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Review

Permeable Concrete Barriers to Control Water Pollution: A Review

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Abstract: Permeable concrete is a class of materials that has long been tested and implemented to control water pollution. Its application in low-impact development practices has proved its efficiency in mitigating some of the impacts of urbanization on the environment, including urban heat islands, attenuation of flashfloods, and reduction of transportation-related noise. Additionally, several research efforts have been directed at the dissemination of these materials for controlling pollution via their use as permeable reactive barriers, as well as their use in the treatment of waste water and water purification. This work is focused on the potential use of these materials as permeable reactive barriers to remediate ground water and treat acid mine drainage. In this respect, advances in material selection and their proportions in the mix design of conventional and innovative permeable concrete are presented. An overview of the available characterization techniques to evaluate the rheology of the paste, hydraulic, mechanical, durability, and pollutant removal performances of the hardened material are presented and their features are summarized. An overview of permeable reactive barrier technology is provided, recent research on the application of permeable concrete technology is analyzed, and gaps and recommendations for future research directions in this field are identified. The optimization of the mix design of permeable reactive concrete barriers is recommended to be directed in a way that balances the performance measures and the durability of the barrier over its service life. As these materials are proposed to control water pollution, there is a need to ensure that this practice has minimal environmental impacts on the affected environment. This can be achieved by considering the analysis of the alkaline plume attenuation in the downstream environment.

Keywords: permeable concrete; mix design; preamble reactive barriers; acid mine drainage; remediation; pollutant removal

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1. Introduction

All life forms on our planet need water to function properly; this need is attributed to the chemical composition of this compound, in which water is a polar covalent molecule, which supports its use as a solvent for many nutrients and, subsequently, their transport throughout living organisms. In addition, water is the main ingredient used to form different fluids that are needed to protect and lubricate biological tissues; it helps in controlling the body's temperature and acts as a medium for the chemical reactions of enzymes [1]. Nearly 97.5% of this natural compound is saline, and only 0.3% is surface fresh liquid water that is distributed in lakes, rivers, swamps, soil moisture, and the atmosphere. The limited amount of easily accessible fresh water and its necessity for life's continuation were the main drivers of the identification of the provision of clean water and sanitation as one of the Sustainable Development Goals (Goal 6). In addition, the availability of freshwater of acceptable quality is the driving force to achieve Goals 2, 3, 8, and 9 (i.e., zero hunger; good health and well-being; decent work and economic growth; and industry, innovation, and infrastructure) [2]. Moreover, preventing and controlling pollution that can spread because of the improper management of solid wastes and wastewater is a key aspect of achieving Goals 11, 14, 15, and 17 (i.e., sustainable cities and communities; life below water; life on Earth; and partnership for goals) [2]. Hence, several efforts are being carried out worldwide that aim to ensure the sustainable provision of water with acceptable quality by preventing and controlling the pollution of surface and ground waters. These efforts focus on the investigation of new materials and/or systems to evaluate their potential implementation in preventing and controlling water pollution, where passive engineering barriers play an important role in this field via the application of permeable barriers that can remove pollutants from different types of water.

Conventional cement-based materials is a class of materials that have been used for decades to support human civilization, and they depend on the use of a hydraulic binder to bond fragmented particles. Upon the hydration of the binder, new hardened materials with enhanced physical and mechanical properties are formed. The properties of the fresh and hardened materials can be tolerated by changing the mix design, i.e., changing the additives and admixtures, water-to-cement ratio, finesse, and type of cement used [3]. The hydration reaction products include major hydration phases (i.e., calcium silicate hydrate (C-S-H), portlandite (CH), and ettringite (AFt)); minor hydration phases (e.g., monosulfate and hydrogarnet); and the heat of hydration. The proportion of these phases is dependent on the mix design and the curing conditions [3–6]. These hydrated phases determine the properties of the hardened cement-based materials and the evolution of these properties overtime [6,7]. Conventional cement-based materials are heavily used in the construction sector and in environmental protection and restoration. In particular, the application of these materials in environmental protection encompass many fields, including rock repair and enforcement, the design of disposal facilities for hazardous and radioactive wastes, the stabilization and solidification of hazardous and radioactive contaminants, and water and wastewater treatment [8–21]. This wide range of applications is supported by the low cost of these materials and their availability, the accumulated knowledge and experience from operating these materials, and the ease of engineering the hardened materials that ensures the attainment of the required performances.

Permeable concrete, also known as pervious concrete, is a subclass of cement-based materials that is tailored to have a characteristic interconnected and tortuous macroporous structure by eliminating the use of fine aggregates [22–25]. This subclass of cement-based materials is receiving increased interest in research to enhance its application in the prevention and control of water pollution due to its following characteristics [5,8,16–20,22,25–31]:

- Input materials that are available and produced through standardized production methods. Additionally, they are low cost, which reduces the cost of pollution prevention and control practices;
- Preparation/construction requiring the use of simple devices (e.g., mixers at room temperature) that have a record of long use and experience;
- Characteristic porous structure allowing for its use as a filter and the passage of water without the need to enforce this passage, in addition to having a high specific surface area that enhances the sorption of containments;
- The presence of the amorphous and crystalline hydration phases providing sites for the chemical and physical entrapment of different anions and cations;
- The hydration of the cement creates highly alkaline conditions leading to the precipitation of most of the metallic contaminants;
- Ecofriendly permeable concrete has the potential to reduce the material and energy footprints of these water pollution prevention and control practices, as well as to reduce greenhouse gas emissions.

The interest of the scientific community in studying permeable concrete, its durability, and its use as barriers can be recognized by analyzing the number of publications indexed in the Scopus database (Figure 1). This analysis was conducted by constraining the search in the database using the Boolean operator "AND", whereby the total number of documents that mentioned the words "permeable" and "concrete", starting from 1944 to 15 September 2023, was 2056 documents. The restriction of the search to the durability of permeable concrete and permeable concrete barriers reduced the total number of documents to 411 and 121, respectively. Figure 1a,b visualize the variation of the annual published works over the past twenty years and their distribution based on the type of document. It is clear that the scientific interest in this research field increased over time, declined in 2014, then increased again, only to decline again in 2022. It should be noted that the records in 2023 are not final, as the search was conducted in the fourth quarter of the year. The average ratios between the number of publications that addressed permeable concrete durability and permeable concrete barriers relative to those that addressed permeable concrete are 0.21 and 0.09, respectively. This indicates that durability studies on these materials are addressed in one-fifth of the publications, and their use as barriers represent approximately10%. Most of the published works are research articles published in journals, as indicated in Figure 1b, in which the average ratios of the publications, independent of the type of document, are fairly constant. The geographical distributions of the published works are illustrated in Figure 1c–e; for this part of the analysis, the national contributions that had less than 1% of the total publications were summed with the undefined category under the name "Other". The United States and China represent the major contributors in the "permeable AND concrete" (Figure 1c) and "permeable AND concrete AND barrier" (Figure 1d) research topics, whereas India and China are the main contributors in the topic "permeable AND concrete AND durability" (Figure 1e). Through applications of permeable concrete materials in low-impact development practices, their efficiency in mitigating some urbanization impacts on the environment has been proven, e.g., urban heat islands, attenuation of flashfloods, and reduction of transportation-related noise [22,32–35]. Some recent review papers have been published that summarize different aspects of these applications, including the state-of-the-art development of these materials and their characteristics, performance, application, and sustainability [32–35]. In addition, research efforts have been directed at disseminating the use of permeable concrete not only for preventing water pollution through their use in low-impact development practices but also for controlling pollution via their use as permeable reactive barriers and their use in the treatment of waste water and water purification [18,21,24,25,30,31,36,37]. These applications for controlling water pollution need to be assessed in light of the acquired knowledge from the application of these materials in low-impact development practices over more than three decades and recently published research in these areas. The aim of this work is to assess the application of permeable concrete materials in the control of water pollution through their application as permeable reactive barriers. In this respect, an overview of both conventional and innovative permeable concrete mix design and their characterization is presented. Permeable reactive barrier technology is reviewed, recent studies on the application of permeable concrete with this technology are analyzed, and gaps in this field are identified.

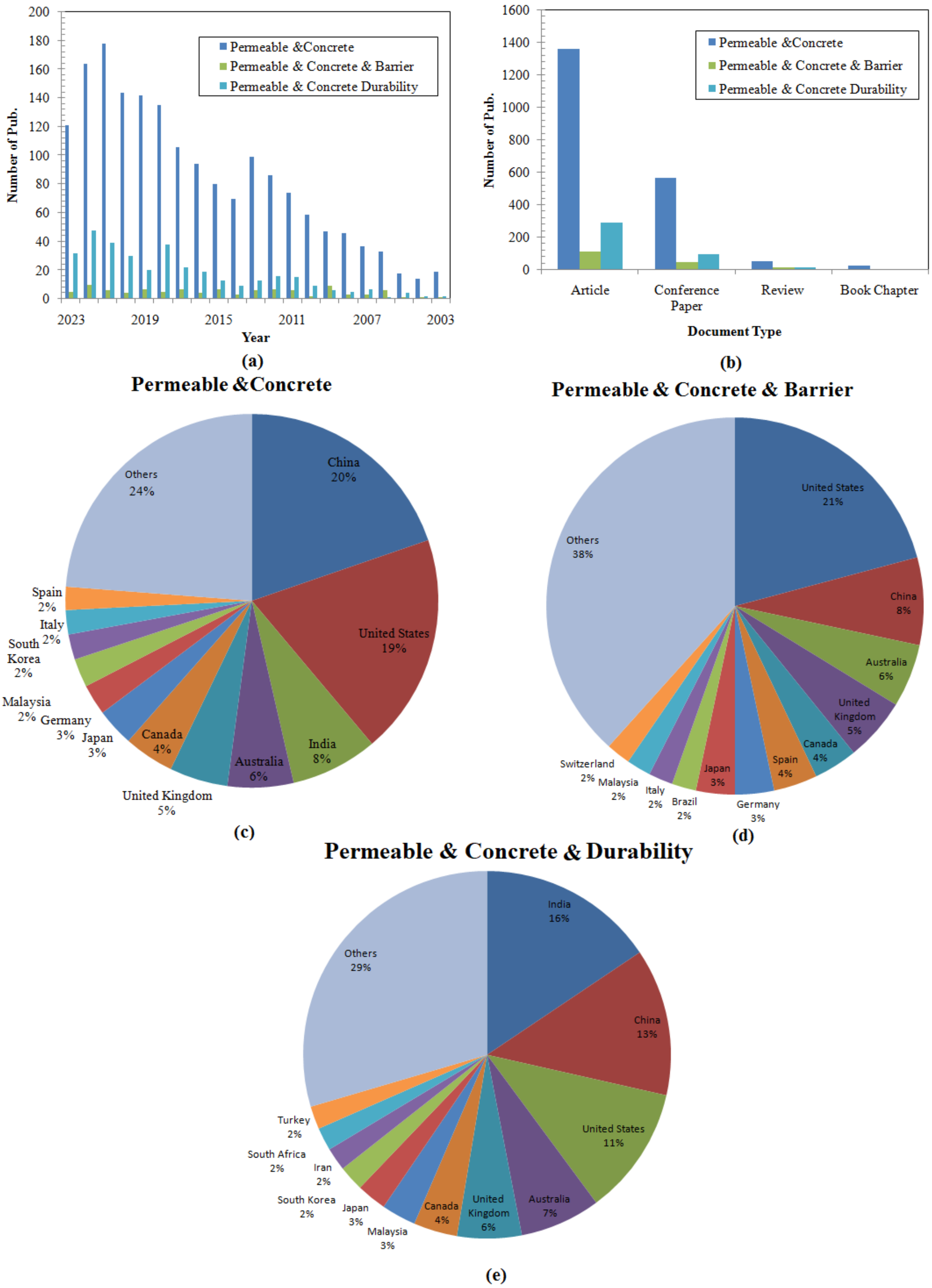


Figure 1. Bibliometric data analysis: (**a**) annual variation of the published works; (**b**) type of the published documents; (**c**–**e**) geographical distribution of the published works.

