



Materials for sustainable metallic iron-based water filters: a review

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Abstract

Water pollution is calling for a sustainable remediation method such as the use of metallic iron (Fe^0) to reduce and filter some pollutants, yet the reactivity and hydraulic conductivity of iron filters decline over time under field conditions. Here we review iron filters with focus on metallic corrosion in porous media, flaws in designing iron filters, next-generation filters and perspectives such as safe drinking water supply, iron for anaemia control and coping with a reactive material. We argue that assumptions sustaining the design of current Fe^0 filters are not valid because proposed solutions address the issues of declining iron reactivity and hydraulic conductivity separately. Alternatively, a recent approach suggest that each individual Fe^0 atom corroding within a filter contributes to both reactivity and permeability loss. This approach applies well to alternative iron materials such as bimetallics, composites, hybrid aggregates, e.g. Fe^0 /sand, and nano- Fe^0 . Characterizing the intrinsic reactivity of individual Fe^0 materials is a prerequisite to designing sustainable filters. Indeed, Fe^0 ratio, Fe^0 type, Fe^0 shape, initial porosity, e.g. pore size and pore size distribution, and nature and size of admixing aggregates, e.g. pumice, pyrite and sand, are interrelated parameters which all influence the generation and accumulation of iron corrosion products. Fe^0 should be characterized in long-term experiments, e.g. 12 months or longer, for Fe dissolution, H_2 generation and removal of contaminants in three media, i.e., tap water, spring water and saline water, to allow reactivity comparison and designing field-scale filters.

Keywords Intrinsic reactivity · Permeability loss · Permeable reactive barriers · Porous media · Water filter · Zero-valent iron

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Introduction

Metallic iron (Fe^0) is a popular and low-cost material used in the water treatment industry for more than 160 years (Bischof 1973; Devonshire 1890; Baker 1934; van Craenenbroeck 1998; Cundy et al. 2008; Mwakabona et al. 2017). Fe^0 has been exploited to remove various contaminants and groups of contaminants from polluted natural waters and wastewaters through various mechanisms such as adsorption, co-precipitation, degradation, ion exchange, reductive transformations and size exclusion (Henderson and Demond 2007; Hu et al. 2021; Lawrinenko et al. 2023a, b; Noubactep 2023). Relevant contaminants include chlorinated solvents (Guan et al. 2015), emerging contaminants (Stieber et al. 2011), halide salts (Antia 2022), metalloids (Lan et al. 2023), radionuclides (Zhang et al. 2021) and trace metals (Li and Liu 2022).

The literature unanimously reports a time-dependent decline in the performance of Fe^0 -based filters (Bigg and

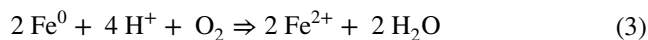
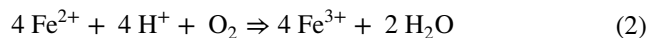
Judd 2000; Lee et al. 2004; Li et al. 2005, 2006; Henderson and Demond 2007; Bartzas and Komnitsas 2010; Li and Benson 2010; Guan et al. 2015). Henderson and Demond (2007) identified two related processes as the major causes of the system's failure: (1) a decline of Fe⁰ reactivity (reactivity loss) and (2) a decline of the hydraulic conductivity (porosity loss and permeability loss) (Firdous and Devlin 2014, 2015; Cormican et al. 2020). Biofouling and hydrogen (H₂) production were the other two possible causes of filter clogging (Noubactep 2013; Plessl et al. 2023; Singh et al. 2023).

Since 2007, the large majority of researchers has been attempting to solve both problems by applying various countermeasures (Bartzas and Komnitsas 2010; Li and Benson 2010; Guan et al. 2015). Addressing the two phenomena separately ignores the fact that the two are interrelated since both are linked to the inherent reactivity of Fe⁰ (Caré et al. 2013; Noubactep 2013; Domga et al. 2015). Reactivity loss and permeability loss are two inherent aspects of iron corrosion in porous media (Stefanoni et al. 2018; Angst 2019; Yang et al. 2021, 2022). In other words, reactivity loss and permeability loss occur in synergy in all Fe⁰-based filters. Consequently, increased preferential flow, reduced available porosity, residence time and reduced treatment performance over time are present in all Fe⁰ filters (Noubactep 2013; Cormican et al. 2020; Tao et al. 2022; Bilardi et al. 2023; Tao 2024).

