

# Natural Coal Aggregates to the Rescue of Fe<sup>0</sup>-Bed Filters in Quaternary Reactive Zones Fe<sup>0</sup>/S/Pz/C<sub>x</sub> to Repel Clogging and Boost Reactivity

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**Abstract:** The absorption and remediation power of natural coal (NC) and its contribution to the harvesting of corrosion products (CPs) resulting from electrochemical processes of Fe<sup>0</sup> oxidation in the reactive zone (RZ) of Fe<sup>0</sup>/S/Pz/C (Iron/Sand/Pozzolan/Natural Coal) in Fe<sup>0</sup>-bed filters have now been proven. As a result, an in-depth study of NC varieties (C<sub>x</sub>) for increased performance of Fe<sup>0</sup>/S/Pz/C<sub>x</sub> (Iron/Sand/Pozzolan/Natural Coals varieties) for water quality was investigated. Nine (9) systems were used, with respective RZs consisting of (1) Fe<sup>0</sup> (Pure Iron), (2) AC (pure Ayous wood's Coal), (3) CC (pure Coconut bark's Coal), (4) MC (pure Movingui wood's Coal), (5) PC (pure Padouk wood's Coal), (6) Fe<sup>0</sup>/S/Pz/AC (Iron/Sand/Pozzolan/Ayous wood's Coal), (7) Fe<sup>0</sup>/S/Pz/CC (Iron/Sand/Pozzolan/Coconut bark's Coal), (8) Fe<sup>0</sup>/S/Pz/MC (Iron/Sand/Pozzolan/Movingui wood's Coal) and (9) Fe<sup>0</sup>/S/Pz/PC (Iron/Sand/Pozzolan/Padouk wood's Coal). OM (orange methyl) of 2 mg/L concentration was used as operative indicator. The experiments lasted forty (40) days per device. We measured the pH, discoloration, dissolved iron content of the collected water, while observing the flow rate and stability of every Fe<sup>0</sup>/S/Pz/C<sub>x</sub>. Thereby, the beneficial effects of the Pz/C<sub>x</sub> combination in the RZ are proven regardless of the variety used. Thus, CC is the best absorbent material almost as good as Fe<sup>0</sup>. PC has the best throughput and therefore the most permeable. All varieties contribute to the minimization of iron released. The pH is in line with WHO recommendations. Fe<sup>0</sup>/S/Pz/C<sub>x</sub> discoloration's performance is thus evolving: Fe<sup>0</sup>/S/Pz/CC > Fe<sup>0</sup>/S/Pz/AC > Fe<sup>0</sup>/S/Pz/MC > Fe<sup>0</sup>/S/Pz/PC > pure Fe<sup>0</sup>; the flow changes inversely. With 98% ≤ 25% Fe<sup>0</sup> / 25% S / 25% Pz / 25% CC ≤ 100% discoloration rate, Fe<sup>0</sup>/S/Pz/CC is the most stable over time. CC appears to be the best non-expansive porous material to be combined with the ternary Fe<sup>0</sup>-based filters, and widely with Fe<sup>0</sup>/H<sub>2</sub>O systems to allow a resurgence of efficiency, to delay clogging and extend lifespan.

**Keywords:** Aqueous Corrosion, Ayous, Fe<sup>0</sup>-bed Filters, Coal, Coconut's Bark, Movingui, Orange Methyl, Padouk

## 1. Introduction

The technology of metal bed filters, which also aims to provide drinking water to many populations far from distribution networks, is increasingly being developed [1-4]. In fact, the filtering power of the zero-valent iron (Fe<sup>0</sup>) and widely Fe<sup>0</sup>-filters, is associated with corrosion products (CPs) generated by the electrochemical process of iron oxidation. CPs are pollutant collectors that allow the elimination of many metals, bacteria and viruses [5-10]. However, pure iron devices are susceptible to clogging [11-

14]. The work of Btatkeu et al., and other authors [15-20] have shown the need to unclog the reactive surface, notably by the Fe<sup>0</sup>/S (Iron/sand) involvement, thanks to the responsiveness of the Si-OH groups present in the silica (SiO<sub>2</sub>). This mixed efficiency has led to the combination of a porous absorbent/adsorbent and non-expansive material, through a Fe<sup>0</sup>/S/Pz filter (Iron/Sand/Pozzolan) capable of collecting these CPs [21]. The effectiveness of this ternary device, its stability and its lifespan can be greatly improved by the concomitant combination of two porous and non-expansive materials in the same reactive zone (RZ).