2. Permeable Concrete Mix Design

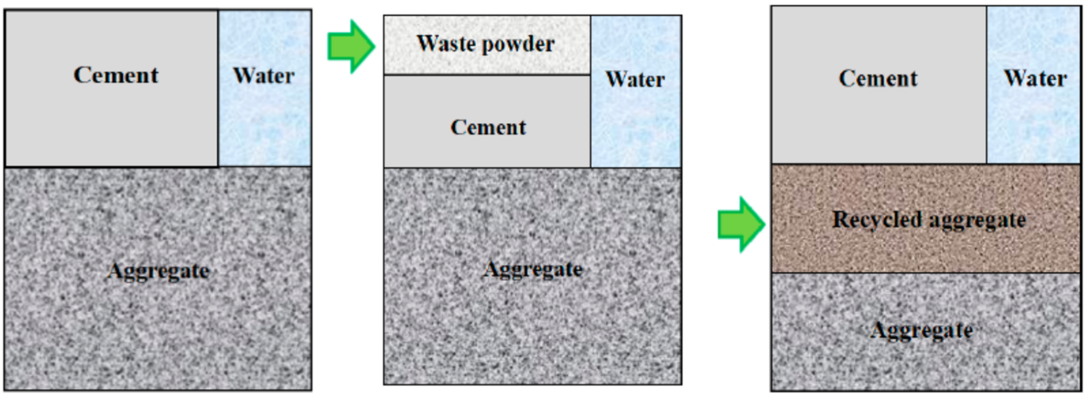
In general, the design of any cement-based material mix is dependent on the identified function of the hardened material. Depending on the application of these materials, processing and performance requirements should be defined [38]. Most of the review articles that addressed the preparation of permeable concrete only focused on the performance requirements of the produced material (i.e., mechanical, hydraulic, and durability requirements) with little focus on their processing requirements [26,32–34,39–43]. Currently, there is only one available valid standard test for this class of cement-based materials that address the measurements of the infiltration rate through the in-place barrier: ASTMC 1701/C1707M-17a. This test is a performance test and is related to the application of the permeable concrete as pavement. Both conventional and innovative cement-based materials were tested for their application in the production of permeable concrete, in which alkali-activated materials were recently investigated for this purpose [26,30,39–43]. In this section, advances in material selection and their proportion in the mix design of conventional and innovative permeable concrete are presented.

2.1. Conventional Permeable Concrete Mix Design

Conventional permeable concrete is manufactured using ordinary Portland cement (OPC), where type 1 is the most used in research investigations, with coarse additives, little or no fine aggregate, admixtures, and water. The hardened materials are designed to have interconnected pores (2–8 mm in size), and dead-end and capillary pores [32]. They should contain 15–25% voids with an acceptable compressive strength in the range of 2.8–28 MPa [44]. In this respect, OPC is used to provide a coating layer on the coarse aggregate, which is required to bond the aggregates, sustain certain mechanical and hydraulic properties, and ensure its durability over its service life. During the optimization of the mix design, the following general requirements maybe considered:

- A water-to-cement (w/c) ratio optimized in the range 0.25–0.45.Increasing the water ratio is an advantage for the formation of porous materials, but it will reduce the thickness of the final cement coat on the aggregates, as the paste will have a high flowability [23,32,44,45];
- A coarse-aggregate-to-cement ratio in the range of 4:1 to 6:1, in which the volume of aggregate in the hardened materials occupy 50–65% [23]. The grading of the aggregates should be optimized to control the void ratio of the hardened product, whereby single-sized coarse aggregate or narrow-grading coarse aggregate (e.g., between 9.5 and 19 mm) can be used [44,46];
- The use of admixtures in the permeable concrete to control the workability of the paste without greatly increasing the water content, retarding or accelerating the hydration process, and improving the freeze–thaw durability [44,46].

Alternative materials have been investigated to replace cement and/or aggregates to reduce the environmental impacts associated with their use and to improve the performance of hardened permeable concrete [33]. In this respect, natural materials and industrial and agricultural wastes have been investigated for their applications as supplementary cementitious materials (SCMs) and/or replacement of aggregates. Figure 2 represents the mix design of permeable concrete with no replacement (Figure 2a) and the partial replacement of cement (Figure 2b) and aggregates (Figure 2c) [47]. Additionally, several materials have been tested for their application as admixture, fibers, or functional materials to enhance certain properties of the paste/fresh and hardened permeable concrete. Table 1 lists some examples of different materials that have been investigated for their application as alternative SCMs, aggregates, and fibers [11,20,25,31,37,48–95]. Recent review articles were devoted to addressing the effect of solid waste reuse [47] and the use of functional materials on the properties of hardened permeable concrete [48,49]. Several research papers investigated the use of different fibers to reinforce permeable concrete, in which natural fibers, industrial wastes, and organic and inorganic chemicals were investigated [51,54,57,67,75,78,81,85,86,90]. Additionally, some research papers employed several admixtures, i.e., superplasticizer and water-reducing and air-entrapping admixtures to control the fresh and hardened permeable concrete properties [25,52,61,94,96]. Finally, functional materials (i.e., chemicals)have been investigated for their effects on enhancing the mechanical properties and pollutant-removal performance of permeable concrete, e.g., reduced graphene [36], nano-iron oxide [52,59,62], and nano-titanium oxide [63]. Limited research has proposed the coating of aggregates to improve the fresh paste concentration at the aggregates' joints [73] or to enhance their mechanical, sorption, and leaching performance [93]. It should be noted that coating the aggregates was reported to be a nonstandard practice for concrete producers and can increase the overall price of the final produced materials [97].



(**a**) No replacement (**b**) Cement replacement (**c**) Aggregate replacement

Figure 2. Schematic presentation of the use of alternative materials to replace the cement and aggregated in conventional cement mix design (copyrighted [47]).

Table 1. Alternative SCMs, aggregates, and fibers investigated to prepare permeable concrete.

	SCMs		Aggregates		Fibers	
	Material	**Refs.**	**Material**	**Refs.**	**Material**	**Refs.**
Natural materials	Metakaolin (MK)	[53,61]	Basalt	[51,56,63,79]	Basalt fibers	[78,86]
	Nano-clay	[81,90]	Lignite	[50]	Jute fibers	[54]
			Limestone	[20,51,52,59,66,71,80]		
			Granite	[31,84,88,92]		
			Pumice	[51,90]		
			Zeolite	[20]		
Industrial wastes	Fly ash	[11,25,31,52,53,55–60,89,94]	Recycled concrete aggregate	[60,64,66,70–73,81,90]	Plastic fibers	[81,90]
	Blast furnace slag	[25,54,64,66–69]	Recycled brick aggregate	[74–77]	Fine saw dust	[57]
	Volcanic ash	[65]	Iron slag	[20]		
	Copper slag	[64]	Steel slag	[79]		
	Silica fume	[25,56,61,80–82,89]	Crumb rubber	[82]		
	Calcium carbide	[84]				
	Sugarcane bagasse ash	[95]				
Agro-waste	Biochar	[37,83]	Petioles from Sterculia foetida plant	[91]	N-A *	
	Rice husk	[84,85]	Oil palm kernel shell	[92]		

Table 1. *Cont.*

	SCMs		Aggregates		Fibers	
	Material	**Refs.**	**Material**	**Refs.**	**Material**	**Refs.**
Chemicals	Nano silica	[59,87,88]	N-A *		Steel and steel wool fibers	[51,75,81,85,86,90]
					Polypropylene	[51,67,75]
					Polyphenylene sulfide	[85]
					Glass	[85]

Note: * N-A not available

2.2. Innovative Permeable Concrete Mix Design

Permeable concretes based on the use of innovative cements has also been prepared and investigated, and this includes geopolymers, magnesium phosphate cement, and calcium sulfoaluminate cement [26,30,39–43,98–102]. Most of these research efforts have focused on the use of geopolymers with limited investigation of the use of magnesium phosphate cements, and only one paper, to the knowledge of the authors, proposed a combination of ordinary Portland cement and calcium sulfoaluminate cement [102].

2.2.1. Permeable Geopolymers Concrete

Geopolymer is a relatively new class of alkali-activated cement-based materials that is prepared using aluminosilicate source(s) and alkali-activating solution(s). Natural minerals, industrial wastes, and chemicals can be used to prepare geopolymers [41,103,104]. Metakaoline (MK) is the most used natural aluminosilicate source, whereas fly ash (FA), slag, red mud, and biomass fly ash are the most used waste materials for the same purpose [39,41,105–108]. Other natural materials have been used in the preparation of geopolymers, including bentonite and feldspar, but to the knowledge of the authors these natural aluminosilicate sources have not been investigated to prepare permeable concrete. Different alkali-activating solutions are used for the preparation of geopolymers, including NaOH, KOH, water glass, or a combination of them [39,41,103]. The main binding phase in hardened geopolymers is aluminosilicate gel, and it is classified based on its Si/Al ratio into poly(si-alate) (Si/Al = 1) and poly(sialatesiloxo) (Si/Al = 2), and poly(sialate-disiloxo) (Si/Al = 3) [103,104]. As in any cement-based materials, the final the properties of the hardened geopolymers are strongly dependent on their mix design and curing conditions, in which the value of the silicon-to-alumina ratio effectively changes the composition of the hardened geopolymer as follow [6,103,105,106]:

- Crystalline zeolite is formed in geopolymers at $Si/Al < 1$;
- Geopolymers of reduced porosity are formed at $1 < Si/Al < 2$;
- The porosity of geopolymers are dependent on the solubility of the Si source at $2 < Si/Al$.

Permeable geopolymers can be prepared using different methods to produce a wide variety of materials with distinctive porous characteristics, and these methods can be categorized into the self-forming method (SFM; Figure 3A), direct foaming method (DFM; Figure 3B), adding filler method (AFM; Figure 3C), and particle stacking method (PSM; Figure 3D) [41]. Table 2 lists the features of these methods, the general pore structure characteristics of the hardened geopolymers, and their applications.

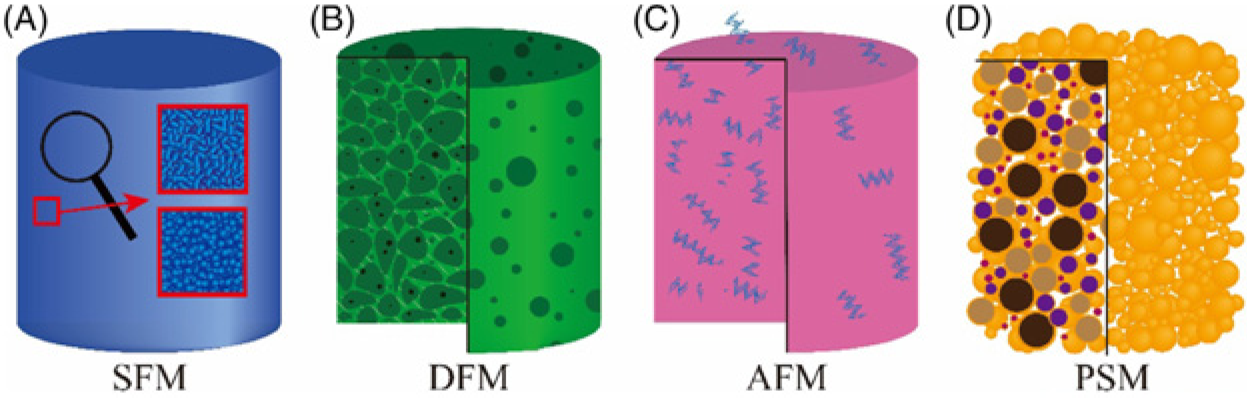


Figure 3. Schematic diagram and photos of the preparation method of porous geopolymer (copy righted from [41]).