Moreover, individual Fe⁰ materials respond differently to different environmental conditions including contamination, pH value, temperature, and water chemistry (Raychoudhury and Scheytt 2013; Sun et al. 2016). Therefore, to understand and predict the long-term performance of a Fe⁰ filter, it is essential to characterize the contaminant-free Fe⁰/H₂O system in a holistic approach. This holistic approach is based on the law of conservation of mass and entails considering the Fe mass balance to discuss three aspects: (1) the time-dependent generation of iron corrosion products (FeCPs) (corrosion rate), (2) the extent to which the pore space is filled (porosity loss) and (3) how the two related processes are impacted by dynamic changes in the pore size distribution. Obviously, material selection (corrosion rate or intrinsic reactivity) has not received the due attention in earlier studies (Velimirovic et al. 2018; Li et al. 2019; Cui et al. 2023; Ren et al. 2023).

The science of the contaminant-free Fe⁰/H₂O system has been presented a century ago in the iron corrosion literature (Whitney 1903) but has been largely overlooked by the pioneers of the Fe⁰ remediation technology (Gillham and O'Hannesin 1994; Matheson and Tratnyek 1994) and the majority of subsequent active researchers as summarized by Lawrinenko et al. (2023a) and Xiao et al. (2023). Whitney (1903) demonstrated that, at pH larger than 4.5, Fe⁰ immersed in water is corroded by water and water alone

(H⁺ from the dissociation of water molecules) (Eq. 1). If molecular oxygen (O₂) is available, Fe²⁺ is further oxidized to Fe³⁺ (Eq. 2). This makes the reaction after Eq. 3 impossible.

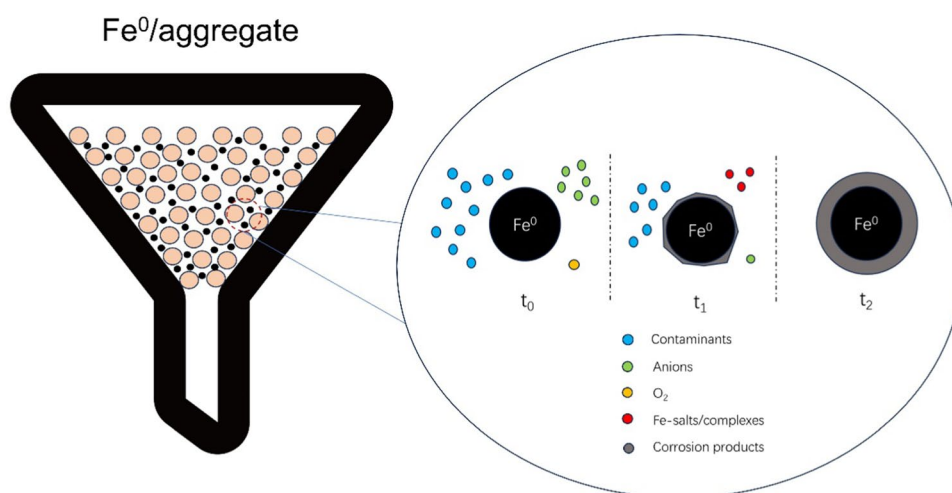


The demonstration by Whitney (1903) was later validated by corrosion scientists during the 1930s while establishing the electrochemical theory of aqueous metal corrosion (Evans et al. 1931; Evans and Hoar 1932; Evans and Mears 1934). Equation 1 clearly shows that the oxidative dissolution of one mole of Fe⁰ produces one mole of Fe²⁺ and one mole of H₂ (gas). In the absence of any organic complexing agent (e.g. natural organic matter), Fe²⁺ and Fe³⁺ form salts, e.g. FeCl₂, FeCl₃, FeSO₄, complexes (FeF₆³⁻), or precipitates, e.g. Fe₂O₃, Fe₃O₄ (Whitney 1903; Wang et al. 2018; Crespo et al. 2023; Tao 2024). Hydrogen (H₂) either escapes or is used in biogeochemical processes within the system. In other words, before the advent of the Fe⁰ remediation technology, it was established that Fe⁰ corrodes in water, even when present as moisture (Stratmann and Müller 1994; Wilson 1995; Tratnyek 1996).