Indeed, joining Pz and natural coal (NC) in a quaternary device  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}$  (Iron / Sand / Pozzolan / Natural Coal) showed an excellent efficiency rate to discoloration in MO (orange methyl) of 98-100% and stability of the device, since  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C} > \text{Fe}^\circ/\text{S}/\text{C} > \text{Fe}^\circ/\text{S}/\text{Pz} > \text{Fe}^\circ/\text{C} > \text{Fe}^\circ/\text{Pz} > \text{C} > \text{Pz}$  [22]. The proportions of materials in the RZ are established taking into account the fact that  $25\% \leq \text{Fe}^\circ \leq 60\%$ ,  $25\% \leq \text{S} \leq 50\%$ ,  $25\% \leq \text{Pz} \leq 50\%$  [23]. The hope generated by the  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}$ , requires a widening of the scope of possibilities by the study of different varieties of coals ( $\text{C}_x$ ) for finding a good absorbent, which is also a means of addressing the difficulty of sourcing Pz [22, 24-25].

Hence, the coconut whose shell is widely used in the form of a natural coal (CC) for many virtues in the fields of health, environment and aesthetics is experienced here. Coconut shells abound on many African beaches, and constitute agricultural pollution. As part of waste recovery, a comparative study of the reactivity of four coals obtained from coconut shells was carried out and confirms the adsorption properties conferred on them. [26]. Since the use of NC as an energy source is part of the daily lives of many African households, it is available [22, 27]. The study of its physicochemical characteristics reveals high porosity and absorption capacity [28, 29]. Wood is a cellular, hygroscopic, anisotropic and compact, more or less hard material that makes up the trunk, branches, and roots of trees and shrubs; it is biodegradable [30]. Some species are abundant in tropical forests, such as Ayous, Moabi, Movingui, Padouk and Tali varieties [31]. Only Ayous (AC) variety, Movingui (MC) and Padouk (PC) were tested here. AC, or *triplochiton scleroxylon*, from the Sterculiaceae family, is a light wood that dries quickly in the air [32]. MC or *Distemonanthus benthamianus* of the leguminosae family is a hardwood used as firewood and for coal production [33]. PC or *Pterocarpus soyauxii*, is a strong and hard wood [34].

The present work is aimed at analyzing the different  $\text{C}_x$ ,

namely (1) AC, (2) CC, (3) MC and (4) PC in order to characterizing  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$  as a safe alternative to the pure  $\text{Fe}^\circ$ , and  $\text{Fe}^\circ$ -based filters, but also to find the best  $\text{C}_x$  as a non-expansive porous material for such a  $\text{Fe}^\circ$ -quaternary system, while using OM discoloration method. The nine (9) following systems are investigated: (1) 100%  $\text{Fe}^\circ$ , (2) 100% AC, (3) 100% CC, (4) 100% MC, (5) 100% PC, (6) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% AC, (7) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% CC, (8) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% MC, (9) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% PC. The performance parameters that are pH, released iron, discoloration, and throughput are measured.

## 2. Materials and Methods

### 2.1. Solutions

OM is used in acid-base dosages as a colored indicator. Its cornering zone is between [3.1-4.4] and allows to mark the presence of an acid medium, it turns red, or a basic medium, it turns yellow. It naturally has a great affinity for solid surfaces of opposite loads such as certain  $\text{Fe}^\circ$ -CPs [35, 36]. The OM comes from KEM LIGHT PVT Laboratories. LTD, Mumbai, India. The solution used has a concentration of 2.0 mg/L; the pH value is 5.2, this concentration is chosen to be close to natural pollution conditions [37]. A standard iron solution, 990  $\mu\text{g}/\text{mL}$  from Aldrich Chemical Company, Inc. (Milwaukee, WI, USA) was used to calibrate the spectrophotometer. The L (+) -ascorbic acid from E. MERCK, Darmstadt. 90% ethanol; sodium acetate from ANALAR; 1.10 o-phenanthroline from NORMAPUR used as a reagent for  $\text{Fe}^{2+}$  complexation require for spectrophotometric reading, 0.2 g/L concentration [38-40].

### 2.2. Solid Materials

Seven materials were used to carry out this work. All samples are from Cameroon.

**Table 1.** Symbol, Granulometry, source, and nature of the materials used - <sup>1</sup>Communal Market, <sup>2</sup>North Region, <sup>3</sup>South West Region, <sup>4,5</sup>Coastal Regions.

N°	Materials	Symbol	Granulometry	Source	Nature
1	Iron	$\text{Fe}^\circ$	$\leq 1 \text{ mm}$	Collected (CM <sup>1</sup> )	Adsorbent Generator
2	Sand	S	1 mm	Collected (NR <sup>2</sup> )	Adsorbent
3	Pozzolan	Pz	2 mm	Collected (SWR <sup>3</sup> )	Porous Absorbent / Adsorbent
4	Coconut bark's Coal	CC	2 mm	Collected (CR <sup>4</sup> )	Porous Absorbent / Adsorbent
5	Ayous wood's Coal	AC	2 mm	Collected (CR <sup>5</sup> )	Porous Absorbent / Adsorbent
6	Movingui wood's Coal	MC	2 mm	Collected (CR <sup>5</sup> )	Porous Absorbent / Adsorbent
7	Padouk wood's Coal	PC	2 mm	Collected (CR <sup>5</sup> )	Porous Absorbent / Adsorbent

#### 2.2.1. Metal Iron

The iron used in this work is iron wool made by the steel mills of Cameroon (Douala, Cameroon) and marketed in the various local markets; its granulometry is less than 1 mm. This material has shown its effectiveness in discoloring methylene blue [41]. It is used without treatment. The X-ray fluorescence analysis reveals: 0.62% Mn, 0.52% Si, 0.23% Cu, 0.2% Cr, and 0.09% Ni.

