Proceedings of the 21st ICSMGE, Vienna, Austria, 14 – 19 June 2026. Pistrol, Adam & Schweiger (eds.)
Published by: ÖGG, Austrian Society for Geomechanics, Salzburg, Austria, ISBN 978-3-9503898-4-5
https://doi.org/10.53243/ICSMGE2026-1197

Performance of multi-layer permeable reactive barriers

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ABSTRACT: A permeable reactive barrier (PRB) consists in the placement of a granular reactive medium in the subsoil in order to intercept a contaminated plume and to remove the contaminants within by means chemical, physical and/or biological mechanisms. The choice of the most suitable reactive medium represents a key point for the correct design of the technology. This choice requires an in-depth knowledge concerning the reactive and hydraulic behavior of the reactive medium in the short and long term. The zero-valent iron (ZVI) is the reactive medium used in over 60 % of PRBs, it can be used as individual material or admixed with inert, porous or reactive granular materials. The main objective of admixing ZVI with other granular materials is to prevent the iron corrosion process from reducing the hydraulic conductivity of the PRB, compromising its functioning. A high dispersion of ZVI particles improves the long-term hydraulic conductivity but can significantly reduce reactivity due to the lower amount of ZVI. In order to find configurations able to optimize the hydraulic and reactive behavior of a PRB, this study illustrates a research activity aimed at evaluating the performance of a two-layer configuration of a ZVI/lapillus granular mixture. The two layers are characterized by a different ZVI content per unit volume. The experiments were carried out by means of column tests with a multi-contaminated solution of copper, nickel and zinc. The results show that a two-layer configuration can improve the long-term hydraulic behavior of a PRB.

KEYWORDS: Environmental geotechnics, groundwater remediation, hydraulic conductivity, granular mixtures.

1 INTRODUCTION

A permeable reactive barrier (PRB) involves placing a reactive and permeable medium into the aquifer downstream of the contamination source to prevent contaminant propagation. A PRB does not provide an immediate corrective solution, but it is designed to manage and eliminate the plume over a long period of time under a natural hydraulic gradient. The "horizontal" configuration, suitable for almost uniform aquifers, is obtained by arranging the reactive medium in a perpendicular direction to the flow of groundwater.

In a funnel and gate configuration, elements with lower permeability (funnels) direct the contaminated flow toward the reactive medium (gate). The funnels cause flow mixing, resulting in lower variability in influent and effluent concentrations and more efficient use of the reactive medium (Bilardi et al. 2023a; Mokif and Faisal, 2023). In the early 1990s, both configurations were thought to operate effectively for several decades without requiring any maintenance; malfunctions were deemed unlikely. In reality, it was observed that a PRB could reduce its reactivity and hydraulic conductivity, necessitating replacement of the reactive medium. Therefore, new configurations, installed primarily in Europe, were developed. These configurations, known as "efficiently controllable" PRBs, are the in-situ vessels, the "Drain-and-Gate" PRBs or significantly modified funnel and gate configuration. In these configurations the barrier is designed to direct the flow to an area that can be effectively monitored and easily accessed for any replacements of the reactive medium (Birke, 2007; Klammler et al., 2010).

Depending on the arrangement of the reactive medium, there are a multi-PRB system and a multi-layer configuration. A multi-PRB system consists of a sequence of PRBs placed one-after-another and each PRB is either filled with the same or different reactive materials based on the target contaminants (Birke, 2007; Singh et al. 2020). While this configuration is designed to tackle a complex contamination, a multi-layer system is designed to avoid reduction in hydraulic conductivity when zero-valent iron (ZVI) is used.

ZVI is a versatile reactive medium supplied by different manufacturers, known and used worldwide in laboratory or in full scale PRB. Although there are examples of a good longevity of ZVI-PRB, there are numerous cases when a significant reduction of the PRB hydraulic conductivity occurred (Bilardi et al. 2023a; Plessl et al., 2023; Sun et al., 2016). This phenomenon is mainly caused by the formation of iron oxides and hydroxides which reduce the porosity and permeability of the PRB (Lawrinenko et al, 2023; Noubactep, 2021.; Ruhl et al., 2014).