Table 2. Features of the porous geopolymer preparation methods [41].

Method	Preparation	Pore Characteristics	Applications
SFM	The porous structure is self-formed without the addition of any material	Porous structure cannot be observed directly Pores are small	Sorption Membrane filtration
DFM	Foaming agents, surfactants, or both are used	Large pore diameter that can directly be observed Noted circular pores on the surface and irregular internal bubbles pores	Building insulation Building lightweight
AFM	Porous filler or materials are added	Reflects the filler's pore structure rather than that between the filler and geopolymer	Various applications including Adsorption Ultralight weight Building insulation
PSM	Bonding of the aggregates	The pore structure is formed in geopolymers or between the aggregates and geopolymers The pore diameter is related to the aggregate size Pores are observed directly	Porous pavement Permeable concrete

2.2.2. Permeable Magnesium Phosphate Concrete

Magnesium phosphate cements are formed through reactions between MgO and phosphates to form a magnesium phosphate salt with cementitious properties [5,6,98,100,101]. Magnesium phosphate cement is characterized by its fast hardening, near-neutral pH, low water demand, high adhesive strength to metals and concrete and high bending and compressive strength [5,6]. Acid phosphate anions (e.g., mono-potassium di-hydrogen phosphate, mono-sodium di-hydrogen phosphate, mono-ammonium di-hydrogen phosphate, and di-ammonium hydrogen phosphate) are used as phosphate sources or aqueous phosphoric acid [5,6,98,100,101,109,110]. The main final phase in hardened magnesium phosphate cement is struvite, where the ratio between the Mg and PO_4 largely affects the produced hardened cement's properties. At a low Mg/PO_4 ratio (<4), the crystallization of the struvite is enhanced and denser microstructure is attained. Because of its fast setting, a retarder is usually used to control the rate of the reaction, e.g., sodium tripolyphosphate, glacial acetic acid, and boric acid [5,98,100,101]. The limited research on the preparation of permeable magnesium phosphate concrete has tailored the mix design of this paste to include an aluminosilicate source (e.g., FA, MK, granulated blast furnace slag [98,101], steel slag [98], and crushed stone [101]) as aggregates and borax as retarder.

2.3. Permeable Concrete Mix Design: Future Prospects

As mentioned in the previous subsections, several research efforts have been directed toward the incorporation of natural materials as SCMs, aggregates, and fibers in conventional permeable concrete. Other research efforts have been directed toward the incorporation of industrial or agricultural wastes for the same purposes or for their use as aluminosilicate sources in the preparation of innovative permeable concrete. These studies have been motivated by the need to reduce the environmental burdens of the conventional cement industry. Within this quest, special attention should be paid to the characteristics of the used solid wastes and their compliance with national regulations. In this respect, it should be noted that some solid wastes (e.g., coal ash, copper slag, rice husk) may contain considerable amounts of heavy metals, sulfur, and chlorine [47,52,111,112]. Subsequently, the extent of the presence of these contaminants and their dissolution and mobility should be assessed to determine their compliance with national regulations. In the case of noncompliance, a pretreatment process should be designed to reduce the risk of the presence of these contaminants and to mitigate their release. These topics have not been fully investigated in the literature and need to be addressed in depth to ensure the sustainable use of permeable concrete materials in preventing and controlling the pollution of water.

Extensive scientific efforts in the preparation and testing of permeable concrete have generated an acceptable range of values for the water-to-cement ratio and the coarse-aggregate-to-cement ratio, as indicated in Section 2.1. Yet, a standardized method that can be followed to produce universal permeable concrete products for specified applications is still missing [23,32,44–46,97]. In this respect, a standard practice to proportionate the used materials, identify the mixing procedure, and determine the optimum curing conditions is missing not only for innovative cement-based materials but also for conventional permeable concrete [97]. This quest to establish a procedure for the optimization of the mix design is also scarce in the literature. To the knowledge of the authors, only one paper elaborated on the development of a mix design procedure that was recommended for the optimization of the permeability and compressive strength of the permeable concrete [113]. The optimization procedure comprises three steps that benefit from a set of constitutive relationships, and the performance of a film-forming ability test to allow for the determination of the optimum mix design, as presented in Figure 4 [113].

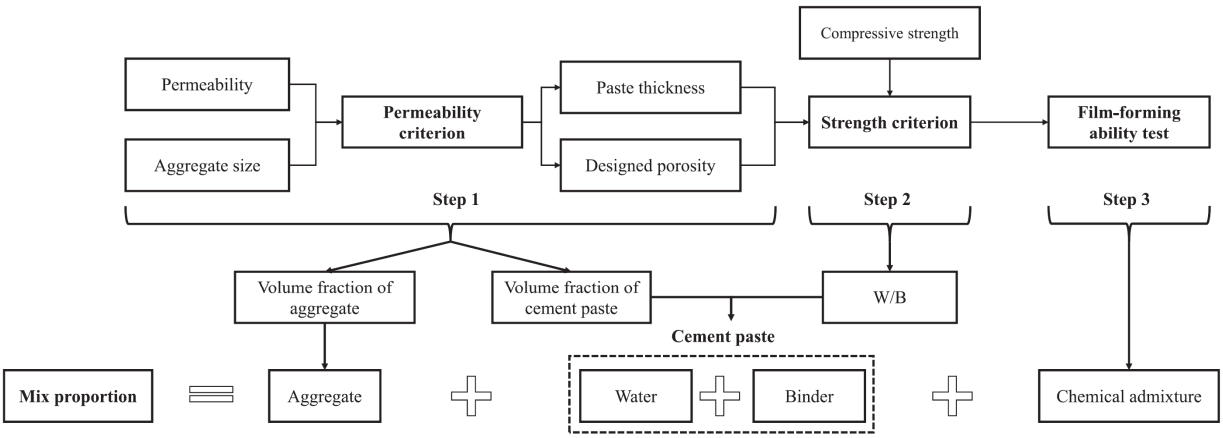


Figure 4. Proposed procedure to optimize the mix design of permeable concrete based on the permeability and compressive strength (copyrighted from [113]).

3. Characterization and Functional and Durability Performances of Permeable Concrete Materials

Different testing and evaluation techniques are used to characterize and evaluate the important properties of cement-based materials. These techniques are used to ensure that the requirements of both of the processing properties of the paste/fresh concrete and the desired performance and degradation resistance of the hardened materials are met [114,115]. On the one hand, standardized and nonstandardized characterization techniques have been developed to qualify the raw materials (e.g., grading), the paste (e.g., rheological properties), and the hardened materials (e.g., pore structure and permeability).

The details of these tests are found elsewhere [114,116,117]. On the other hand, functional performance evaluation and durability tests have been developed to ensure that the hardened cement-based materials meet the functional and durability requirements, respectively. For instance, permeable concrete used in permeable pavement applications is required to exhibit adequate hydraulic, mechanical, thermal, and sorption performances that enable the final hardened materials to effectively allow for the management of rainfall during their service life, meet the requirements on their strength (i.e., compressive, tensile, flexural, and abrasion resistance), and have improved sound absorption and temperature mitigation [32]. For permeable reactive barriers, their hydraulic performance requires that the flow is maintained under a natural hydraulic gradient without considerable retention within the barrier and to maintain good pollutant removal over their service life. In this section, the available characterization techniques and functional and durability tests applied to the permeable concrete materials are presented, and their features are summarized with some highlights provided regarding the effect of variations of the mix design on the measured properties.

3.1. Rheological Properties of the Paste

The composition of the mix design will affect the rheological properties of the paste and, subsequently, its adherence on the aggregates and the overall quality of the hardened material [113,118]. As the rheology of the paste will affect the thickness of the formed cement-based materials on the aggregates, it will affect its mechanical and hydraulic performances. In the case of permeable concrete, as the minimum amount of binder is applied, the importance of adjusting the rheological properties of the pastes becomes a necessity to avoid the formation of inhomogeneous paste and its segregation and the formation of hardened nonporous material or materials with low mechanical strength. As flowability is an important paste property, the adequacy of this mix design to produce a paste with adequate flowability can be checked via the application of the following tests [113,118–131]:

- Hand compacting method: This is a qualitative, easy method for testing the adequacy of the water in the paste and, hence, provides an indication of the paste's flowability (Figure 5a–c). Scarce water will yield a crumbling of the ball (Figure 5a), and excess water will yield an accumulation of paste on the glove, leaving the aggregates with a minimum coat of cement (Figure 5c). Adequate water will lead to the formation of a ball without excess paste accumulated on the glove (Figure 5c);
- Slump flow test: This test is used to examine the horizontal flow of the paste (ASTM 143/C143M-12) [126]. In this respect, the slump is recommended to be adjusted to near zero and less than 5 cm [122,123,126,128];
- The flow table test: this is used to measure the flowability of the paste (ASTM C230/C230M-14), where acceptable values are in the range 15–23 cm [87,129–131].

Additionally, the viscosity of the paste, yield stress, and adhesive force of permeable concrete pastes are measured using a rheometer or viscosimeter, and some researchers have studied the effect of these parameters on the compressive strength, thickness of the cement-based material on the aggregates, and on the pore structure of the hardened material [118,124,125,129,131]. There is an available standardized test to characterize the rheological properties of the paste using a rotational rheometer: ASTM C1749. Furthermore, there are available methods to measure the thickness of the formed paste layer on the aggregate: ideal paste thickness or actual paste thickness methods [113]. A modification of the actual paste thickness method was proposed using a flow table, and Figure 6 illustrates the procedure for conducting this method [124]. Another modification of this procedure was proposed by applying vibration or limiting the number of drops to 10 [113]. Additionally, image processing can be used to calculate the thickness.

Figure 5. Hand compacting method: (**a**) scarce water content; (**b**) adequate water content; (**c**) excess water content (copyrighted from [124]).