Iron corrosion by water (Eq. 1) is electrochemical in nature. Whitney (1903) also demonstrated that the presence of dissolved O₂ and carbonic acid (H₂CO₃) accelerates iron corrosion by: (1) consuming Fe²⁺ for the chemical reduction of O₂ (Eq. 2) and (2) supplying H⁺ for Fe⁰ oxidation (via H₂CO₃ dissociation). Clearly, O₂ reduction in a Fe⁰/H₂O system is a chemical reaction (electrons from Fe²⁺) (Eq. 2). The rationale for this is that the Fe⁰ surface is instantaneously shielded by an oxide scale, acting as a diffusion barrier for O₂, and other potential dissolved oxidizing agents, including contaminants (Noubactep et al. 2012a; Noubactep 2016a; Konadu-Amoah et al. 2022). The oxide scale also acts as a conduction barrier for electrons from Fe⁰ (Fig. 1) (Hu et al. 2021; Xiao et al. 2023).

Here, we show that assumptions sustaining the design of current Fe⁰ filters are not valid. Equation 1 summarizes the alternative approach already adopted by some few scientists (Ghauch 2015; Vollprecht et al. 2020; Gheju and Balcu 2023; Bilardi et al. 2023; Lan et al. 2023), including our research group (Noubactep 2015, 2023; Noubactep 2023; Xiao et al. 2023; Tao 2024; Tao et al. 2024). Herein, Eq. 1 is used to demonstrate the inconsistency of the popular view summarized by Lawrinenko et al. (2023a, b). The following sections start with an overview description of Fe⁰ corrosion in porous media ("[Metallic iron corrosion in porous media](#)")

Fig. 1 Contaminant removal in $\text{Fe}^0/\text{H}_2\text{O}$ systems in the presence of co-solutes, e.g. anions. Individual processes, e.g. adsorption, co-precipitation, chemical reactions, are not shown



section), followed by a summary of the fundamental flaws in designing Fe^0 filters ("Fundamental flaws in designing metallic iron filters" section). The section "Next-generation metallic iron filters" discusses how to better design such filters by drawing lessons from earlier studies.

Metallic iron corrosion in porous media

A reliable approach for a knowledge-driven design of sustainable and stable Fe^0 filters goes through a holistic analysis of the $\text{Fe}^0/\text{H}_2\text{O}$ system (Noubactep 2009; Cao et al. 2021). A Fe^0 filter is a porous bed containing granular Fe^0 in a pure (Fe^0 alone) or hybrid system (e.g. Fe^0 /pyrite, Fe^0 /sand) (Moraci and Calabrò 2010; Firdous and Devlin 2015; Tao et al. 2022). On the one hand, a porous bed is characterized by an initial porosity, e.g. 36% for compact spheres (Caré et al. 2013), which necessarily decreases over time as contaminants are eliminated from inflowing polluted waters. On the other hand, iron corrosion is a volumetric expansive process, meaning that the volume of each oxide (V_{oxide}) is larger than that of the parent metallic iron (V_{iron}): $V_{\text{oxide}} > V_{\text{iron}}$ (Pilling and Bedworth 1923; Caré et al. 2008; Zhao et al. 2011). This inherent feature of Fe^0 materials implies that Fe^0 filters experience permeability loss even in the absence of any contaminants (Caré et al. 2013; Luo et al. 2013; Tao et al. 2022). Luo et al. (2013) have documented this evidence using X-ray computed tomography. The results of Luo et al. (2013) also recalled that Fe^0 corrodes in deionized water (contaminant-free) as reported by Whitney (1903).

Iron corrosion in a Fe^0 filter is an electrochemical process as discussed above (Eq. 1). The four components are: (1) iron dissolution at an anodic site of Fe^0 , i.e. releasing Fe^{2+} and leaving electrons behind, (2) transport of released Fe^{2+} through the electrolyte (polluted water) away from the Fe^0

surface, (3) transfer of electrons to a cathodic site on the surface of uncorroded Fe^0 and (4) H^+ reduction at the cathode (Fig. 2). The corrosion rate is influenced by several factors including: (1) the rate with which species H^+ and Fe^{2+} are consumed or released (intrinsic reactivity), (2) their rate of transport through the pore system within the filter (system porosity), (3) the occurrence of further chemical reactions such as complexation, oxidation, precipitation (i.e. pH value, solution chemistry), and (4) the occurrence of physical processes such as adsorption, ion exchange and size exclusion (pH value, solution chemistry) (Nesic 2007; Noubactep 2015; Sun et al. 2016; Tao 2024; Tao et al. 2024).