#### 2.2.2. Sand

Sand (S) is a conductive natural aggregate used to modify

the hydraulic conductivity of geotechnical systems. The sand used herein is a natural material taken in the Vina River (Cameroon), washed and rinsed with water boiled at 100°C for 3 hours and then dried at 110°C for 4 hours, it constitutes the different layers  $L_1$  (upper),  $L_2$  (intermediate or RZ) and finally  $L_3$  (lower). For its availability and mixing agent, sand was used in  $\text{Fe}^\circ/\text{H}_2\text{O}$  systems [42-43]. The average chemical composition per X-ray diffraction reveals: 81.5%  $\text{SiO}_2$ , 5.60%  $\text{Al}_2\text{O}_3$ , 4.71%  $\text{Fe}_2\text{O}_3$ , 3.86%  $\text{CaO}$ , 1.75%  $\text{TiO}_2$ , 0.91%  $\text{K}_2\text{O}$ , 0.48%  $\text{P}_2\text{O}_5$ , 0.26%  $\text{SO}_3$ , 0.32%  $\text{MnO}$ , 0.08%  $\text{SrO}$ , 0.03%  $\text{V}_2\text{O}_5$ .

### 2.2.3. Pozzolan

The pozzolan (Pz) used comes from Idenau (Southwest, Cameroon) and has undergone the same pre-treatment as sand. It is L<sub>2</sub> layer (RZ). Pozzolan has a porosity of 60% which serves as a reservoir for Fe<sup>o</sup>CPs [21, 23, 44-45]. and has adsorption and absorption properties. The average chemical composition per X-ray diffraction is: 81.18% SiO<sub>2</sub>, 10.00% Al<sub>2</sub>O<sub>3</sub>, 2.19% Fe<sub>2</sub>O<sub>3</sub>, 0.59% CaO, 0.46% TiO<sub>2</sub>, 3.60% K<sub>2</sub>O, 0.05% MnO, 0.02% SrO, 0.02% ZrO<sub>2</sub>.

### 2.2.4. Natural Charcoal

As part of this work, coconut shells come from the reserves of farmers in Bakoko village in Cameroon's coastal regions. The Ayous, Movingui and Padouk wood samples also come from the same wooded area of Bakoko village. The wood is made up of three (3) main constituents: cellulose, lignin and water. Cellulose and lignin, plus other substances, are strongly linked to form the material called wood. The first step in carbonization is drying at 100°C until the anhydrous state. The temperature of the dry wood is then raised to about 280°C, a temperature at which the dry wood begins to decompose spontaneously to give charcoal. At 500°C, the carbon content is 85%, and 10% of volatile elements. The carbonization was carried out in the coal mines of the local market, with temperature regulators. The resulting coals were calibrated as mentioned in Table 1 and have undergone the same pre-treatment (washed and dried) as the S and the Pz. It is the L<sub>2</sub> layer (RZ). Hence, pure Coconut bark's CC, is studied here. AC, or *triplochiton scleroxylon*, from the Sterculiaceae family, is a light wood that dries quickly in the air.

MC or *Distemonanthus benthamianus* of the leguminosae family is a hardwood used as firewood and for coal production. PC or *Pterocarpus soyauxii*, is a strong and hard wood. Adsorption tests were performed on all samples outside the filters. However, only the performance of the RZs is taken into account in this manuscript Table 2 below. The adsorption tests performed are used to assess the amount of adsorbed.

$$Q_e = \left( \frac{Co - Cr}{m} \right) \times V$$

Q<sub>e</sub> is the amount adsorbed per gram of coal in mg/g. Cr is the concentration of the species in solution at the moment t in mg. Co is the initial concentration of the solution in mg/L. V is the volume of the solution initially introduced in L. Mass (m) of the adsorbent in g [22, 26, 28-34, 46-48].

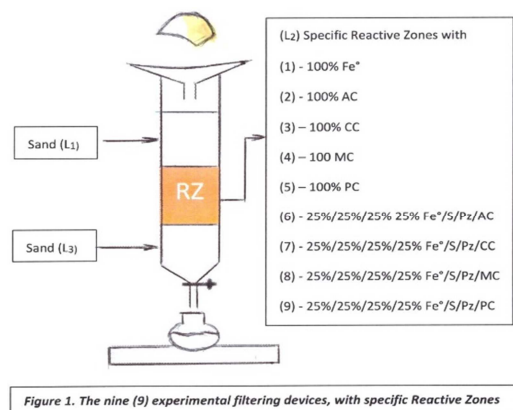
The chemical composition is determined by the contribution of MIPROMALO (Mission for the Promotion of Local Materials) in Yaounde- Cameroon.

