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## Pb removal in pervious concrete filter: Effects of accelerated carbonation and hydraulic retention time



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#### HIGHLIGHTS

• Pervious concrete was seen to be an effective medium to remove Lead from aqueous solutions.

• Accelerated carbonation technology use on pervious concrete reduced the propensity for calcium leaching during the filtration process.

- Hydraulic retention time was found to be vital in the filtration performance of pervious concrete.
- Image analysis technique was found to be a powerful tool in analyzing the pore structure of pervious concrete.

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#### ABSTRACT

In this study, the effects of accelerated carbonation and hydraulic retention time (HRT) on the lead (Pb) removal in pervious concrete filter (PCF) were investigated. The Pb containing solution of 8 mg/L concentration was constantly passed for 60 min through three PCF monoliths that were arranged in series with progressively decreasing water heads. This three-tier filtration experiment was conducted on PCFs, which were tested right after initial curing as well as after an accelerated carbonation exposure. Cumulatively, the Pb removal in 7-day cured and carbonated PCFs were found to be 97.2% and 99.1%, and thus it showed that the CO<sub>2</sub> uptake by PCF marginally improved the Pb fixation, and further these results strongly recommend the HRT consideration in concrete filter design for real-field applications. Moreover, the leachability test results suggests that the change in physical characteristics of treated water from carbonated PCF was found lesser than 7-day cured PCF regardless of the inlet solution pH. The Ca ions removed from both PCFs post-treatment were found well below drinking water acceptable limits. The cost of both PCFs were estimated, which appeared inexpensive unlike treatment units that are currently in practice. On the whole, this study effectively demonstrated the potential of 7-day cured and carbonated PCFs for cost-effective Pb removal.

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#### 1. Introduction

Among the heavy metals, lead (Pb) is of particular interest because of its toxicity and widespread presence in the environment. The Pb discharge from industries (battery manufacturing, metal plating, soldering, paint and pigment production and oil refining) into surface and ground waters is increasing significantly [1,2]. As per EU Drinking Water Directive [3] and IS 10500 [4], the maximum acceptable limit of Pb in drinking water is  $10 \mu g/L$ . In May 2014, Central Water Commission, Govt. of India reported that the Pb concentration in eight Indian rivers (Brahmani, Ganga, Gha-

\* Corresponding author. *E-mail address:* manusanthanam@gmail.com (M. Santhanam). ghara, Gomati, Mahanadi, Ramganga, Rapti and Yamuna) exceeded the IS 10500 [4] acceptable limits. This polluted water on consumption can cause severe health disorders like kidney diseases, chromosome and nervous damage, and perhaps even leading to death [5]. The heavy metals from industries are typically treated either by adsorption, electrolysis, precipitation, ion-exchange, oxidation, reduction, and reverse osmosis. However, the major drawbacks in practising these methods are high initial capital cost, operational and maintenance costs [2].

Pervious concrete filter (PCF) is a special type of concrete that allows the waste water to pass under gravity through its coarse, interconnected pore-structure. The highly alkaline PCF neutralised the acid mine drainage (from pH of 2.8 to 7) and additionally trapped 99% Fe, 83% Mn and 30%  $SO_4^{2-}$  from such aggressive aque-

ous media [6]. The PCF also fixated metals Co, Ni, Fe, Mn and Al from gold mine and coalfield drainage [7]. Laying pervious concrete (18-35% porosity, 2-8 mm voids size, 2.7-27 MPa compressive strength, and 80–720 L/min/m<sup>2</sup> infiltration rate) as road pavement reduces the storm water volume and simultaneously fixate heavy metals Pb, Cd, Cu and Zn, which can contaminate the ground water source if not strongly trapped [8–10]. Vadas et al. [11] demonstrated that the tortuous pore network pre-occupied with iron oxide and fine organic matter could improve the metals fixation in PCF. The choice of PCF is advantageous and economical than treatment methods that are currently in practice, which is due to its ease in mass production, the ability to fabricate at any location, and lesser requirement of skilled labour and sophisticated machines for preparation. The use of crushed limestone in PCF preparation seems to improve the metals fixation [12]. Jiang et al. [13] pointed out that the HRT increase could improve the metals fixation in PCF, which has not been strongly explored and presented in the literature, to the best of our knowledge.

The Pb ions are immobilised better in PCF than Cu and Zn and these fixated metals could easily leach when the pH of flowing water reduced to 8 or lesser [7,11,14,15]. The Pb ions fixate in PCF because of complexation and solids formation with the OHand  $CO_3^{2-}$  species (i.e. ion-exchange) that are attached to calcium in the concrete [15]. Moulin [16] demonstrated batch adsorption studies to determine the sorption isotherms of Pb(II) ions onto the pure C<sub>3</sub>S, C<sub>3</sub>A, C<sub>4</sub>AF, and pre-hydrated C<sub>3</sub>S and pre-hydrated C<sub>3</sub>A by adding lead nitrate (Pb(NO<sub>3</sub>)<sub>2</sub>). Although C<sub>3</sub>A showed low sorption and consequently resulted in lead oxide (PbO) precipitation, the phases like C<sub>4</sub>AF, pre-hydrated and unhydrated C<sub>3</sub>S showed higher Pb affinity. Up to 0.03 mmol of Pb were adsorbed on 1 g of unhydrated C<sub>3</sub>S, and this high adsorption could stop the C<sub>3</sub>S hydration [17]. The OH<sup>-</sup> species in PCF can deplete because of its tendency to naturally carbonate in ambient conditions [15,18]. However, this carbonation improved metals fixation in cement-solidified hazardous wastes [19].