A well-established strategy to prevent this phenomenon is mixing ZVI with another granular medium, such as sand or volcanic materials (e.g. pumice, lapillus, zeolites), to separate iron particles and prevent the aggregation of ZVI particles from following their expansion which can cause clogging phenomena (Bilardi et al. 2023a).

The optimum choice of ZVI per unit volume when using pumice or lapillus is strictly dependent on the flow velocity of the groundwater through the PRB, the type of contamination and the initial concentration of contaminants. In particular, by reducing the initial contaminant concentration and/or the flow velocity, the ZVI dispersion rate should increase in order that the contaminant removal does not occur in the first few centimeters of the reactive medium and the risk of hydraulic conductivity reduction is lowered (Bilardi et al., 2019, 2022; Calabrò et al., 2021; Madaffari et al., 2017).

In order to avoid clogging phenomena that occurs at the inlet section of the PRB, a multi-layer configuration was implemented in two full scale PRBs (Gavaskar et al. 2000; Li & Benson, 2010; Morrison, 2003).

2 MULTI-LAYER PRBS

The multi-layer configuration was implemented at Monticello (UT, USA) and at Dover Air Force Base (DE, USA). The configuration consists of two layers: the first containing ZVI mixed with an inert material (i.e. sand or gravel) and the second composed of ZVI only. The scope of the first layer is to preserve the permeability of the reactive zone of the barrier by chemically pre-treating the groundwater. The main characteristics of the two PRBs (i.e. reactive medium, thickness and contaminants) are summarized in Table 1.

At Monticello site (UT, USA), a pre-treatment zone composed of ZVI and gravel was placed upstream of the reactive zone, composed of 100% ZVI and of a third zone composed of crushed gravel with the function of distributing the treated water to the aquifer. Field studies have documented the greatest loss of hydraulic conductivity occurring within the center of the 100% ZVI zone (Bartlett, 2005). Much of the decrease was attributed to precipitation of calcium carbonate and other ZVI corrosion products (Li and Benson, 2010; Morrison, 2003).

At Dover Air Force Base (DE, USA) the pre-treatment zone composed of ZVI and sand precedes the ZVI reactive zone and the exit zone composed of coarse sand. The pre-treatment zone was installed to limit the entry of oxygen into the reactive zone and monitoring after 18 months confirmed its depletion before entering the reactive cell. This indicates that by incorporating a pre-treatment zone upstream the real reactive one may improve barrier longevity (Gavaskar et al., 2000).

Table 1. Main characteristics of multi-layer PRB installed at Monticello (UT, USA) and at Dover Air Force Base (DE, USA).

N° layer	Reactive medium	Thickness (m)	Contaminants	Site
1	ZVI/gravel (13:78)*	0.6	As, Mo Se, U, V	Monticello (UT, USA)
2	ZVI	1.2		
3	gravel	0.6		
1	ZVI/ sand (10:90)*	0.6	Chlorinated solvents	Dover Air Force Base (DE, USA)
2	ZVI	1.2		
3	sand	0.6		

*volumetric ratio

The first layer, or "pre-treatment zone", can be easily installed using different technologies, such as the caisson technology employed at Dover Air Force Base. In this case, a temporary divider was used to separate the pretreatment zone from the rest of the reactive medium during filling (Gavaskar et al. 2000). At the Monticello site, the barrier was built by driving steel sheet piling into the bedrock forming a rectangular box where the native soils were replaced with the reactive media.

The in-situ experiences revealed how a pre-treatment zone does not exclude the occurrence of clogging phenomena over time. For example, in-situ tests at Monticello site revealed that hydraulic conductivity values had remained nearly constant within the gravel/ZVI zone but decreased about two orders of magnitude within the ZVI zone approximately 6 years after installation (Bilardi et al., 2023b).