### 2.3. Procedure

We experimented with nine (9) filter devices including two (5) 100% material devices and four (4) quaternary devices. The column is made of polyethylene and each filter has a total material mass of 220 g, including the top layer L<sub>1</sub> (m = 60 g of sand), intermediate layer L<sub>2</sub> (m = 40 g RZ) and lower layer L<sub>3</sub> (m = 120 g of sand). All devices are equal in total mass of materials.

**Table 2.** Composition of the reactive zone (RZ) of each filtering device. The masses of materials are expressed in gram (g) and percentage (%).

N°	Devices	Fe <sup>o</sup> (g)	Fe <sup>o</sup> (%)	S (g)	S (%)	Pz (g)	Pz (%)	AC (g)	AC (%)	CC (g)	CC (%)	MC (g)	MC (%)	PC (g)	PC (%)
1	Fe <sup>o</sup>	40	100	00	00	00	00	00	00	00	00	00	00	00	00
2	AC	00	00	00	00	00	00	40	100	00	00	00	00	00	00
3	CC	00	00	00	00	00	00	00	00	40	100	00	00	00	00
4	MC	00	00	00	00	00	00	00	00	00	00	40	100	00	00
5	PC	00	00	00	00	00	00	00	00	00	00	00	00	40	100
6	Fe <sup>o</sup> /S/Pz/AC	10	25	10	25	10	25	10	25	10	25	10	25	10	25
7	Fe <sup>o</sup> /S/Pz/CC	10	25	10	25	10	25	10	25	10	25	10	25	10	25
8	Fe <sup>o</sup> /S/Pz/MC	10	25	10	25	10	25	10	25	10	25	10	25	10	25
9	Fe <sup>o</sup> /S/Pz/PC	10	25	10	25	10	25	10	25	10	25	10	25	10	25



**Figure 1.** The nine (9) experimental filtering devices, with specific Reactive Zones.

L<sub>1</sub> ensures a distribution of raw water over the entire filtering device, decreases the energy or strength of the incoming contaminated water that can disturb the reactive layer, ensures the constant height of the polluted water; L<sub>3</sub>, however avoids the phenomenon of air bubbles under the RZ, regularize or improve the flow [49], Figure 1.

### 2.4. Analytical Methods and Experimental Results

The wavelength for the iron solution is 510 nm, the tanks used are 1.0 cm. Residual iron is determined according to the 1.10 Orthophenanthroline protocol. The OM content in the filter is determined by UV-visible spectrophotometer at λ= 461nm Ling type 9100-9400 whose spectral range extends over a range ranging from 320-1100 nm. The effectiveness in discoloring the initial OM filter (c<sub>o</sub>) depending on the residual OM content, the effectiveness (E) to discoloration is

given by the relationship:

$$E(\%) = \left(1 - \frac{C_e}{C_0}\right) \times 100.$$

The pH values are obtained by an electrode pH (WTW Co, Weilheim, Germany) [38-40, 50].

### 3. Results and Discussion

#### 3.1. Discoloration (Absorption / Adsorption) and Mitigation of Iron Release with $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_X$

Figures 2, 3, 4 and 5, respectively, depict discoloration in OM (2 mg/L) and iron released in filtered water for single-material RZs, (1) 100%  $\text{Fe}^0$ , (2) 100% AC, (3) 100% CC, (4) 100% MC, (5) 100% PC, and four-material RZs (6) 25%  $\text{Fe}^0$  / 25% S / 25% Pz / 25% AC, (7) 25%  $\text{Fe}^0$  / 25% S / 25% Pz / 25% CC (8) 25%  $\text{Fe}^0$  / 25% S / 25% Pz / 25% MC and (9) 25%  $\text{Fe}^0$  / 25% S / 25% Pz / 25% PC. These results show that the pure RZs provide only partial remediation, about 70% for the pure  $\text{Fe}^0$ -filter; 40%  $\leq C_X \leq 65\%$ , and  $\text{CC} > \text{AC} > \text{MC} > \text{PC}$ . There is a loss of efficiency beyond twenty-five (25) days. The iron released is however high for the pure  $\text{Fe}^0$ -system, and negligible for coals. When the RZs are made up of  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_X$ , there is a renewed responsiveness, such as  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC} > \text{Fe}^0/\text{S}/\text{Pz}/\text{AC} > \text{Fe}^0/\text{S}/\text{Pz}/\text{MC} > \text{Fe}^0/\text{S}/\text{Pz}/\text{PC}$ , and the discoloration rate intervals are  $90\% \leq \text{Fe}^0/\text{S}/\text{Pz}/\text{C}_X \leq 100\%$ , and  $98\% \leq \text{Fe}^0/\text{S}/\text{Pz}/\text{CC} \leq 100\%$ , for the best quaternary filtering system. The iron released in the filtered water is significantly reduced. There is no loss of reactivity during the forty days.