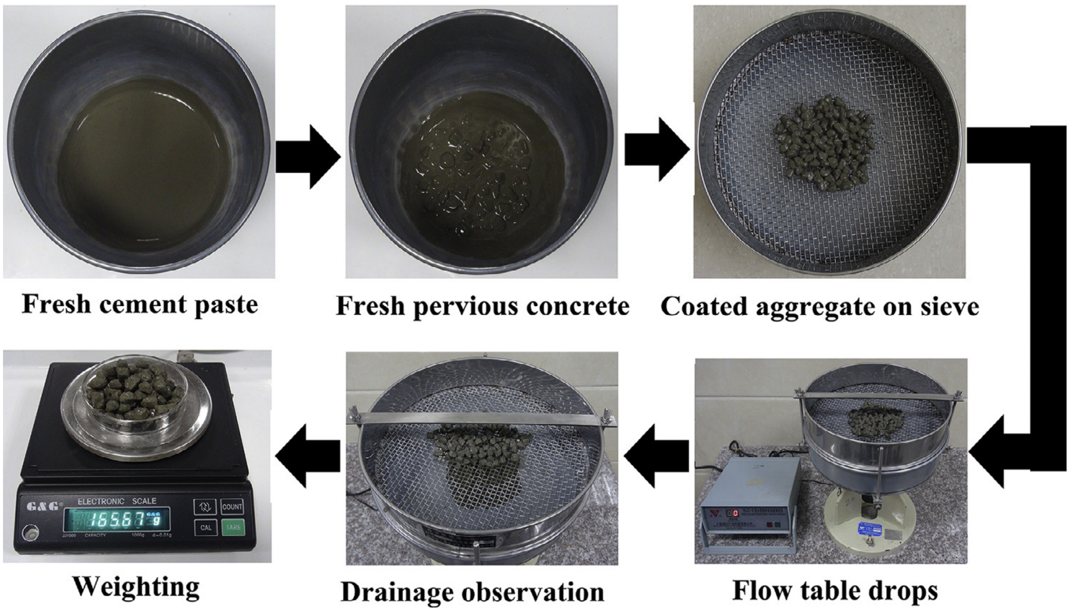


Figure 6. Procedure to evaluate the paste coating thickness on the surface of the aggregates (copyrighted from [124]).

Figure 7a–d illustrate the effect of the variation of the conventional cement mix design on the flowability, i.e., slump spread, yield stress, viscosity, and the maximum paste coating thickness on the aggregates [124]. Figure 7a,c show the effect of the variation of the w/c ratio in a simple permeable concrete samples composed of 100% cement as binder on these properties. The general behavioral trend can easily be deduced in these simple systems, where the flowability (i.e., slump spread) increases by increasing the w/c ratio and the rest of the properties display decreasing behaviors [124]. The behavior of these properties can be described using polynomial equations with correlation coefficients larger than 0.99. The effect of the variation of the SCM incorporation percentage in the permeable concrete (i.e., granulated blast furnace slag (GBFS)) on the same properties has a limited effect compared to that of the w/c ratio, shown in Figure 7a–c, as reported by Xie et. al. [124]. These studied samples were prepared at a lower w/c ratio (i.e., 0.28) and contained superplasticizer at a dosage equal to 0.4%. For those samples, the flowability slightly increased with the increased incorporation of the GBFS up to 50% and with the rest of the properties being reduced. The increasing correlation between the thickness of the cement-based material on the aggregates and the yield stress and the viscosity of the paste were confirmed for all of the studied mix designs (Figure 7a–d insert) [113,124,125].

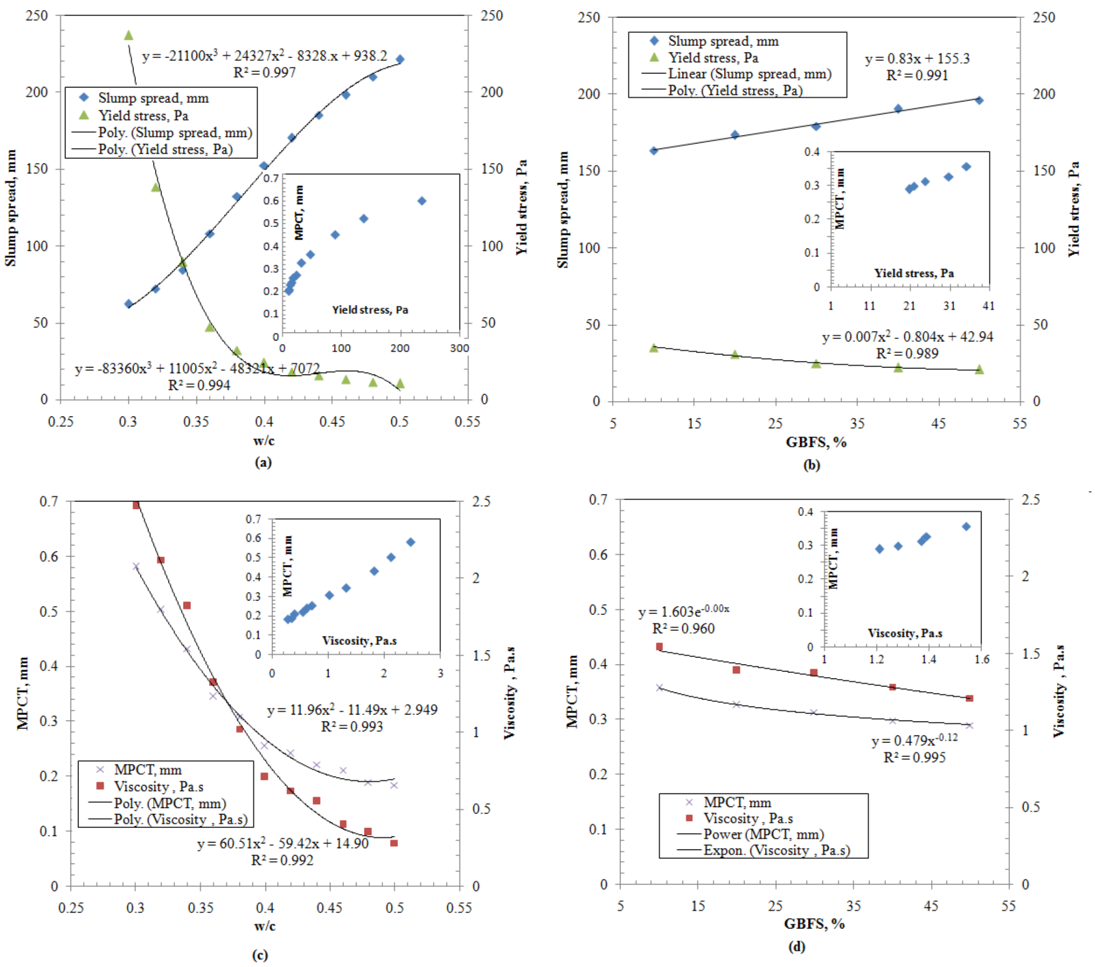


Figure 7. Effect of the mix design variation on the paste properties in conventional permeable concrete: (**a**,**c**) effect of the w/c ratio variation; (**b**,**d**) effect of the GBFS in cooperation (source data were extracted from [124]).

The effect of the variation of the sodium content in the alkali activator of the mix design of the permeable geopolymer concrete on the flowability of the paste, viscosity, shear stress, and the thickness of the hardened geopolymer coat was studied by Geng et al. [131]. The studied samples were BFS-based geopolymers prepared with water glass and sodium hydroxide in the presence of a retarder and limestone as coarse aggregate. Figure 8a–c illustrate the results in which the flowability, viscosity, and coat thickness increased with the increase in the Na_2O content up to 8%; then, the values of these properties decline. The increased behavior of the flowability and reduced behavior of the viscosity and shear stress of the paste is attributed to the formation on an electric double layer with repulsive forces on the precursor particles (i.e., BFS) due to the adsorption of activator silicate anions (Figure 8a,b). A further increase in the amount of Na_2O is claimed to lead to the enhanced formation of C-S-H gel that reduces the flowability and increases the viscosity and shear stress [131]. The variation of the geopolymeric coat's thickness on the aggregates (Figure 8c) shows a similar trend to that of the flowability [131]. The effect of the admixture on the rheological properties of the geopolymeric paste is an active area of study that aims to identify the effect of various admixtures on the workability of the paste. A study was devoted to investigating the effect of the water-reducing admixture and the molar strength of the alkali activator on the rheological properties of the fly-ash-based geopolymer paste [132]. The studied systems included both fine and coarse aggregates, as well as the alkali activator composed of a mixture of sodium hydroxide and sodium silicate. The results revealed that there was a critical molar strength (i.e., 4 M) beyond which the plasticizer and the superplasticizer had contradicting effects on the plastic viscosity, slump spread, and yield stress of the studied samples. In this respect, above this threshold value (i.e., <4 and =10 M), a clear decrease in the slump spread and increase in the yield stress and viscosity were recorded [132]. This study indicates that the employment of a lignin-based plasticizer leads to a better performance in terms of the paste's workability over that of a polycarboxylic–ether-based high-range water reducer.

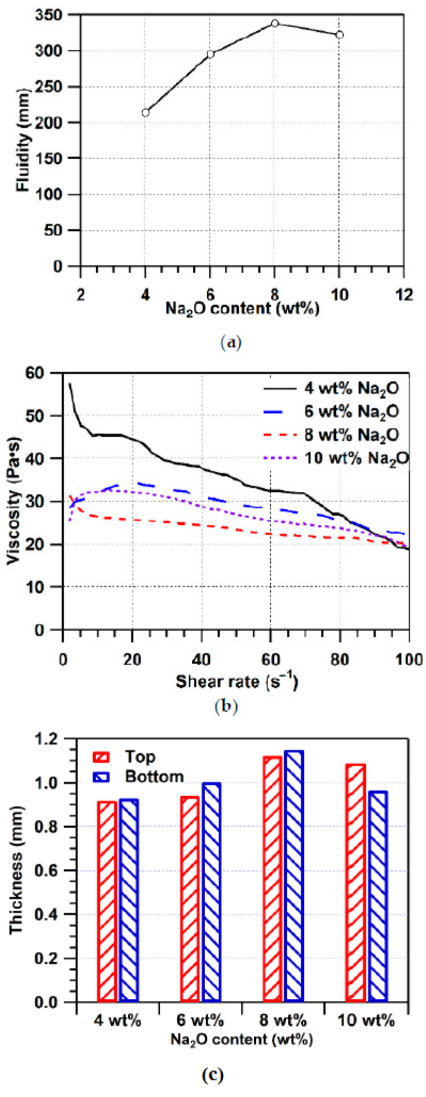


Figure 8. Effect of the variation of the mix design on the paste's properties in innovative permeable concrete: (**a**) fluidity; (**b**) viscosity and shear stress; (**c**) coat thickness (copyrighted from [131]).

3.2. Hydraulic Properties of Hardened Permeable Concrete

There are different standardized tests that have been issued to measure the pore structure and permeability of hardened cement-based materials, and there are other non-standardized tests that have been developed and used. These tests are used to measure the porosity, specific surface area, total connected porosity, pore volume, and air and water permeability [114]. These tests include:

- Gravimetric techniques: These are employed to measure the porosity using general standardized ASTM tests: ATSM C457/C457 M-16. It should be noted that a standardized test to measure the porosity of permeable concrete (ASTM C1754) was recently withdrawn;
- Absorption tests: These include the BET and MIP, and standardized ASTM tests are available for fragmented materials: ASTM D5604-96 and ASTM D4404-18, respectively. In addition, auto-clam and Figg tests are used to measure in situ water and air permeability [114];
- Ultrasonic techniques: These can be used to determine both the permeability and compressive strength of materials [116,133];
- Imaging techniques: These are used to construct 3D models for a sample, either using X-ray computed tomography or 2D scanning images and suitable image processing software. These models are used to drive empirical relationships for calculating the pore size and distribution and to model the mechanical and hydrological behavior of the material [97,114–138].