Components (1) to (4) locally affect the electrolyte chemistry at the Fe^0 surface, which in turn affects the electrochemical reaction kinetics of the uncorroded Fe^0 . It is essential to mention that H_2 and Fe^{2+} (Eq. 1) are stand-alone reducing agents for several organic (Jiao et al. 2009) and inorganic pollutants (Farrell et al. 2001). Moreover, several Fe^{II} and $\text{Fe}^{\text{II}}/\text{Fe}^{\text{III}}$ species generated in $\text{Fe}^0/\text{H}_2\text{O}$ systems are stand-alone natural reducing agents (Charlet et al. 1998;

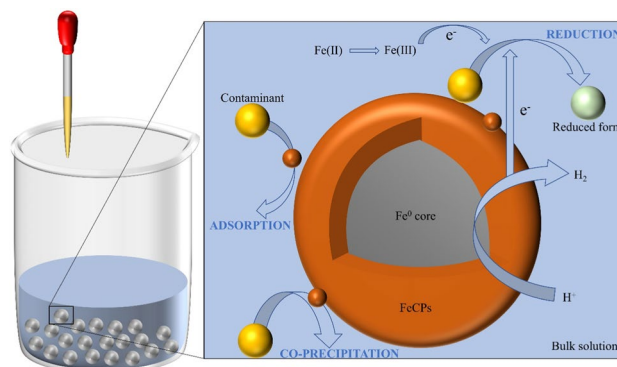


Fig. 2 The $\text{Fe}^0/\text{H}_2\text{O}$ system, indicating key features relevant for contaminant removal

Chaves 2005; Henderson and Demond 2007). For example, Fig. 2 shows that iron corrosion begins at a location where Fe^{2+} is generated (anode). Fe^{2+} goes into the aqueous solution and two electrons left behind migrate to another location (cathode), where they are taken up by H^+ from water dissociation. The resulting hydroxide ions (OH^-) react with the Fe^{2+} to initially form hydrous iron oxides that precipitate as $\text{Fe}(\text{OH})_2$. Depending on the environmental conditions, $\text{Fe}(\text{OH})_2$ is oxidized and transformed into various $\text{Fe}^{\text{II}}/\text{Fe}^{\text{III}}$ oxides that form the oxide scale (Charlet et al. 1998; Liger et al. 1999). The oxide scale acts as a barrier that restricts quantitative contaminant transfer to the Fe^0 surface.

Another characteristic feature of Fe^0 filters is the presence of a porous medium which significantly influences all processes involved in the progression of iron corrosion (Eq. 1). The most important being that: (1) there is limited space to store expansive corrosion products, and (2) transport processes are all limited, e.g. restricting convection. A potential beneficial role of a porous medium adjacent to the Fe^0 surface is that different local properties at the $\text{Fe}^0/\text{H}_2\text{O}$ interface create new electrochemical cells to sustain iron corrosion.

Summarized, to design a Fe^0 filter, the following is needed: (1) a filter container, (2) a reactive Fe^0 sample, and (3) one or several filling materials (e.g. gravel, pumice, sand) or reactive aggregates, e.g. manganese dioxide (MnO_2), pyrite (FeS_2) (Tao et al. 2022; Bilardi et al. 2023; Tao 2024). Because Fe^0 corrosion is volumetric expansive in nature, a pure Fe^0 filter (100% Fe^0) can be efficient but not sustainable (Hussam 2009; Domga et al. 2015). In other words, sustainable Fe^0 filters are hybrid in nature, e.g. Fe^0 /sand (Tepong-Tsindé et al. 2019; Tao et al. 2022).

