model with COMSOL Multiphysics to simulate mass transfer through packed column in the bioadsorption process of Cr(VI) in the S-layer of immobilized *Bacillus sphaericus* pellets, whereas Sachdev et al. [58] used the modern computational fluid dynamics (CFD) code COMSOL Multiphysics 4.2a for modeling and simulation of packed bed reactors. A detailed description of the flow behavior and heat transfer aspects within the bed was established and validated with the literature data. Furthermore, Faisal and Hmood [59] developed a 1D model solved numerically by finite difference for description of Cd(II) transport through zeolite barrier. In addition, Faisal et al. [60] used COMSOL Multiphysics 3.5a software for simulating the Zn(II) transport through sandy soil in the presence of ZVI barrier. The experimental and predicted results proved that the barrier was able to restrict Zn(II) migration. Benner et al. [61] evaluated and analyzed the performance of a permeable reactive barrier, designed to remove metals and generate alkalinity by promoting sulfate reduction and metal sulfide precipitation, by means of chemical analysis coupled with geochemical speciation modeling using MINTEQA2 code. This analysis result in that the pore water in the barrier becomes supersaturated with respect to amorphous Fe sulfide in addition to the accumulation of Fe monosulfide precipitates in solid phase with the shifting in the saturation states of carbonate, sulfate, and sulfide minerals. They reported that the dominant changes in water chemistry in the barrier and down-gradient aquifer can be attributed to bacterially mediated sulfate reduction. Weber et al. [62] used an enhanced version of the geochemical simulation code MIN3P to simulate dominating processes in chlorinated hydrocarbons (CHCs) treating ZVI PRBs including geochemical dependency of ZVI reactivity, gas phase formation and a basic formulation of degassing. A laboratory column test experiments with distinct chemical conditions were simulated to parameterize the model. The calibrated model was applied on the field site (i.e. Bernau, Germany) for the prediction of the long-term performance of ZVI-PRB installed to treat the groundwater contaminated with the chlorinated hydrocarbons (CHCs). The results of model of field site demonstrated that temporarily enhanced groundwater carbonate concentrations caused an increase in gas phase formation due to the acceleration of anaerobic iron corrosion. Indraratna et al. [63] prepared a geochemistry model with geohydraulics model that are coupled to simulate the remediation of acidic groundwater using an alkaline permeable reactive barrier (PRB). In this work, a geochemical algorithm using the transition state theory was developed for treating acidic groundwater using recycled concrete filled PRB. A laboratory column test was accomplished to simulate a real one-dimensional reactive flow that occurs in real reactive barrier whose results are used thereafter to assess the numerical model predictions. The developed algorithm calculates the saturation indices (SI) from PRHEEQC software that in turn used with the governing equations that are incorporated into commercial numerical codes, MODFLOW and RT3D. Using this model, chemical clogging due to secondary mineral precipitates was monitored with a good agreement between both laboratory model results and numerical model predictions and it was found that the hydraulic conductivity reduction due to mineral precipitation occurs at the start of permeation and continues until halfway through the testing phase. Other modeling techniques are also used to simulate the processes and performance of PRBs with desired reliability. Heuristic methods are one of these techniques that are used recently worldwide in many environmental modelling policies. Artificial neural networks (ANNs)-based model was developed by Santisukkasaem et al. [64] which enables evaluation of long-term permeability losses that occur in permeable reactive barriers (PRBs) used in groundwater remediation. The results of this model were compared with the multiple regression analysis (MRA) which is a statistical analysis method. MRA-based linear and nonlinear regression model results were used for comparison to assess their performance. The encouraging results lead authors to decide that ANN modeling is a promising tool for the simulation and assessment of the permeability decline in PRBs.

2. CONCLUSION

The new concepts related to sustainable (green) technology and use of waste (by-product) materials in the field of environmental remediation with the assistance of physical and numerical simulation provide considerable and wide horizons for scientific research. PRB is a promising technology, and studies about the possibility of using different reactive gates composed of strong chemicals, zeolites, surfactants, iron, adsorptive substances, organisms, and bioactive materials are still underway. In this study, several sorbents have been described, which are actually used for treating of water contaminated with inorganic and/or organic compounds. Accordingly, extensive studies and extra attempts are required for selecting new waste (by product) reactive materials, determining their properties and behavior in the removal of contaminants from groundwater and, consequently, identifying their appropriateness for use in PRBs.

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CONFLICT OF INTEREST

None

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