The permeability reflects the ability of the material to allow water to flow through it; it is dependent on the pore characteristics of the material, i.e., pore size, shape, connectivity, and tortuosity [22]. The permeability (k, cm/s) can be measured using the constant head method (Equation (1)) or the falling head method (Equation (2)) [22,97,139–141]:

$$k = \frac{QL}{Ah} \quad (1)$$

$$k = \left(\frac{aL}{At}\right) ln\left(\frac{h_1}{h_2}\right) \quad (2)$$

where Q is the flow rate, h is the head in the constant head method, A and L are the surface area and length of the sample. In the falling head method, a and t are the cross-sectional area of the pipe encasing the sample and the time required for the water pressure head to drop within predetermined levels (h_1 and h_2), respectively. Research papers that compare the validity and accuracy of both methods indicate that the permeability measured using the falling head method is lower than that of the constant head method, in which the latter is reported as viable and provides economic benefits [139,140]. It should be noted that the flow rate measured in permeable concrete is claimed to be in the transient flow regime between laminar and turbulent flow, which necessitates the use of a valid equation to describe the flow, i.e., Darcy–Forchheimer [124,140]. Several researchers have fitted the experimental data to obtain a relationship between the porosity and the permeability of the permeable concrete, in which linear, power, and exponential equations are derived [111]. It should be noted that these equations should be treated as mix-design dependent, and their validity should be tested before their application.

The effect of the variation of conventional cement mix design on the porosity and permeability of hardened permeable concrete is illustrated in Figure 9a–c [123]. The studied samples were prepared at a constant aggregate-to-cement ratio of 4 and different w/c ratios (0.24–0.32) and admixture dosages (0.4–1.1%) to control the final porosity. The investigations of the effect of the admixture on the values and relationships of the porosity and permeability at a fixed w/c ratio revealed that the use of a 0.4% admixture yielded a lower porosity and permeability of the hardened sample. The authors attributed this behavior to the paste's drainage into the interconnected voids [123]. The known effect of increasing the w/c ratio on the porosity and permeability was not clear in this study (Figure 9b). The visualization of the statistical distribution of the permeability of these samples, as shown in Figure 9c, indicates that the permeability decreases with an increase in the w/c. This is attributed by the authors in that work to the increase in the degree of lubrication, which results in a better densification of the mixture, consequently reducing the permeability [123]. It should be noted that the sample with the highest water content was prepared without any admixture, and the samples with the lowest water content were prepared with the highest admixture ratio; this clearly shows the importance of considering the rheological properties of the paste and their critical role in tailoring the hydraulic properties of the hardened permeable concrete.

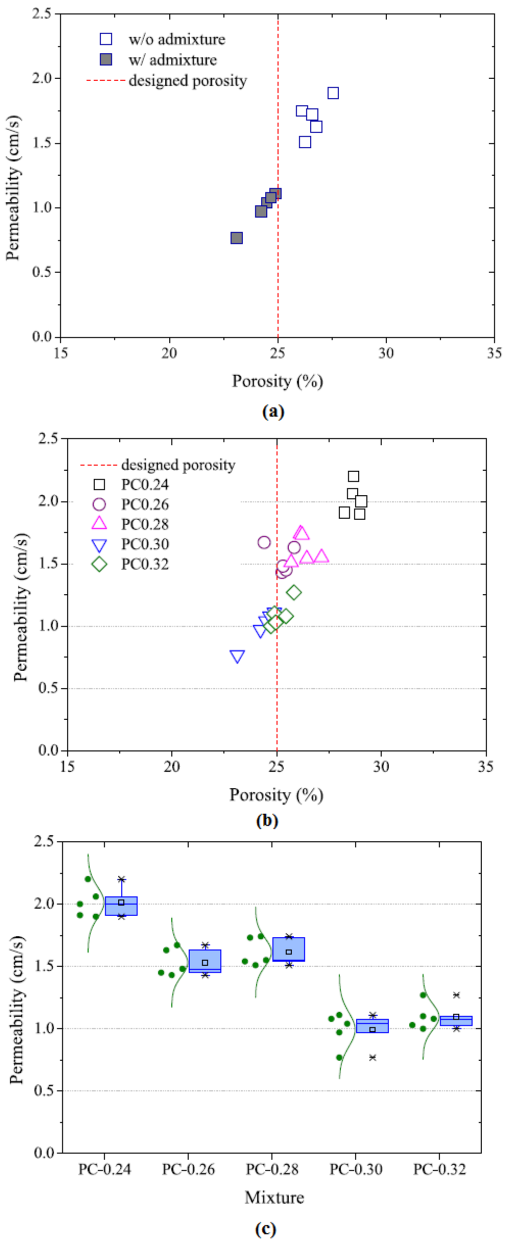


Figure 9. Effects of the mix design on the hydraulic properties of the hardened permeable concrete of different water to cement ration PC-X (X = 0.24; 0.26; 0.28; 0.3; 0.32): (**a**) effect of the incorporation of the admixture; (**b**) effect of the w/c ratio; (**c**) statistical distribution of the permeability at different w/c ratios (copyrighted from [123]).

The porosity and water permeability in the innovative permeable concrete is affected by the mix design of the innovative cement. In this respect, increasing the Na_2O content led to a reduction in the total and connected porosity and the permeability of BFS-based geopolymer prepared with water glass and NaOH in the presence of a retarder using limestone aggregates [133]. Another study was dedicated to investigating the effect of the substitution of GBFS with red mud to produce permeable geopolymer concrete using a combination of NaOH, Na_2SiO_3, and water glass as an alkali activator at an aggregate-to-binder ratio of 5 [40]. This study indicates that a 30%substitution led to an increase in the total void ratio and permeability by 7.69 and 6.35%, respectively [40]. Further, an increase in the red mud incorporation did not affect the void ratio and led to a reduction in the increase permeability to half of its value [40]. This behavior was not further discussed in that work. The substitution of the GBFS with red mud was investigated in another study, which confirms the increase in the porosity and permeability with an increase in the substitution by up to 50% [31].

3.3. Mechanical Properties

Mechanical properties are among the most important performance measures that need to be optimized during the design of any cement-based material. The hardened material strength should be preserved during its service life to allow for the sustainable functionality of the barrier. ASTM C109/C109M-20b [142]; ASTM C39 [143]; ASTM C496 [144]; and ASTM C293 [145] were developed to measure the compressive strength for cubic and cylindrical samples, splitting tensile strength; flexural strength, respectively. Several authors have used measured experimental data to deduce empirical models that can describe the relation between mix design components (e.g., water-to-cement ratio, cement content, aggregate size, and their porosity) and the compressive strength at a specified age (e.g., 28 days), flexural strength, tensile strength, elastic modulus, and fatigue [97].

As permeable concrete is designed to attain a specified mechanical performance and because of the limited amount of cementitious material in these composites, several authors have investigated the enhancement of this performance using different SCMs, fine aggregates, and fibers [25,51–61,79–89,94,95]. In this respect, it should be noted that the mechanical performance does not show a linear relationship over a wide range of these materials' incorporation. Therefore, an extensive number of review papers have addressed these effects in a comprehensive way [22,33,35,39,41,47,97,104]. For conventional permeable concrete, using fine aggregates was reported to improve the mechanical properties of the concrete. Thus, the reactivity of the used material plays an important role in determining their contributions to the build-up of the mechanical properties. In particular, the use of pozzolanic materials (e.g., volcanic ash, BFS, and FA) contributes to the long-term build-up of the strength of the hardened materials [47]. The inclusion of the fibers was proposed to improve the mechanical properties and durability of the permeable concrete, and plastic fibers were reported to have a limited positive effect on the mechanical performance of the hardened material [22,51]. Similarly, the use of permeable geopolymer concrete can be affected negatively with the use of low reactive materials in the mix design, and the use of red mud in GBFS geopolymers led to a reduction of the compressive strength [40]. This reduction increased with an increase in the GBFS substitution [30,40].

3.4. Durability of Hardened Permeable Concrete

In general, cement-based materials, as any other material, are affected by their presence in the environment, and the porous nature of these materials facilitates the penetration of water, gases, and aggressive materials leading to the activation of different reactions with the hydrated phases in the hardened materials affecting their durability. Both physical and chemical reactions occur between cement-based materials and ambient environmental components, leading to a reduction in their permeability [141,146], e.g., carbonation [79];sulfate attacks [80];induction of cracks through, for example, freeze–thaw cycles [61,73,80,88,128]; aggregate reactions [69]; and loss of materials via, for instance, leaching [24,36,61], abrasion [64,79,84,86,89], and erosion. In addition, the performance of these materials might be affected by the applied loads on them that can, in conjunction with environmental conditions, lead to serious failures. Subsequently, it could be concluded that both the hydraulic and mechanical performances of the cement-based barriers are affected by these conditions. Methods to solely assess the effects of these conditions are standardized, e.g., sulfate resistance (ASTM C452-21 [147]), aggregate reaction (ASTM C1778 [148]), freeze and thaw (ASTMC666 [149]), and aggregate soundness (ASTM C88 [150]). It should be noted that the standard test ASTM C1747 has been withdrawn.

Another important durability aspect of permeable concrete material is the clogging of the pores that affects the drainage performance of these barriers [20,22,35,49,124,151]. An evaluation of permeable concrete clogging is not only important for permeable pavement but for any permeable barrier, as it can seriously affect one of its main characteristic properties, i.e., permeability. Clogging is very similar to the well-known fouling phenomena in membranes, in which it can be traced to [22]:

- Physical clogging: an accumulation of suspended particles within the porous structure; this phenomenon does not include a chemical reaction;
- Chemical clogging: which occurs because of the penetration of chemical components into the flow of water through the barrier, leading to scale formation that clogs the porous structure;
- Biological clogging: which occurs because of the reproduction of algae and bacteria within the porous structure of the material.

As the clogging is mainly affected the hydraulic performance of the barrier, it is usually evaluated by measuring the permeability of the sample, as mentioned above, or by relying on the imaging techniques.

The effects of the variation of the mix design on the durability of the permeable concrete have been investigated [59,61,73,79,80,84,86,89,128]. The effect of using a combination of SCMs (i.e., nano silica (22.8%), spent fluid catalytic cracking catalyst (11.4%), and paper sludge waste (2.86%)) in conventional permeable concrete prepared using three gravel sizes at an aggregate-to-binder ratio equals to 5.6:1 on the leaching and freeze and thaw resistance of the hardened materials in the presence of melamine-based superplasticizer was addressed [128]. The study revealed that the use these materials enabled a reduction in the portlandite leaching and increased the resistance to freezing–thawing even after 50 cycles [128]. Another study addressed the effect of using nanomaterials on the physical durability of the hardened permeable concrete [59]. In this context, different samples were prepared using varying nano silica (0–4%) and nano iron (6%) contents in the IP and GU Portland cement in the presence of water-reducing admixture (0–1%) and cement substitution with FA in the range of 10–50% [59]. A multivariant method was followed to optimize the mix design to achieve a specified mechanical and hydraulic performance based on the maximum achievable compressive strength and target permeability equal to 8.8mm/s. The recommended optimum mix design comprised 24% fly ash, 1.9% nano silica/fly ash, and 0.35%admixture. The study concluded that the use of the nanomaterials improved the physical durability of the hardened materials but increased their costs [59]. The effect of modifying the surface of the recycled aggregates to improve the durability of the hardened permeable conventional concrete waste was investigated [73]. The study utilized a simple mix design composed of cement and recycled aggregate. The study indicated that the surface modification of the aggregates using a hydrophobic silicone membrane improved the durability of the hardened materials due to the following [73]:

- Its role in preventing the water accumulation on the aggregate surface, which allowed for the increased formation of dense C-S-H in the interface transitional zone;
- Its ability to mitigate the water absorption and migration into the aggregate that suppresses the ice pressure.