In practice, Fe^0 proportion, Fe^0 type, Fe^0 shape, initial porosity, nature, shape and size of admixing aggregates, and water quality are interrelated parameters which all influence the progressive generation and accumulation of iron corrosion products (FeCPs) within the pores of any Fe^0 filter (Ruhl et al. 2014; Cormican et al. 2020; Tepong-Tsindé 2021). The filter function can also be influenced by sediment influx and deposition. Consequently, determining the rate of sedimentation and the period before sediment accumulation obstructs the filter's ability to function effectively is crucial in Fe^0 filter design. While designing a Fe^0 filter, enough storage capacity for FeCPs should be considered so that FeCPs accumulation does not affect the filter's function during its planned operational life. One proven efficient approach has been the use of porous Fe^0 materials (Bischof 1973; Hussam and Munir 2007; Hussam 2009; Rahman et al. 2013; Bilardi et al. 2023; Plessl et al. 2023).

Fundamental flaws in designing metallic iron filters

The literature reports on "detailed analysis of the rate of surface precipitate buildup" (Wilkin et al. 2002) in Fe^0 filters as a critical step in understanding how long-term systems can be sustainably designed (Li et al. 2005, 2006; Li and Benson 2010). However, the reasoning was rooted on the Fe^0 amount available for the reductive transformation of selected contaminants (Sarr 2001; Wilkin et al. 2002). In this approach, depending on the environmental conditions (e.g. aquifer chemistry, composition of Fe^0 filter), different types of minerals and surface coatings have been discussed (Vikesland et al. 2003, 2007; Li et al. 2006; Li and Benson 2010). The impacts of microbiological processes (e.g. presence of bacteria and their mediated processes) have also been largely discussed (Henderson and Demond 2007; Raychoudhury and Scheytt 2013; Fu et al. 2014; Obiri-Nyarko et al. 2014). In this context, Wilkin et al. (2002) present a subsurface Fe^0 filter as a "large reservoir of iron" for contaminant transformation whose action can be enhanced by microorganisms including iron-reducing, sulphate-reducing or methanogenic bacteria. Wilkin et al. (2002) also suggested that such enhancements may occur at the "expense of faster corrosion," thereby inducing a faster clogging of the Fe^0 filter.

The approach presented by Wilkin et al. (2002) is commonplace in the Fe^0 literature and was mainly a recommendation of the Interstate Technology & Regulatory Council (ITRC 2005, 2011). Accordingly, to explore the biogeochemical processes occurring within a Fe^0 filter, and thus understand the reasons for reactivity loss and permeability loss, sophisticated structural analytical techniques and detailed coring and water sampling programmes are needed (Gaspar et al. 2002; McGuire et al. 2003; Chekli et al. 2016; Li et al. 2019; Wielinski et al. 2022). However, in the absence of any theory of the system ("Introduction" section), there is no guide to orient the discussion of the achieved results from such monitoring studies. In addition, most of the observations made from coring and water sampling programmes are just static snapshots and their measurements are inaccurate (Brenner 2010). Thus, they cannot enable the generation of non-trivial models of the dynamic processes within Fe^0 filters, especially as these occur over an enormous range of timescales, for example, from a few days or weeks (laboratory experiments) to several months or years (pilot studies and field implementations). Moreover, each Fe^0 material has its own intrinsic corrosion rate (Reardon 1995), meaning that two different materials will fill the same pore space within different timescales (Fig. 3).

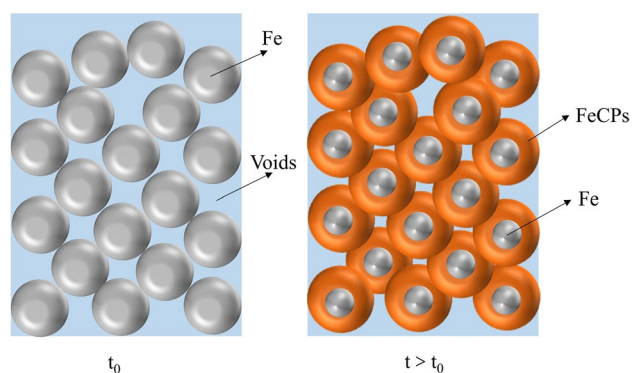


Fig. 3 Changes in the residual porosity in a bed filled with spherical Fe^0 particles: left: at the start of the operation ($t_0=0$) and right: at some time later ($t > t_0$, but $t < t_\infty$) before Fe^0 complete exhaustion at t_∞ . The time required for complete exhaustion (t_∞) depends on various factors, including Fe^0 intrinsic reactivity, initial porosity, water chemistry and Fe^0 ratio

The approach recommended by the Interstate Technology & Regulatory Council (ITRC) for subsurface Fe^0 -based permeable reactive barriers (Fe^0 PRBs) aimed at: (1) characterizing the surface precipitates and minerals formed within and downgradient of the Fe^0 PRBs, (2) developing conceptual models to predict the type and rate of precipitate formation, (3) identifying relevant microbiological activities impacting the service life of Fe^0 PRBs, and (4) developing applicable and cost-effective protocols for long-term performance monitoring of Fe^0 PRBs (ITRC 2005; 2011).