Actually, such a result is an excellent report on the contribution of the RZ nature over the lifetime of the filtering system. These results are consistent with the disposal process of  $\text{Fe}^0$ -based filters. Pure, they are prone to rapid clogging [55-57]; associated with S [20], Pz [16, 21, 23, 44-45, 51-52], or CN [22, 28-29, 53-54], they are improved [11-14, 21-23]. The reactivity of the  $\text{Fe}^0$ , and largely that of  $\text{Fe}^0$ -based filters, involves electrochemical reactions of the wet corrosion of iron, and generates adsorbents that collect contaminants responsible for the discoloration. Some studies were found that the presence of different functional groups in the dyes such as OM, can be an important factor for selective interaction with iron oxide nanoparticles [9, 18, 21, 58]. The combination of two non-expansive porous materials that use their absorbent/adsorbent properties to serve the system as receptacles for  $\text{Fe}^0\text{CPs}$ , makes  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_X$  stable devices, which allow to divide by four (4) the proportion of  $\text{Fe}^0$  in the RZ (Figures 2, and 3) and in the filtered water (Figures 4 and 5), while the rate of discoloration increases from 70% ( $\text{Fe}^0$ ) to 100% ( $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC}$ ) [22]. The iron released comes from  $L_1$  and  $L_3$  for systems (2), (3), (4), and (5); it comes from  $L_1$ ,  $L_2$ , and  $L_3$  for systems (1), (6), (7), (8) and (9). Even though the pH value significantly impacts the extent of  $\text{Fe}^0$  dissolution, it has been demonstrated elsewhere and refs cited therein that in this pH range, the solubility of iron is minimal [65]. So, S, Pz, AC, CC, MC, and PC are not only admixing agent in  $\text{Fe}^0/\text{H}_2\text{O}$  systems, but they actually contribute to the elimination process as explained above. Therefore, CC's

absorption capacity makes  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC}$  an excellent device. AC comes from a light wood that dries quickly, and seems to be a good absorbent. The  $\text{Fe}^0/\text{S}/\text{Pz}/\text{AC}$  is stable and efficient. Hardwoods such as MC and PC produce fewer absorbent coals,  $\text{Fe}^0/\text{S}/\text{Pz}/\text{MC}$  and  $\text{Fe}^0/\text{S}/\text{Pz}/\text{PC}$  are just as stable over time [26, 30-34], but with discoloration rates slightly below those of CC and AC.  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC}$  is the best rate quaternary filtering system,  $98\% \leq \text{Fe}^0/\text{S}/\text{Pz}/\text{CC} \leq 100\%$ , making it a safe alternative for the  $\text{Fe}^0/\text{H}_2\text{O}$ , and  $\text{Fe}^0$ -bed filters.

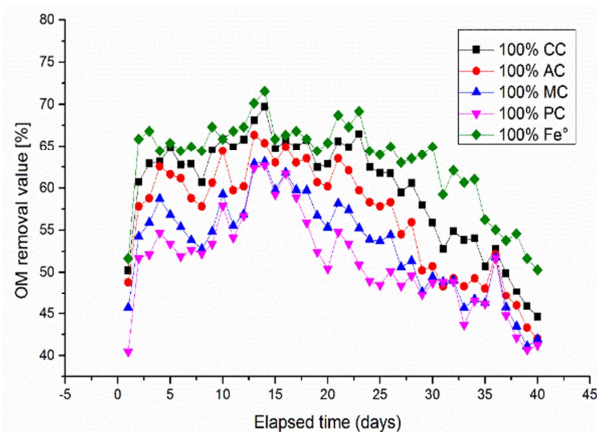


Figure 2. OM removal values, one-material RZs with pure AC, CC, MC and PC compared to pure  $\text{Fe}^0$  RZ.

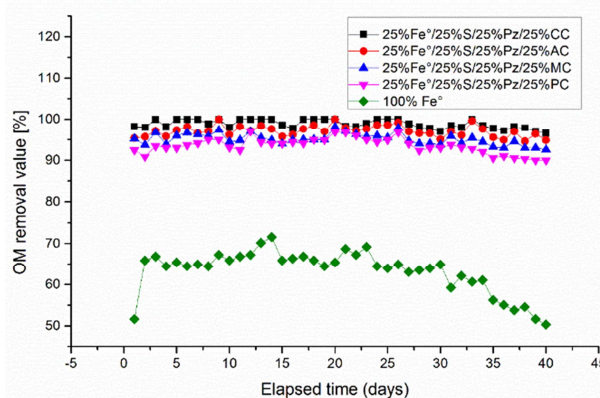


Figure 3. OM removal values, quaternary RZs with  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_X$  compared to pure  $\text{Fe}^0$  RZ.