Well-optimized mix designs for innovative cement-based materials have been reported to provide beneficial durability performances under varying conditions including different chemical and freeze and thaw resistances [39,41,103,104,146]. In particular, the effect of the type of permeable geopolymer concrete (i.e., GBFS, FA, and MK) on the durability of the hardened materials was considered [26]. These three investigated systems at a target porosity of 20% were prepared and tested, including a control conventional permeable concrete and two permeable geopolymer concretes prepared from MK-GBFS and FA-GBFS. The MK geopolymer was found to have the best mechanical performance in terms of compressive and tensile strength and durability in terms of freeze and thaw resistance. The study indicated that the FA geopolymer provided remarkable benefits in terms of reducing energy consumption and greenhouse gas emissions.

3.5. Pollutants Removal Performance

As mentioned in Section 1, the presence of amorphous and crystalline hydration phases within the hardened permeable concrete provides sites for the chemical and physical entrapment of different anions and cations. This structural feature is employed in permeable concrete to remove different organic and inorganic pollutants from the water and entrap them within the material structure [6,8,12,15–19,21,24,25,27–31,36,38,40,41,48,49,62,63,93,103,151–153]. In this respect, monolith samples of permeable concrete are prepared either in the form of disks, cylinders, or cubes, characterized, and then tested for their potential application in removing pollutants (Figure 10). Both static [24,25,40,63] and dynamic [30,31,36,40,62] tests can be applied to test the ability of these materials to remove the pollutants of interests, including heavy metals, CODs, BODs, DODs, dyes, etc. In both types of experiments, the performance of the monolith material is expressed by recording the concentration measured in the aqueous phase or calculating the removal percentage (P, %) or the reduction ratio (Equation (3)) [40],

$$P\ ,\ \% = \left(\frac{C_o - C_m}{C_o}\right) \times 100 \qquad (3)$$

where C_x is the concentration of the studied pollutant before the first sorption cycle ($_{x=o}$) and after m number of cycles ($x = m$). Some of these investigations were directed toward assessing the effect of adding materials of a known sorption capacity [48], e.g., zeolite [20,154,155], iron oxide [62], grapheme oxide [36], or the addition of materials of known photocatalytic effects, such as iron oxide or titanium oxide to enhance the sorption ability. Finally different nanomaterials were investigated for the same purpose [59,62,63,151,156].

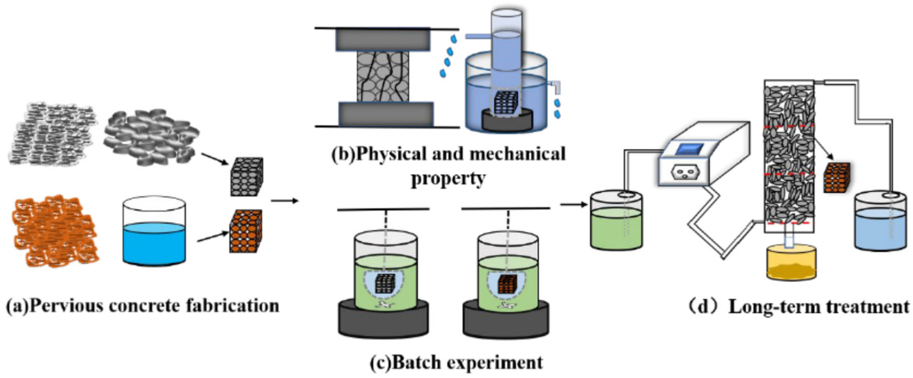


Figure 10. Experimental testing scheme for potential application of permeable materials in pollutant removal: (**a**) preparation of the monolith sample; (**b**) physical and mechanical characterizations; (**c**) static removal test; (**d**) dynamic removal test (copyrighted from [30]).

The performance of conventional permeable concrete prepared at a specified w/c ratio (0.3) and varying coarse aggregate-to-cement ratios in the presence of silica fume (SF; 10%) and FA (20%) aimed at the removal of total phosphorus (TP) and total nitrogen (TN) was investigated [25]. The study indicated that higher removal performances were obtained for samples that contained smaller sizes of aggregates and higher void contents. In this respect, 1.7 and 2.8 times more TP and TN concentrations were reported to be removed using permeable concrete containing 5–10 mm aggregates. The amount of attached microorganisms on the permeable concrete was concluded to control the amount of removed TP and TN [25]. In the context of the use of permeable concrete for water purification, the use of low pH concrete was suggested to reduce the leaching of the alkaline components that can affect an aquatic ecosystem [25]. The utilization of conventional permeable concrete prepared with OPC, basalt, FA, and SF for water purification was investigated [56]. In their work, Wang et al. investigated the performance of different mix designs of permeable concrete in the removal of suspended solids (SS), ammonia –nitrogen (AN), and total phosphorus (TP). The study concluded that SS removal was the highest followed by AN then TP, and the increased incorporation of FA led to a slight reduction in the removal performance. The study indicated that there is a need to improve the removal performance of AN and TP using auxiliary purification materials [56]. The performances of different permeable conventional concrete in the removal of some heavy metals were studied using batch static procedures by some investigators [24,40,155]. The studied systems were OPC-based permeable concrete with different types of aggregates, such as gravel, limestone, soda lime glass beads, and pumice, in the presence and absence of additive materials, e.g., FA and silica fumes. Table 3 lists the features of the investigated mix designs and the removal experiment conditions: initial contaminant concentration (C_o, ppm), time (t, h), and percentage removal (*P*. %). Holmes et al. confirm that there are several mechanisms that contribute to the removal of the heavy metals from the aqueous solutions onto the conventional permeable concrete [24]. These mechanisms include both chemical and physical sorption, precipitation, co-precipitation, and internal diffusion. Their study indicates that the use of calcareous aggregates improved the removal of heavy metals and reduced their leachability [24]. Finally, the dynamic removal of nitrate was studied using conventional permeable concrete prepared at varying coarse-pumice-to-cement (3–5), w/c (0.26–0.35), fine aggregates (0–20%), and nano-silica (0–6%) ratios [157]. The results indicate that the removal performance increased from 18.5–29% to 53.5–64.2% with an increase in the incorporation of nano silica [157].

Table 3. Contaminant removal performances for conventional permeable concrete.

Contaminant	Permeable Concrete Mix Design				Removal Conditions		P, %	Refs.
	Aggregates	A/C	w/c	Additives	C_o, ppm	t, h		
Pb	Gravel	5.5	0.4	-	2-207.2	72	84–91	[24]
	Limestone	5.5	0.4	-	2-207.2		87–88	
	GB *	5.5	0.4	-	2-207.2		88.5–92	
	Gravel	5.5	0.4	FA, 33.5%	2-207.2		31.5–92	
	Limestone	5.5	0.4	FA, 33.5%	2-207.2		87–88	
	GB *	5.5	0.4	FA, 33.5%	2-207.2		68.5–95.5	
	Na	5	0.37	-	50	0.5	38	[40]
Cd	Gravel	5.5	0.4	-	0.11-112.4	72	95–97	[24]
	Limestone	5.5	0.4	-	0.11-112.4		56–80	
	GB *	5.5	0.4	-	0.11-112.4		16–99	
	Gravel	5.5	0.4	FA, 33.5%	0.11-112.4		48–97	
	Limestone	5.5	0.4	FA, 33.5%	0.11-112.4		54–64	
	GB *	5.5	0.4	FA, 33.5%	0.11-112.4		39.5–78	
Cu	Pumice	3	0.35	Pumice, 10% SF **, 5%	Na	Na	97	[155]

Table 3. *Cont.*

Contaminant	Permeable Concrete Mix Design				Removal Conditions		P, %	Refs.
	Aggregates	A/C	w/c	Additives	C_o, ppm	t, h		
Ni	Pumice	3	0.35	Pumice, 10% SF, 5% **	Na	Na	71	[155]
Zn	Gravel	5.5	0.4	-	0.65-65.38	72	96	[24]
	Limestone	5.5	0.4	-	0.65-65.38		72–80	
	GB *	5.5	0.4	-	0.65-65.38		67–100	
	Gravel	5.5	0.4	FA, 33.5%	0.65-65.38		56–96.5	
	Limestone	5.5	0.4	FA, 33.5%	0.65-65.38		76	
	GB *	5.5	0.4	FA, 33.5%	0.65-65.38		32–88	

Notes: * GB soda lime glass beads. ** SF silica fume.

An investigation of the potential use of innovative permeable concrete in the removal of different heavy metals was conducted [40]. In comparison with conventional permeable concrete, the innovative permeable concrete was found to have higher removal performance, and this increase is contaminant specific, e.g., Cd (18.75%), Pb (17.91%), Cu (25.07%), and Cr (39.18%) [40]. This performance was further increased by increasing the substitution of GBFS with red mud due its fine particle and high surface reactivity [40].

4. Permeable Reactive Concrete Barriers

The remediation of contaminated ground water is an essential activity to ensure the sustainability of this source of water, and both active and passive ground water remediation technologies have been implemented. Specifically, passive remediation via permeable reactive barrier have been applied since 1995.The basic idea behind this technology is to install a porous reactive material below the ground surface in the down gradient direction of the plume of the contaminated ground water, which allows for its passage under a natural hydraulic gradient [156–163]. Figure 11 illustrates the configuration of the funnel and gates (F&GPRB) and the continuous trench (CPRB) installation of the permeable reactive barrier [161]. This technology provides several economic and technical benefits compared to active ex situ groundwater remediation technology, i.e., pump and treat. These benefits include the following [158,161–163]:

- Low energy consumption, which reduces the carbon footprint of the process and the operating costs;
- Requires monitoring its activity with only minimum scheduled maintenance after a specified period of operation, if needed;
- Easily installation and removal procedures;
- Provides efficient and targeted remediation, where these barriers convert specified pollutants to fewer toxic species and/or retains them.

The major limitations of this technology include the depletion of the reactive chemical compounds over time due to their reaction with the contaminant and the clogging of the barrier pores. These limitations can be addressed by relying on the efficient design of the barrier that addresses the plume and site characteristics.

These barriers are designed to activate several mechanisms to decontaminate the groundwater plume, e.g., precipitation, sorption, and degradation [162]. Conventional reactive materials have been investigated and implemented for this purpose including zero-valent iron (ZVI), carbonaceous materials, sulfate-reducing bacteria, metal oxide/sulfides, mineral materials, and industrial wastes [159,161,162,164–166]. Innovative materials include the use of single materials, e.g., meso-zero-valent iron, permeable concrete, basic oxygen furnace slag, or modified/composite materials [164,166]. The factors that affect the selection of materials for permeable reactive barriers include [159]:

- Reactivity: the capacity of the material should be high enough to allow for the precipitation and/or sorption and/or degradation during its service life;
- Hydraulic conductivity: the barrier should have adequate permeability to allow for the passage of the contaminated ground water without considerable retardation in its velocity;
- Environmental compatibility: the used material should not have the potential to release toxic species into the host environment;
- Long-term physical and chemical stability: the material should have adequate long-term stability to eliminate the need for maintenance of the barrier during its service life.

Table 4 lists illustrative examples of the large-scale implementation of permeable reactive barriers in remediating different contaminants [164,167–172]. In this respect, it is clear that this technology has been successfully implemented to remediate different types of contaminants including both organic and inorganic. Only one of these examples, based in Willisau, Switzerland, claimed to require additional remediation action, as it did not achieve its target performance, and this was attributed to the complicated hydrogeological conditions at the site and the geometry of the installed barrier [164,170].