In order to investigate the long-term behavior of multi-layer barrier systems and try to improve their performance, the research activity, carried out in this study, aimed at evaluating the performance of a two-layer configuration of a ZVI/lapillus granular mixture. The two layers are characterized by a different ZVI content per unit volume. In particular, the ZVI is more dispersed in the first layer in order to reduce the risk of clogging phenomena and to ensure the permeability necessary for groundwater flow. Conversely, the ZVI is less dispersed in the second layer in order to assure greater longevity in terms of reactivity.

The experiments were carried out by means of column tests with a multi-contaminated solution of copper, nickel and zinc.

3 MATERIALS AND METHODS

3.1 *ZVI and lapillus*

The ZVI used (FERBLAST RI 850/3.5, distributed by Pometon S.p.A., Mestre Italy) is mainly made of iron (>99.74%) and impurities include Mn, O, S and C.

Lapillus (distributed by "Società Estrattiva Monterosi s.r.l.", Viterbo, Italy) is a sedimentary pyroclastic material originated from the explosive volcanic activity in the Sabatini Mountains (Italy). It mainly consists of silica (SiO_2, 47% mass) and oxides of various elements (Al_2O_3, 15%; K_2O, 8%; Na_2O, 1%; Fe_2O_3-FeO, 7–8%; MnO, 0.15%; MgO, 5.5% and CaO, 11%). The grain size distribution of lapillus specimen has been selected in function of that of ZVI and taking into consideration the filter design criteria (Moraci et al. 2022). The lapillus grains were washed, the retained grains in sieve No. 40 (> 0.42 mm) and the passing to sieve No. 200 (< 0.074 mm) were discarded in order to obtain a grain size distribution more similar to that of ZVI.

The coefficient of uniformity (U) the mean grain size (d_{50}), the particle density of ZVI and the apparent particle density of lapillus are summarized in Table 2.

Table 2. Main characteristic of ZVI and lapillus.

Reactive medium	d_{50} (mm)	U (-)	ρ (g/cm^3)
ZVI	0.5	2	7.87
Lapillus	0.4	3.2	2.2

3.2 *Column tests*

Laboratory scale polymethyl methacrylate (Plexiglas) columns with an internal diameter of 5 ± 0.1 cm and height equal to 50 cm, were used in this research. The columns were filled up to about 28 cm and the empty area of the columns was filled with quartz gravel. Column tests were carried out using a multi-channel precision peristaltic pump (Watson Marlow 205S) in up-flow mode under constant flow rate of 0.5 ml/min (Figure 1).

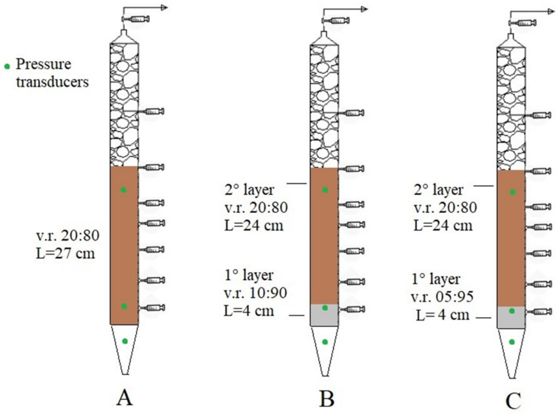

Figure 1. Schematic diagram of column test apparatus.

The contaminated solution was prepared by diluting Copper Nitrate (Copper(II) nitrate hydrate, purity > 99%; Sigma-Aldrich), Nickel Nitrate (Nickel(II) nitrate hexahydrate, purity > 99 %; Sigma-Aldrich) and Zinc Nitrate (Zinc(II) nitrate hexahydrate, purity > 99 %; Sigma-Aldrich) in distilled water in order to obtain a concentration of about 18 mg/l for each metal. The aqueous samples withdrawn from sampling ports located at distance of 3, 5, 8, 13, 18, 23 and 28 cm from inlet were analyzing using Inductively Coupled Plasma – Optical Emission Spectroscopy (ICP-OES - Perkin Elmer Optima 8000).