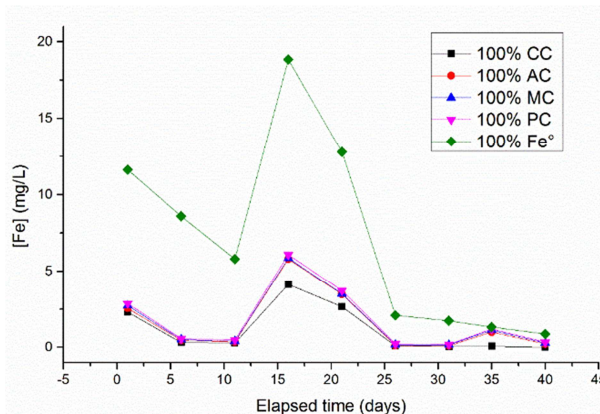
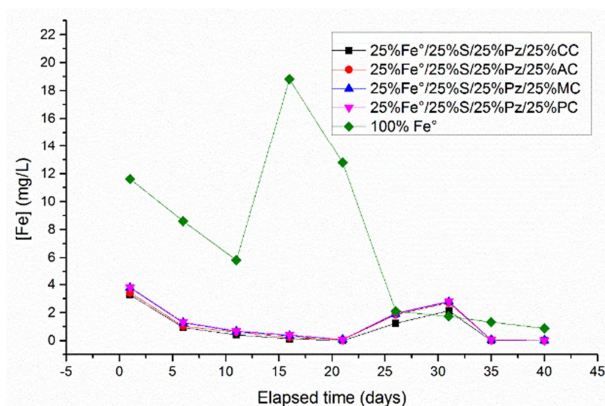


Figure 4. [Fe] released, one-material RZs with pure AC, CC, MC and PC compared to pure  $\text{Fe}^0$  RZ.



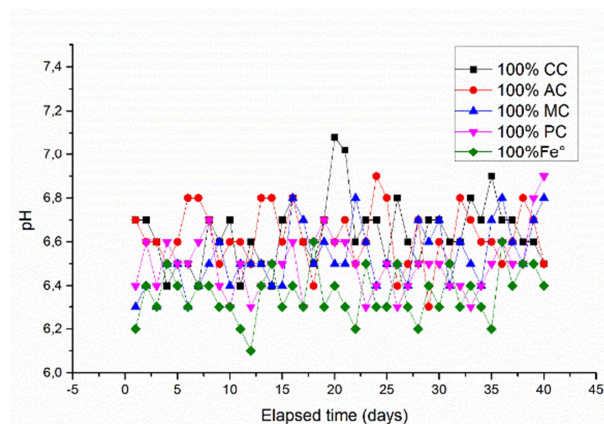


**Figure 5.**  $[\text{Fe}]$  released values, quaternary RZs with  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$  compared to pure  $\text{Fe}^\circ$  RZ.

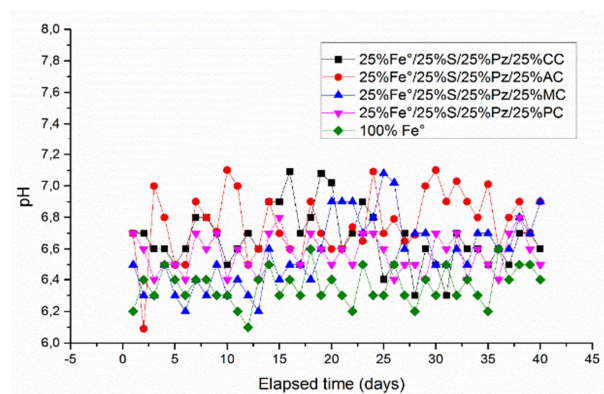
### 3.2. Study of pH and Performance of $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$

Figures 6 and 7 depict the time-dependent pH, measured for forty (40) days from nine (9) experimental systems using OM (2 mg/L). Specific RZs are (1) 100%  $\text{Fe}^\circ$ , (2) 100% AC, (3) 100% CC, (4) 100% MC, and (5) 100% PC, and RZs of four (4) materials (6) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% AC, (7) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% 25% CC, (8) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% MC and (9) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% PC. The initial pH is 5.2. We are seeing an increase in pH for the nine (9) systems studied, but more for  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$  with material ratio four (4) times lower [38-39].