The CO<sub>2</sub> ingress in cement creates chemical imbalance between pore solution and hydrates, and thus primarily turns portlandite into calcite, in the following order (1) portlandite dissolution, (2) CO<sub>2</sub> absorption and carbonate ions formation, and (3) chemical reaction and precipitation [20,21]. Next to portlandite, the remaining phases AFt, AFm and C-S-H also carbonate. The net effect of carbonation is a reduction in the pH of hydrated cement, and thus it results in rebar depassivation, which is a major concern in case of structural concrete. However, this seemingly negative effect can potentially be used to improve the metals fixation in concrete-based filters. This CO<sub>2</sub> treatment can also reduce the leaching of fixated metals from PCF. Pandey et al. [22] demonstrated the utilization of carbonated cement for immobilizing Cd, Cr(III), Pb and Zn unlike Cu. These fixated metals could bond strongly with carbonated C-S-H [23-26], which in-turn could decrease the degree of metal leaching from cement-solidified waste [27]. Van Gerven et al. [28] pointed out that this positive effect could reverse when the cement was fully carbonated, in which the carbonate species turn into weak bicarbonates. The choice of accelerated carbonation technology is preferable for PCF and its application. It is a controlled accelerated form of the naturally occurring process, where the green product, i.e. cement is exposed to a gaseous CO<sub>2</sub>-rich environment for shorter period and thereafter it rapidly stiffens into a structural medium, can strongly fixate the heavy metal ions, and also provides the way for CO<sub>2</sub> sequestration [29]. The logarithm of the stability constants for the formation of the solid hydroxide and carbonate species for Pb are 10.9 and 13.1, as reported in Stumm and Morgan [30], indicating a higher affinity for the formation of lead carbonate than lead hydroxide solids.

This study was focussed to investigate the effects of accelerated carbonation and HRT on the Pb removal in PCF. The choice of Pb for this study was mainly because of its potential to strongly adsorb in cement-based PCF, which has been previously demonstrated by many researchers successfully in the literature [10,14,31]. However, this experimental study for the first time proposed a filtration arrangement comprising three PCF monoliths (100 mm diameter and 150 mm length each) that were arranged in series with progressively decreasing levels of water heads (H, H/4 and trickling head). The Pb containing solution of 8 mg/L concentration simulating the real-field environment was constantly passed through this arrangement for 60 min. This filtration experiment was conducted on PCFs, which were tested right after initial curing as well as after subjecting the concrete elements to an accelerated carbonation exposure. As a consequence, this investigation can provide valuable insight into the use of accelerated carbonation technology in PCF for Pb removal, and additionally recommends the need for HRT consideration in the concrete filter design for real-field applications.

On the other hand, the effect of different solvents (dilute HCl solution or NaOH solution or tap water or distilled water) on the leaching characteristics of 7-day cured and carbonated PCFs post-treatment were investigated, and thus it can provide useful information on limiting the solvent types that can be treated effectively using these PCFs.

#### 2. Materials and methods

#### 2.1. Specimen preparation and accelerated carbonation exposure

53 Grade Portland cement (62.16 wt% CaO, 20.95 wt% SiO<sub>2</sub>, 5.14 wt% Al<sub>2</sub>O<sub>3</sub>, 3.06 wt% Fe<sub>2</sub>O<sub>3</sub>, 3.08 wt% SO<sub>3</sub>, 1.33 wt% MgO, 0.009 wt% Cl, 1.63 wt% loss on ignition, and 2.63 wt% insoluble residue) and crushed granite coarse aggregate conforming to IS 12269 [32] and IS 383 [33] respectively, distilled water and polycarboxylic ether (PCE)-based superplasticizer were used in the preparation of PCF monolith whose proportions are presented in Table 1.

The specific gravity (saturated surface dry condition) and water absorption of coarse aggregate were found to be 2.77 and 0.42% as per the methods specified in IS 2386-3 [34]. For experimental investigations on PCF, cast iron moulds (100 mm diameter and 200 mm length) were used in the monolith preparation. The compaction was performed with a Marshall Hammer having foot diameter, drop weight and free fall of 98.4 mm, 4.54 kg and 457 mm. The fresh concrete was filled into the mould in three layers, which were weighed exactly before filling. Twenty-six blows were given using the Marshall Hammer after each layer of fresh concrete was poured inside mould.

For better compaction of the top layer, a collar arrangement was added to the top of iron mould, in order to avoid the spillage of the concrete outside the mould. After the compaction of top layer, the surface was carefully finished using trowel [19]. The monoliths were demolded from moulds after 24 h and then cured in tap water at room temperature  $(25 \pm 2 \circ C)$  for 7 days. The bleed water from fresh concrete used to fill the coarser voids in top and bottom sections of monolith, and therefore the actual permeability could be affected, which was resolved by removing top and bottom slices of 25 mm thick from 7-day cured monolith using diamond-tipped saw. This sectioned PCF monolith (100 mm diameter and 150 mm length) was then air dried at 25 °C and 65% R.H. for the next 14 days, and consequently introduced into the carbonation chamber, which was controlled at 25 °C, 65% R.H. and 1% CO<sub>2</sub> concentration for 56 days. The PCF after this CO<sub>2</sub> treatment was referred as 'carbonated' in this paper.

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