The column was equipped with pressure transducers (TE Connectivity US300, pressure range of 0 – 200 kPa, ± 0.1 % accuracy) to study the hydraulic behavior. They are located before the column inlet, and at 3 and 25 cm from column inlet (Figure 1) in order to determine the hydraulic conductivity in a first (0-3 cm) and a second layer (3-25 cm).

Three column tests were performed. One column was filled with a single layer of the ZVI/lapillus mixture at a volumetric ratio (v.r.) equal to 20:80 (Column A).

The second column was filled with a first layer of the ZVI/lapillus mixture at v.r. of 10:90 and with a second layer of the ZVI/lapillus mixture at v.r. of 20:80 (Column B).

The third column was filled with a first layer of the ZVI/lapillus mixture at v.r. of 05:95 and with a second layer of the ZVI/lapillus mixture at v.r. of 20:80 (Column C).

The A column test was interrupted after 131 days due to the reduction in the hydraulic conductivity which was not compatible with the imposed flow rate. The B and C column tests were interrupted after 265 and 330 days, respectively, due to exhaustion of the reactivity.

4 RESULTS AND DISCUSSION

The event that could affect the lifetime (or longevity) of the reactive medium is the loss of its reactivity or hydraulic conductivity. While the reduction in reactivity can be managed with the possible replacement of the reactive medium (if configurations that allow its replacement are used), a reduction in hydraulic conductivity due to iron corrosion processes creates the aggregation/cementation among ZVI particles and a more difficult replacement of the material. The results of the three column tests will be discussed by examining the hydraulic and reactive behavior of the three configurations.

4.1 *Hydraulic behavior*

The hydraulic behavior of first (0-3 cm) and second layer (3-25 cm) of the three columns is illustrated in Figure 2. It shows the hydraulic conductivity calculated at time t (k(t)) divided by the initial value (k_0) as a function of time.

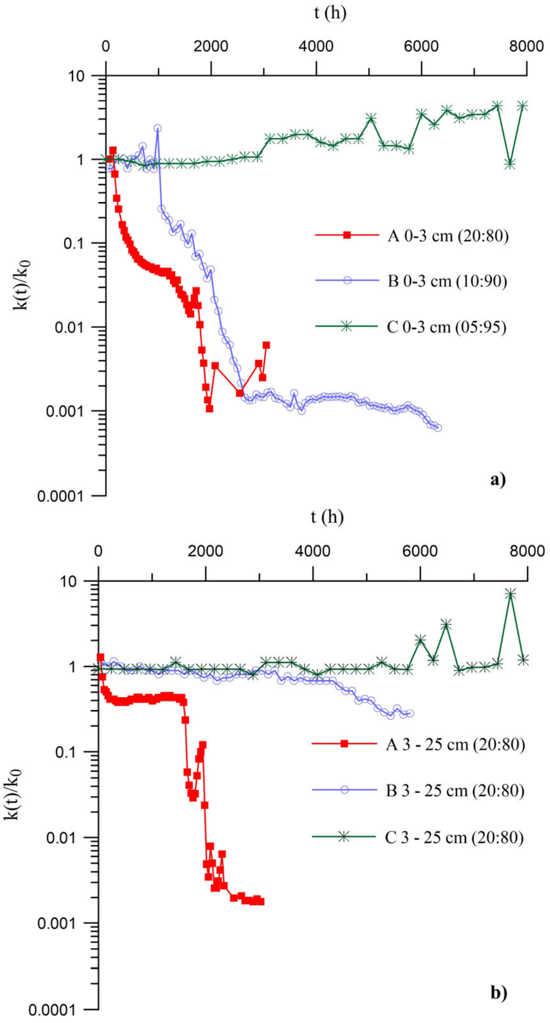

Figure 2. Normalized hydraulic conductivity profile of a) the first (0-3 cm) and b) second (3-25 cm) layer of the reactive media contained in columns A, B and C.