The increase in pH is the result of the elimination of OM. In fact, the RZs are made up of  $\text{Fe}^\circ$ , S, Pz and  $\text{C}_x$  or four (4) at once.  $\text{Fe}^\circ\text{CPs}$  are adsorbents that participate in decontamination processes [11-14, 18, 21-23, 37]. RZs contain sand (S) that is provided by adsorption, and an affinity for opposite load species such as protons and vice versa [20-23, 55-57, 66]. Pz reacts by absorption/adsorption due to its very high porosity [16, 44-45, 51-52, 21-23, 66]. According to some authors, low pH values may be due to acidification generated by the dissolution of  $\text{SiO}_2$  since S and Pz are mainly made of it. This does not seem to have a significant impact on systems studied since quaternary systems contain  $\text{SiO}_2$  in  $\text{L}_1$ ,  $\text{L}_2$  and  $\text{L}_3$ , unlike single-material systems which contain it only in  $\text{L}_1$  and  $\text{L}_3$ . As for Coals, CC, AC, MC, and PC, they react by absorption/adsorption, due to their porous structure [22, 26, 28-29, 30-34, 53-54, 66]. All  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$  systems have a higher pH than pure  $\text{Fe}^\circ$ . Some authors attribute the increase in pH only to  $\text{Fe}^\circ\text{CPs}$ . This seems partially true since all one-material systems also show an increase in pH, Figure 6. In addition, it is clear that the reduction in the  $\text{Fe}^\circ$  rate from 100% to 25% in all  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$  systems always ensure the increase in pH, which then goes from an initial pH of 5.2 to an average pH of 7, especially for  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{CC}$  and  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{AC}$ , Figure 7. Hence the interest of RZs with four (4) materials but with good adsorbents/absorbents like CC or light woods like AC.  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{CC}$  and  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{AC}$  appear to be a safe alternative to the  $\text{Fe}^\circ/\text{H}_2\text{O}$  and  $\text{Fe}^\circ$ -bed filters in general. This combination of the four materials (4) is undeniably beneficial and requires four (4) times less  $\text{Fe}^\circ$ .



**Figure 6.** pH values, one-material RZs with pure AC, CC, MC and PC compared to pure  $\text{Fe}^\circ$  RZ.



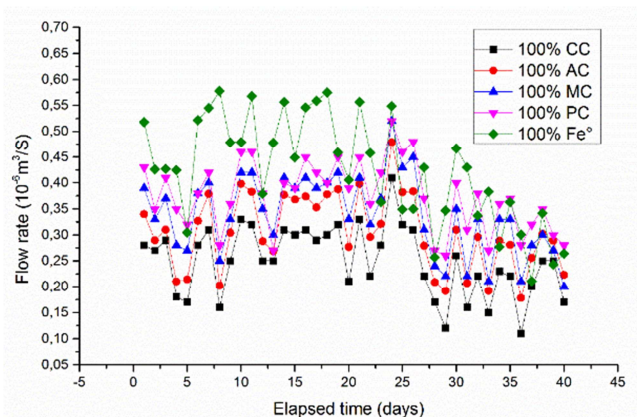
**Figure 7.** pH values, quaternary RZs with  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$  compared to pure  $\text{Fe}^\circ$  RZ.

### 3.3. The Performance of $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{C}_x$ and Flows of Different coal Varieties

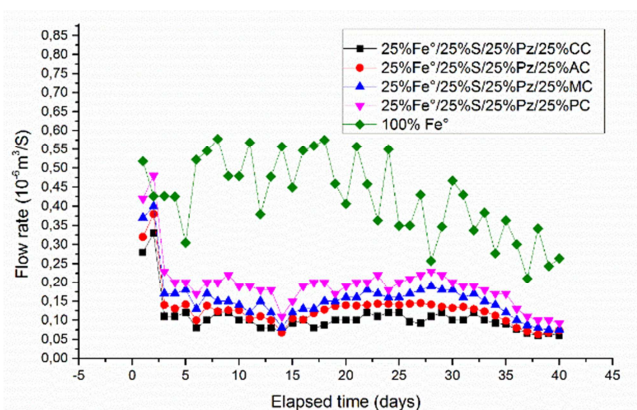
Figures 8 and 9 depict the flows of nine experimental systems using of OM (2 mg/L). Specific RZs are (1) 100%  $\text{Fe}^\circ$ , (2) 100% AC, (3) 100% CC, (4) 100% MC, (5) 100% PC, (6) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% AC, (7) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% CC, (8) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% MC and (9) 25%  $\text{Fe}^\circ$  / 25% S / 25% Pz / 25% PC. We observed the increasing order of flow rates as followed: Flow (100% CC) < Flow (100% AC) < Flow (100% MC) < Flow (100% PC) < Flow (100%  $\text{Fe}^\circ$ ) and Flow ( $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{CC}$ ) < Flow ( $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{AC}$ ) < Flow ( $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{MC}$ ) < Flow ( $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{PC}$ ) < Flow (100%  $\text{Fe}^\circ$ ).