Permeable concrete has been investigated for its potential use in the construction of permeable reactive barriers for ground water remediation [30,31,154,156,173–183]. In particular, these research efforts were directed toward the assessment of the potential use of these materials for the remediation of acidic contaminated groundwater, in particular contamination from acid mine drainage, in which the high buffering capacity of these materials will sustain its performance over long periods of time. Published studies in this field are very limited compared to those related to the application of these materials in the low-development impact practices. These studies addressed the pollutant removal performance of these materials under acidic conditions in the mine acid drainage plume, where the effect of the incorporation of the aggregates and SCMs was studied, the permeable concrete performance in removing the pollutants was compared to that of the mature zero-valent iron (ZVI) materials, and the use of these materials in combination with the active method was proposed. Most of the published works focused on conventional concrete, where the use of innovative permeable concrete was addressed in a limited research. In this section, the published research in this area is summarized.

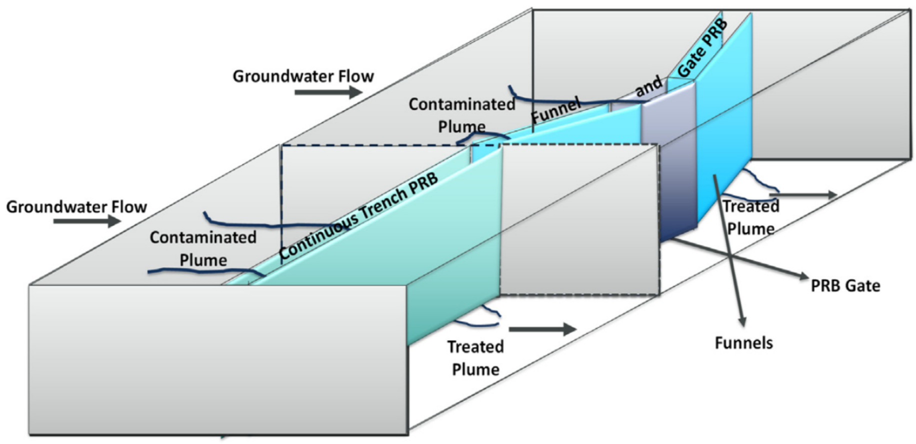


Figure 11. Configuration of the PRB designs: (copyrighted from [161]).

Table 4. Large-scale application of permeable reactive barriers in the remediation of different contaminants.

Location	Nature of the Contamination			PRB Specification		Performance	Refs.
	Target Contaminant	Site Characteristics	Plume Characteristic	Dimension	Barrier Materials and Specification		
Ontario, Canada	Perchloroethene (PCE) and trichloroethene (TCE)	Medium–fine sand underlain by a clayey silt deposit at 9 m below ground level	1 m wide and1 m thick, C_o = 270ppm (TCE) C_o = 50 ppm (PCE)	L = 5.5m, W = 1.6 m, D = 2.2 m	CPRB mixture of ZVI and commercial coarse sand (22:78)	After 299 days, the TCE and PCE were reduced by 90% and 85% with an uncertainty of less than ±5%.	[167]
Northern Ireland, UK	TCE	Sand and gravel over Sherwood sandstone	C_o = 390 ppm	Full scale D = 8 m	F&GPRB with ZVI as the active material	Calcite precipitation observed in the upstream improved the ZVI reactivity. No biological fouling.	[164,168]
Willisau, Switzerland	Chromate	Clayey and silty sand underlined by sand and gravel	C_o < 10 ppm	Full scale	Double array of vertical piles containing iron shavings and gravel	Did not achieve its removal target due to its location in nearly oxygen and calcium carbonate saturated aquifer in a regime of high groundwater velocities.	[169,170]
Florida, USA	Iron and manganese	Clayey sand	C_o = 30 ppm (Fe) C_o = 1.62 ppm (Mn)	L = 6 m, W = 0.9 m, D = 4.6 m	Limestone and crushed concrete	In the first year, Fe removal efficiency 91–95% Reduced performance after three years showed due to clogging.	[171]

4.1. Effect of the Aggregates

The effect of the aggregates type on the pollutant removal performance of permeable concrete was investigated by evaluating the nitrate sorption onto permeable concrete samples prepared with different aggregates [173]. In this respect, type 2 OPC, coarse aggregates (3.8 in no. 4), and fine aggregate (no. 6 and no. 8) were used to prepare the studied samples, and pumice, zeolite, and perlite were tested. The water absorption, alkali–silica reactivity, and permeability were measured for all samples, and it was found that pumice showed the lowest water absorption (7.84%), maximum permeability (1.64 cm/s), and lowest expansion during the alkali–silica reactivity tests (0.02%). In batch experiments, the reaction between the different samples and nitrate solution (C_o = 70 ppm) was studied, it was reported that the reactions reached equilibrium in 30 min for samples containing perlite and pumice aggregates and required longer times for the samples containing zeolite aggregates. In addition, the activation of the aggregates using HCl and H_2SO_4 was concluded to improve the nitrate removal performance, but it did not affect the time required to reach equilibrium. In this regard, the activation of pumice aggregates using HCl was found to improve the nitrate removal from 39% to 50%.

The removal performance of permeable concrete containing granite (Gr) or dolomite (D) coarse aggregates was compared for their potential use in the management of acid mine drainage [182]. The study indicated that granite aggregates resulted in better treatment of manganese (+32%) compared to that of the dolomite aggregates. Both types of aggregates had a fairly equal removal performance for sulfates (Gr = 30% and D =29.3%); calcium (Gr = 84.7% and D = 85%); and iron (Gr = 99.5% and D = 99.6%). Another study assessed the hydraulic performance of permeable concrete of different aggregate types that was used in the treatment of acid mine drainage [180].In that work, the effect of using 30% fly ash as an SCM was addressed at different water-to-cement ratios (0.25–0.27), and different coarse aggregates were used, including dolomite (67 and 95), granite (67–132), shale (67), and andesite (67) [180]. The studied samples were reported to have effective removal performances after 90 days for Al (98.1%), Mg (86.5%), Mn (99.8%), Zn (97.4%), and Fe (99.4%), with an increased chromium concentration due to chromium leaching (−112%) from the cement and fly ash. The samples showed high pore connectivity (95–99.7%), which reflects the maintenance of the hydraulic performance. In addition, it was reported that the isolated porosity increased from 0.1% to 0.86%, while the pore connectivity was reduced from 99.7% to 95.3% for the samples that contained fly ash. A study was dedicated to addressing the effect of the aggregates on the removal performance and costs of the remediation project [24]. In that study, the permeable reactive barrier mix design relied on the use of gravel, limestone, or soda lime glass beads in the presence or absence of fly ash as supplementary material to remediate mine acid drainage. A fixed water-to-cement ratio of 0.4 was used and a fly ash substitution of 25% was considered to prepare permeable concrete with a 25% void content. The results revealed that with high cement content, the hydration products buffered the water leading to the precipitation of lead, zinc, and cadmium. Barriers containing limestone were proved to provide a better removal performance for the studied heavy metals (see Table 3). The study analyzed the costs of the needed materials for the permeable concrete barriers in comparison with the aggregate alone and activated carbon to remediate a plume consisting of a single heavy metal, in which the lime stone aggregate alone had the least costs (USD 35,000,000 for zinc removal assuming a total mass of 1650 $\times$ 10^6 kg). It should be noted that these preliminary cost estimates did not consider the breakthrough characteristics of the barrier or any other kinetic data [24]. In addition, the study revealed that the calcite leaching from the concrete will coat the co-precipitates and provide an additional increase in the reactive surface sites that can increase the life service of the barrier. The leaching of the heavy metals from the prepared concrete was reported after 72 h and was found to be less than 5%. All of the studied samples were found to have a considerable buffering effect on the acid mine drainage solution that increased the initial pH from 5.6 to values higher than 11.4 after 3 days of contact between the samples and the solutions [24].

4.2. Use of Supplementary Cementitious Materials

Red mud, fly ash, nano silica, and a mixture of silica fume, zeolite, and iron oxide were tested to investigate their effects on the pollutant removal performance of permeable concrete samples. Table 5 summarizes these effects. Permeable concrete samples were prepared using OPC, coarse aggregates, water, superplasticizer, and red mud and were investigated to evaluate their performance in the removal of the contaminants from acid mine drainage [30]. The strength, porosity, and permeability of the samples were measured, the strength was found to be correlated negatively with the increase in the red mud content in the sample, and the permeability and porosity had a positive correlation with the red mud content. These results are attributed to the highly alkaline nature of the red mud and higher Na_2O content that led to a fast reaction with the water at the onset of the hydration reaction. In addition, the use of red mud was reported to improve the removal reaction kinetics. At an optimum influent pH = 4 and hydraulic retention time = 24 h, the complete removal of Cu, Mn, Cd, and Zn was achieved, and the removal mechanism was explained by the hydrolysis of the portlandite upon its reaction with the sulfate ions to produce gypsum that led to the precipitation of these ions accompanied by the sorption onto the C-S-H and hematite in the red mud. The prepared material was reported to have had its efficiency in the remediation of heavy metal from acid mine drainage samples collected from a mining area in China proven.

Table 5. Effect of using SCMs on the performance of permeable reactive concrete barriers.

Material	Mix Design			Performance	Refs.
	W/C	A/C	Supplemented Material		
Red mud	0.21–0.23	3.25	Red mud 25, 50% superplasticizer	Reduced the compressive strength, effective at pH = 4 hydraulic retention time of 20 h	[30]
FA	0.27	4.02	Fly ash 30% superplasticizer	Enhanced the real acid mine drainage treatment	[31]
FA+ nano silica	0.26	5	Fly ash 20%, Nano silica 6%	Enhanced the nitrate removal and compressive strength	[157]
SF+ zeolite+ iron oxide	0.25	4	SF 5.05% zeolite 5.45% iron oxide 0.5%	Enhanced the heavy metal removal	[155]

A mix design containing type 1 OPC, granite (95 mm) coarse aggregates, and fly ash as SCM was tested to treat acid mine drainage collected from a gold mine and coalfield [31]. The study investigated the role of fly ash in enhancing the reactive performance of the barrier by improving the pollutant removal. The hardened concrete with the supplementary material was found to enhance the pollutant removal performance for Al, Fe, Mn, Co, and Ni, and this behavior was attributed to the high buffering capacity of the material that raised the acid mine drainage pH to 12 and, subsequently, led to the precipitation of the pollutants' hydroxides. The portlandite in the permeable concrete reacted with the sulfate ions in the contaminated solutions leading to the formation of the expansive gypsum. The presence of the fly ash was concluded to mitigate the damage that could be initiated by the formation of the gypsum. The combined effect of the use of fly ash and nano silica on the properties of the hardened permeable reactive concrete was investigated [157]. In this regard, the optimization of the mix design of permeable concrete consists of OPC type 2, coarse aggregates (No. 4), fine aggregates (No.6 and8), nano silica and fly ash was conducted according to Taguchi multivariant procedure (L9). The compressive strength, density, and void ratio, and permeability were measured according to the ASTM C39, ASTM C 1754, and ACI522, respectively. The enhanced nitrate removal was attributed to the provision of new surface functional groups, Si^{2+} and Si oxide, due to the addition of the SCM, and FTIR investigations indicated that the hydroxyl stretching peak at 3430 cm^{-1}, silicon tetrahedral peak at 1050 cm^{-1}, and Si-O bending vibration peak at 446 cm^{-1} were affected by the sorption. Another study investigated the effect of the flow configuration—gravity and down-up configuration—on the pollutant removal kinetics of permeable concrete supplemented with fly ash and silica fume and concluded that the gravity flow had two orders of magnitude less liquid–concrete contact time to have a similar acid mine drainage treatment quality [179]. This conclusion should be considered carefully, as the use of pumps in the down–up configuration will change the nature of the application from a passive to active mode.