Column A shows the lowest longevity because a rapid reduction in the hydraulic conductivity starting from the column inlet is observed (Figure 2a).

The hydraulic conductivity profile of the first layer of column B is almost the same as that observed in column A, but the reduction in the hydraulic conductivity occurs later due to the lower iron content per unit volume (Figure 2a). The strong similarity of the hydraulic conductivity profile of the first layer of columns B and A suggests a very precise kinetics of iron corrosion. While, for column C, the hydraulic conductivity remains constant up to 3000 hours and subsequently a slight increase is observed, the latter probably due to gas venting or to the formation and removal of solids precipitates.

The hydraulic conductivity of the second layer (Figure 2b), decreases after 1600 hours for column A, slightly decreases after 4200 hours for column test B, whereas it remains constant for column test C.

As observed in Column B, the "pretreatment" layer improved long-term hydraulic behavior compared to Column A because it delayed clogging near the inlet section. However, it should be emphasized that the presence of the pretreatment zone did not prevent clogging of the reactive medium. Meanwhile, as observed in Column C, the "pretreatment" layer not only was not subject to clogging but also preserved the hydraulic conductivity of the subsequent layer.

4.2 *Reactive behavior*

The reactive behavior derived from the three column tests can be studied in terms of the normalized concentrations of contaminants at a given sampling port (or a given thickness of the reactive medium) as a function of the duration of the test. From this curve, it is possible to identify the breakthrough time (T_b) that is the time at which a clear and rapid increase in the contaminant concentration is observed. It is also the maximum time within which the system is able to ensure that the remediation objective is achieved.

For copper, the breakthrough time, or the time the system is able to keep the concentration of the pollutant below the Italian regulatory limit (i.e. 1 mg/l), at 3 cm of the reactive medium thickness is similar for columns B (i.e. 3696 hours) and C (i.e. 3528 hours). While the breakthrough time was never reached for column A which had a shorter duration due to the excessive reduction in hydraulic conductivity.

The concentration of copper remains below 1 mg/l in all sampling ports located after 3 cm for the three column tests.

Unlike copper, nickel and zinc are removed with greater difficulty from the three configurations of the reactive medium. For these two contaminants, the breakthrough times at different reactive medium thicknesses are shown for nickel (Figure 3a) and zinc (Figure 3b) for the three tests. When the Italian regulatory limit for nickel (i.e. 0.02 mg/L) or zinc (i.e. 3 mg/l) was exceeded in the first sampling ports starting from the beginning of the test, it was not possible to observe the breakthrough time.

As shown in Figure 3a breakthrough times for nickel do not always increase as the thickness increases, suggesting the existence of an optimal thickness of the PRB for nickel removal. In particular, neglecting the hydraulic behavior (i.e. assuming that the hydraulic conductivity remains constant over time), the optimal thickness represents the minimum thickness value for which no further increase in breakthrough time is observed. The optimal thickness, obtained on the x-axis of Figure 3a, is equal to 13, 18, and 23 cm for columns A, B, and C, respectively. Defining an optimal thickness for the treatment layers translates into an optimization of the resources required for the construction of a PRB in terms of quantity and cost of materials, as well as in terms of the removal efficiency of individual contaminants.

The presence of an optimal thickness in the case of nickel removal suggests that when ZVI is crossed by uncontaminated water, it is in any case corroded and this leads to a reduction in its reactivity over time. This behavior was further explored by the Authors in Bilardi et al., 2025.

For zinc (Figure 3b), the breakthrough time linearly increases with the reactive medium thickness and data can be fitted with a linear equation having $R^2 > 0.87$.