The gradual drop in flow is observed on all single-material systems in the RZ, (1), (2), (3), (4) and (5) with a curve deflection beyond the twenty-fifth day. The decrease of flow is attributed to the progressive clogging due to accumulation of OM in the initial pores and interstices systems. It is now agreed that pure  $\text{Fe}^\circ$  systems are efficient but not sustainable [67, 68]. The replacement of a fraction of  $\text{Fe}^\circ$  by a non-expansive porous material was therefore found as alternative approach [21-23, 69, 70]. Testing another material in  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{AC}$ ,  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{CC}$ ,  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{MC}$  and  $\text{Fe}^\circ/\text{S}/\text{Pz}/\text{PC}$  shown that AC, CC, MP, PC and Pz enable to in situ control of expansive  $\text{Fe}^\circ\text{CPs}$  which are the main cause of permeability loss. Indeed, the best absorbents (CC and AC)

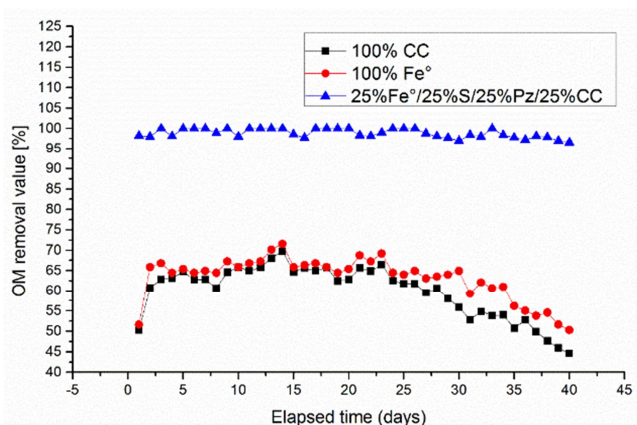
have the lowest flows, Figures 2, 3, 8 and 9. But  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_x$  become more stable, and the lifespan is extended. Hard and resistant woods such as MC and PC, contribute more to this effect. In addition, the proportions of materials are four times lower in quaternary systems, which put these differences into perspective, and highlight the benefit. The flow rate can be greatly improved by offering the systems a larger reactive surface, while retaining the same proportions of the materials already establish here.



**Figure 8.** Flow rate values, one-material RZs with pure AC, CC, MC and PC compared to pure  $\text{Fe}^0$  RZ.



**Figure 9.** Flow rate values, quaternary RZs with  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_x$  compared to pure  $\text{Fe}^0$  RZ.



**Figure 10.** Performance of the quaternary device 2 %/25%/25%/25%  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC}$  for OM discoloration.

Figure 10 shows that the  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC}$  is an excellent filtering system. Lonely in the RZ, 100%  $\text{Fe}^0$  and 100% CC are less efficient, with a loss of net reactivity from the twenty-fifth day. Associated with S, Pz and CC, in the same proportions, there is a resurgence of reactivity and a delay in clogging. Coconut bark is an excellent porous material.

## 4. Conclusion

This work consisted, through the enhancement of local materials, of studying different natural coal aggregates for the optimization of the performance of  $\text{Fe}^0$ -bed filters in quaternary reactive zones  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_x$ , with a view to repel clogging and boost reactivity. Therefore, we have introduced in respective RZs,  $\text{Fe}^0$ , S, Pz, AC, CC, MC and PC. Nine (9) devices were investigated using OM for forty (40) days: (1)  $\text{Fe}^0$ , (2) AC, (3) CC, (4) MC, (5) PC, (6)  $\text{Fe}^0/\text{S}/\text{Pz}/\text{AC}$ , (7)  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC}$ , (8)  $\text{Fe}^0/\text{S}/\text{Pz}/\text{MC}$  and (9)  $\text{Fe}^0/\text{S}/\text{Pz}/\text{PC}$ . The results obtained show that lonely  $\text{Fe}^0$ , AC, CC, MC and PC provide partial OM discoloration, respectively. When associated with the  $\text{Fe}^0$  in RZs  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_x$ , there is a resurgence of responsiveness. The AC, a light wood that dries quickly, is an excellent absorbent, but CC is the best material for a  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC}$ . As a result, the filtering power evolves according to the decreasing order as follows:  $\text{Fe}^0/\text{S}/\text{Pz}/\text{CC} > \text{Fe}^0/\text{S}/\text{Pz}/\text{AC} > \text{Fe}^0/\text{S}/\text{Pz}/\text{MC} > \text{Fe}^0/\text{S}/\text{Pz}/\text{PC} > \text{pure } \text{Fe}^0$ . The OM discoloration rate of  $98\% \leq 25\% \text{Fe}^0 / 25\% \text{S} / 25\% \text{Pz} / 25\% \text{CC} \leq 100\%$ , makes CC the best non-expansive porous material to associate with  $\text{Fe}^0/\text{S}/\text{Pz}$  ternary systems, to delay clogging, and extend lifespan. However, AC, which comes from a light wood, also gives very good results. MC and PC derived from hardwood and hard-resistant, offer the best flow. All varieties contribute to the minimization of iron released. The pH is in line with WHO recommendations.  $\text{Fe}^0/\text{S}/\text{Pz}/\text{C}_x$  can be used as a safe alternative to the pure  $\text{Fe}^0$ , and  $\text{Fe}^0$ -based filters. There appears to be a dual ecological and economic interest. It is an excellent way to revalue the agricultural waste that is coconut shells. It is also a way to give coals a second life while providing drinking water to people far from the distribution networks.

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