4.3. Performance Comparison with Zero-Valent Iron

A batch experiments was conducted to compare the pollutant removal performance of permeable concrete materials against that of zero-valent iron [176]. The permeable concrete was prepared from type 1 OPC and 67 mm granite aggregates at a water-to-cement ratio equal to 0.27. That study investigated the performance of three samples, namely, permeable concrete (CEM1), permeable concrete with 30% FA SCM (30%FA), and zero-valent iron (ZVI), in the treatment of two types of acid mine drainage: from a gold mine and from a coal mine. The ZVI sample was found to buffer the acid mine drainage solutions from 4.15–5.79 to 6–8, whereas the cement-based samples buffered the solutions to higher pH values of 9–12.The results of the batch reactor tests conducted for 43 days, as shown in Figure 12a,b, indicate that the removal rates for Al, Fe, Ni, Co, Pb, and Zn from both acid mine solutions were higher than 80% for all of the studied permeable reactive barrier samples. The permeable-concrete-based samples had higher removal performances for the removal of Mg and Mn from both solutions. The high removal performances of these samples are attributed to the pH-driven metal precipitation of the pollutants on the surface of the permeable concrete and the formed gypsum. It should be noted that all of the studied materials led to the release of sulfate, which was higher in the ZVI sample [176]. In another batch study, the two permeable concrete samples and the zero-valent iron sample were tested to investigated their pollutant removal performance in the treatment of acid mine drainage collected from the coal and gold mines [178]. The first permeable concrete sample was non supplemented, i.e., type 1 OPC and granite aggregates, and the second was supplemented permeable concrete with fly ash (30%). The results of the batch experiment were compared against effluent discharge standards and revealed that Zn, Fe, Ni, Co, Pb, and Al were effectively removed by the three tested material. Both permeable concrete materials had a better performance at removing Mn and Mg. However, the treated solutions with the permeable concrete were buffered to a high pH and contained higher Cr^{6+} concentrations that affected their overall quality. In a third study, the hydraulic and pollutant removal performances of the permeable concrete and zero-valent iron were evaluated under dynamic flow conditions in a column experiment [167]. In this work, the performance concrete barrier buffered the initial acid mine pH from 2.99 to 11, whereas the ZVI barrier buffered the pH to 9. The permeable concrete containing 30% fly ash was reported to improve the retardation factors over that of the zero-valent iron. In terms of the hydraulic performance, the hydrodynamic dispersion coefficient for the 30-FA permeable concrete was higher than that of the zero-valent iron. Finally for a barrier of 1.5 m thickness, the estimated life service of the permeable concrete was double that of the zero-valent iron.

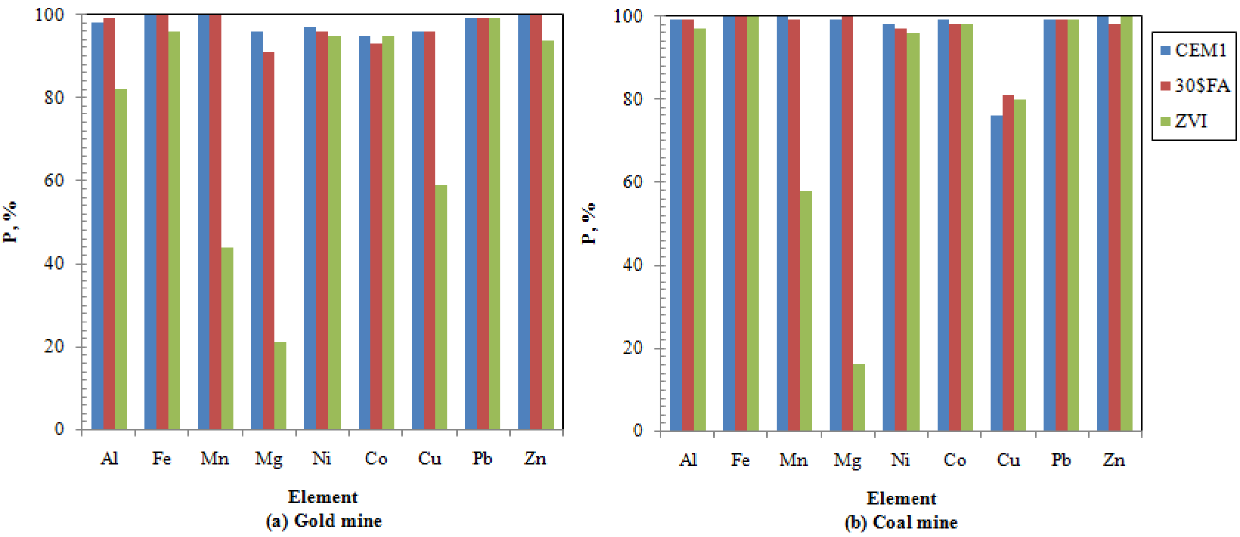


Figure 12. Removal performance of the different metals in the acid mine drainage from (**a**) gold and (**b**) coal mines (source data extracted from [176]).

4.4. Combined Treatment Method

A combination of active and passive processes were proposed for the treatment of acid mine drainage, and these processes included the use of permeable concrete as a pre-treatment passive barrier and in active anaerobic digestion for the treatment of effluent collected from a coal mine in South Africa [175]. The mix design of the permeable concrete barrier was as follows: 4% silica fume, 8% fly ash, and granite coarse aggregate (13.2 and 9.5 mm). The use of the passive concrete barrier enabled the efficient removal of iron (99%), potassium (94%), and alumina (42%), whereas the bioreactor enabled the removal of COD (89.7%) and sulfate (99%). This study revealed that the iron precipitated in a respectively short contact time of 37 s and the satiability of the suspended ions presented an opportunity of its removal from the sludge.

4.5. Gaps in Investigating Permeable Reactive Concrete Barriers

As indicated above, a limited number of published research papers have been directed toward the investigation of the potential use of permeable concrete materials as permeable reactive barriers for ground water remediation. These efforts have mainly focused on studying their pollutant removal performance under the challenging operating conditions of the remediation of the mine acid drainage plume characterized by high sulfate content and acidity. In general, these conditions are known to affect the durability of cement-based materials, yet the obtained results from these research efforts reflect the potential feasibility of their application. This feasibility can be emphasized by addressing some gaps that exist in this field as follows:

- The beneficial use of fly ash to improve the mechanical strength of permeable reactive concrete barriers and to provide more active sites for the removal of pollutants is promising [31,174,178,179]. Yet, it was concluded the cement can contribute to an increase in the Cr concentration in the treated plume [178,180]. This point needs to be addressed in depth by investigating the stability of fly ash using a standardized test (e.g., TCLP) to assess the feasibility of this material's use. In addition, an in-depth analysis of the stability of the hardened material and the deduction of the chromium-leaching mechanism are needed.
- The reported mechanisms of pollutant removal were precipitation due to the alkalinity of the permeable reactive concrete media combined with physical and chemical sorption onto the hardened cement phases, the aggregates, and the SCM [30,173,176,178,179,181]. In particular, the reaction of the sulfate with the portlandite was identified as enhancing the precipitation of the pollutants. The effect of the portlandite reaction with the sulfate on

the permeability of the barrier needs to be assessed, as the formation of expansive phases can affect the long-term hydraulic performance of the barrier.

- Concrete is known to have a high alkaline capacity, which has been reported to buffer the treated solution's pH to an unacceptable value, i.e., 11 [178]. This point can be addressed by using low-pH cement that can buffer the treated solutions to pH values comparable to that of the ZVI. Different types of additives can be used in this respect that should be studied in depth to ensure that the hardened material will meet the hydraulic and pollutant removal performances.
- Innovative permeable concretes have not been tested extensively for their applications in groundwater remediation and acid mine drainage treatment. In particular, magnesium phosphate cement is known for its fast hardening, near neutral pH, low water requirements, and high adhesive strength [5,6]. These materials have been tested for their application in low-development practices but not in permeable reactive barriers.
- The effect of the rheological characteristics of permeable concrete paste and their effects on the hydraulic and pollutant removal performances have been not investigated for permeable reactive concrete barriers. Most of the conducted studies relied on the use of the minimum amount of water (0.21–0.33) in conventional permeable concrete [30,31,156,179,180]. This limited range compared to that used in the low-development impact (0.25–0.45) practices reduces the porosity of the hardened material. This can affect the hydraulic performance of the barriers used in the remediation of the acid mine drainage over the long term, and the formation of expansive phases can lead to a further reduction in the porosity.
- The available experimental data on the performance of permeable concrete reactive materials were conducted within limited time frames of less than a year [21,24,176,178,180,183]. The long-term performances of these materials are required to be studied in depth to ensure the sustainable performance of these barriers throughout their service life and to identify threshold values for their reduced performances affecting their efficiency as a barrier.
- The durability of permeable reactive concrete barriers needs to be addressed to evaluate the effect of harsh operating conditions (i.e., high sulfate and acidic solutions) on the durability of such barriers. Future research needs to address the feasibility of performing scheduled maintenance or substitutions of the barrier material if the barrier does not reach its target remediation prior to reaching unacceptable reduced performances.
- Current research efforts in this field are focused at the lab-scale testing and mainly focus on conducting static experiments, with limited research studying dynamic conditions [30,31,36,181]. There is a need to address the performance of these materials under more realistic conditions that address upscale practical applications of these materials.
- The cost of the remediation technology is an important aspect that affects the decision-making process [21,31,177,180]. The cost estimate for these materials are limited, and these studies have addressed the costs of these materials and trench-type installation costs [24,30,179]. Detailed cost analyses have not been conducted; subsequently, there is a need to address the life cycle costs of the large-scale application of these materials.
- The environmental impacts of the use of permeable reactive concrete barriers need to be addressed. In this respect, the effect of the beneficial use of recycled wastes as SCMs and aggregates in conventional performance concrete should be quantified in terms of the reduction of the environmental footprint. In addition, the impacts of the buffer pH after treatment should be addressed.

5. Conclusions

The feasibility of using permeable concrete as a reactive barrier for the remediation of contaminated groundwater and the treatment of the acid mine drainage was addressed. This class of materials has been long implemented in low-impact development practices and their efficiency has been proven, but these materials are still considered innovative for their application as permeable reactive barriers, with a limited number of studies that have been published. These published studies have proved the efficiency of this class of materials for the removal of pollutants under the harsh acidic conditions of acid mine drainage, but some problems that affect the quality of the treated water have been identified. On the basis of the conducted literature review in this work on the mix design of conventional and innovative permeable concretes; their characterization and performance and durability assessment tests; and the feasibility assessment for their application as permeable reactive barriers, the gaps and recommended future research direction were identified to emphasize their feasibility toward pilot-scale applications. In this respect, the effect of the variation of the mix design of conventional and innovative permeable concretes on the rheology of the paste and the hydraulic, mechanical, and removal performances of the hardened material were identified. The practicality of the application of these materials in the remediation of contaminated groundwater and acid mine drainage were evaluated by reviewing and analyzing the published research in the field and identifying the gaps and suggestion of future research directions. The main conclusions that can be drawn from this work can be summarized as follows:

- The optimization of the mix design of the permeable reactive concrete barrier needs to be guided not only according to the required hydraulic and removal performances but also by the durability of the hardened materials over the designed service life;
- Identifying the environmental impacts of the optimized permeable reactive concrete barrier is crucial to ensure the sustainability of these materials. These impacts should consider the dynamics of the attenuation of the alkaline plume downstream of the barrier of the affected environment.

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