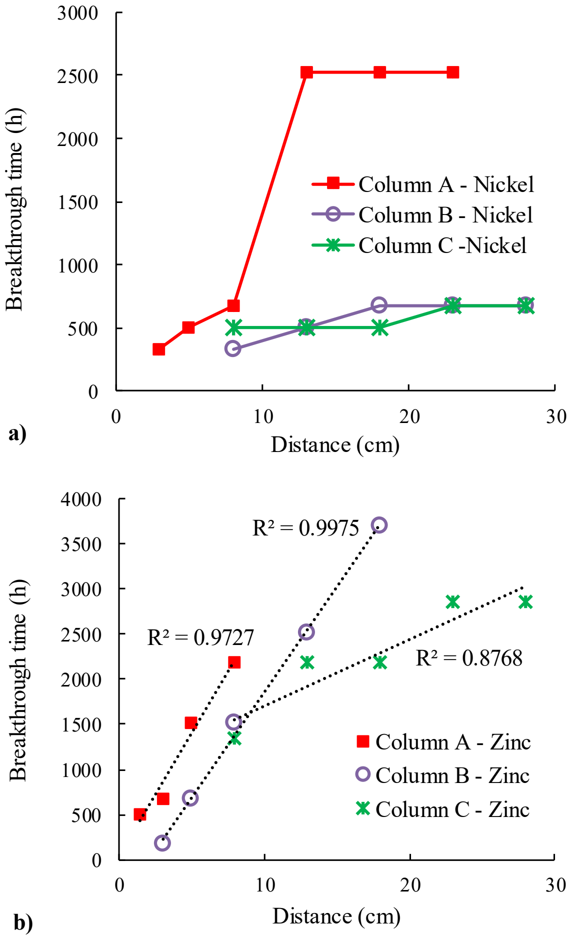

Figure 3. Breakthrough times (h) vs. column distance (cm) for a) nickel and b) zinc.

4.3 *Longevity*

As previously mentioned, the events that could affect the reactive medium longevity are the reactivity or the hydraulic conductivity reduction.

It is important to note that the reactivity and hydraulic conductivity of the reactive medium are closely related. The better removal of the contaminants observed in Column A (see in Figure 3 the higher breakthrough values of column A compared to columns B and C with the same thickness) was attributed to the formation of iron corrosion products, which contribute to the removal of nickel and zinc by co-precipitation (i.e. dissolved species are mechanically entrapped in the matrix of oxyhydroxides during their precipitation) and adsorption onto available iron oxides. Whereas the constant value of the hydraulic conductivity and the complete exhaustion towards nickel and zinc of the first layer of column C in the long term suggests a lower formation of iron corrosion products and the consequent reduction of the heavy metal removal capacity.

Regarding adsorption, contaminants can be adsorbed at the surface of solid iron corrosion products, such as $Fe(OH)_3$; this mechanism involves aged Fe oxyhydroxides, whereas co-precipitation involves nascent Fe oxyhydroxides. The relation between the reactive behaviour showed in Figure 3a (column A) and the hydraulic reduction showed in Figure 2a, suggests that nickel removal is removed by nascent Fe oxyhydroxides. These iron corrosion products are therefore responsible for the removal of nickel and the consequent reduction of hydraulic conductivity (Bilardi et al., 2025).

The events affecting longevity of the three column tests are summarized in Table 3 for the three column tests. Hydraulic longevity was identified with the reduction of hydraulic conductivity by an order of magnitude, while reactivity longevity was identified with the achievement of breakthrough time. In particular, regarding the hydraulic behavior, it was assumed that a reduction in the hydraulic conductivity of one order of magnitude is not compatible with the correct operation of a PRB.

As regards reactive behavior, the lifetime was considered upon reaching the maximum breakthrough time (with reference to the Italian regulatory limit). The events were identified both in terms of distance from the column inlet (or thickness of the reactive medium) and in terms of time during which the events were observed.

Table 3. Time (T), and distance from column inlet (L) in which a hydraulic conductivity reduction of one order of magnitude or the contaminant breakthrough are observed.

Test	T (h)	L (cm)	Event
A	400	3	Hydraulic conductivity reduction of one order of magnitude
	2184	8	Zinc breakthrough
	2520	13	Nickel breakthrough
	3144	3	Copper breakthrough not observed until end of the test
B	1500	3	Hydraulic conductivity reduction of one order of magnitude
	3696	3	Copper breakthrough
	3696	18	Zinc breakthrough
	672	18	Nickel breakthrough
C	3528	3	Copper breakthrough
	2856	23	Zinc breakthrough
	672	23	Nickel breakthrough
	7932	3	Hydraulic conductivity remained constant until the end of the test

According to the hydraulic behavior, Column A would have required the substitution of the reactive medium after about 400 hours. According to the reactive behavior, the maximum longevity for columns B and C is 672 hours, which is the breakthrough time obtained for nickel.

From a comparison among the three tests, it emerges that the presence of the pretreatment zone increases barrier longevity by 68 %.

The lifetime of column C is linked to nickel removal and to the regulatory limit that must be reached, but the results highlighted in Figure 3a show that an increase in the PRB thickness does not allow an increase in its longevity. In the absence of nickel, it is reasonable to assume that the longevity of column C is related to the removal of zinc and that it can be increased by increasing the thickness of the PRB (see design curves in Figure 3b).

Figure 4. ZVI/lapillus mixture inside column A after disassembly.

5 CONCLUSIONS

A multi-layer PRB configuration is designed to avoid reduction in hydraulic conductivity in the inlet section of the PRB when zero-valent iron (ZVI) is used. The in-situ experiences of two full scale PRBs revealed how a pre-treatment zone does not exclude the occurrence of clogging phenomena over time.

In order to improve the hydraulic and reactive behavior of multi-layer barrier systems, the research activity, carried out in this study, aimed at evaluating the performance of a two-layer configuration of a ZVI/lapillus granular mixture.

Three column tests were performed; a column was filled with a single layer of the 20:80 ZVI/lapillus mixture (Column A), two columns were filled with an initial 4-cm-thick layer of a ZVI/lapillus granular mixture with a v.r. of 10:90 (Column B) or 5:95 (Column C) and a second layer with a v.r. equal to 20:80. The columns were permeated with a multi-contaminated solution of copper, nickel and zinc.

Column A showed the lowest longevity due to a rapid reduction in the hydraulic conductivity starting at the inlet section of the column and propagating up to the second layer. Due to the higher iron content, the loss of reactivity of the reactive medium contained in column A, was slower than the reactive media contained in columns C and B. Therefore, the lower longevity of column A was linked to the reduction in hydraulic conductivity.

Once the test was completed, during the dismantling phase of column A, it was noted that the iron corrosion products had caused aggregation/cementation of the reactive medium near the inlet section, making its extraction difficult (Figure 4). This phenomenon highlights the importance of appropriately choosing the degree of dispersion of the zero-valent iron to avoid not only the premature reduction of hydraulic conductivity within a PRB, but also the greater difficulty in replacing the cemented reactive medium, which must be crushed before removal.

The presence of the double layer in columns B and C allowed to avoid the drastic reduction in hydraulic conductivity observed in the single layer contained in column A. For columns B and C, due to the simultaneous presence of copper, nickel and zinc and due to the greater difficulty in removing nickel, the longevity of the two mixtures was linked to the removal of nickel and an increase in PRB longevity of 68 % compared to column A was calculated.

In the three columns, copper and zinc were removed for longer period than nickel and the breakthrough time generally increased with barrier thickness. For nickel, an optimal thickness of the PRB equal to 13, 18 and 23 cm for column A, B, and C respectively was identified. In this case, an increase in the thickness of the reactive medium did not correspond to a significant removal of the contaminant. In cases where breakthrough times increase linearly with reactive medium thickness and hydraulic conductivity remains constant over time, PRB longevity can be increased by increasing PRB thickness.

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