Ideally, a Fe^0 PRB should support complete groundwater remediation while maintaining an acceptable hydraulic conductivity over its planned service life (Bilardi et al. 2023). However, inherent temporal and spatial variability of chemical, microbial and physical processes within the Fe^0 PRB is difficult to correlate with permeability loss and reactivity loss using the conventional approach (Wielinski et al. 2022; Tao et al. 2024). This correlation has not been successfully established by the ITRC approach (Santisukksaem and Das 2019; Wielinski et al. 2022; Berardozzi et al. 2023), suggesting that an alternative approach is needed to design sustainable Fe^0 filters, including Fe^0 PRBs (Wielinski et al. 2022).

The service life of a Fe^0 filter is essentially determined by how fast minerals precipitate within its pore space, which in turn determines the rate of porosity loss and permeability loss. This reveals the paramount importance of the corrosion rate (Fe^0 intrinsic reactivity) of used materials in aqueous environments (Lufingo et al. 2019; Cui et al. 2023; Noubactep 2023; Noubactep et al. 2023a; b). Since 1995, the corrosion rate of Fe^0 materials has been widely characterized (Reardon 1995, 2005; Velimirovic et al. 2018). However, this has been done in a too pragmatic approach (Lufingo et al. 2019) and for too short

experimental durations (Li et al. 2019; Konadu-Amoah et al. 2023). This points to the need for: (1) an alternative approach based on the fundamentals of iron corrosion science and (2) considering reasonably long timescales representative of real field applications. The presentation herein insists on the fact that the oxidative dissolution of each individual Fe^0 contributes to reducing the pore space (porosity loss) and inducing reactivity loss. This occurs to different extents depending on the prevailing environmental conditions, e.g. availability of molecular O_2 , Fe^0 mass, Fe^0 ratio, pH value, water chemistry (Sun et al. 2016; Tao et al. 2023, 2024).

A proof that the conventional approach has not worked is the development of artificial neural network for evaluating permeability decline in Fe^0 PRBs (Santisukksaem et al. 2015; Santisukksaem and Das 2019; Berardozzi et al. 2023). These authors clearly stated that all other approaches were not successful as independently reported by Moraci et al. (2016) and Noubactep (2016b). However, the artificial neural network is not a physically based approach as it relies on the network's ability to understand given information and outputs from physically based relationships derived using other methods (Santisukksaem et al. 2015). Fortunately, a physical approach has been presented which is able to address both permeability loss and reactivity loss in a very simple and low-cost experimental setup: the methylene blue discoloration method (Btateku-K. et al. 2016; Konadu-Amoah et al. 2021; Tao 2024; Tao et al. 2024).

The methylene blue method uses methylene blue as an indicator of reactivity for the $\text{Fe}^0/\text{H}_2\text{O}$ system (Btateku-K. et al. 2016; Konadu-Amoah et al. 2021; Konadu-Amoah 2023; Tao 2024; Tao et al. 2024). The methylene blue method is hinged on the historic findings by Mitchell et al. (1955) that the efficiency of pure sand to adsorb methylene blue declines as iron oxides progressively cover the surface of sand (Fig. 4). The suitability of the methylene blue method to characterize $\text{Fe}^0/\text{H}_2\text{O}$ systems is based on the evidence that: (1) sand is a good adsorbent for methylene blue, and (2) iron oxide-coated sand has a poor affinity for methylene blue discoloration (Miyajima 2012). Zarime et al. (2024) have recently independently reproduced the results of Miyajima (2012) using granitic residual soil-supported nano- Fe^0 . The method characterizes methylene blue discoloration by sand as it is in-situ coated by iron corrosion products (FeCPs) in $\text{Fe}^0/\text{H}_2\text{O}/\text{sand}$ system (Miyajima 2012). In other words, the methylene blue method enables the detection of the decline of Fe^0 reactivity (reactivity loss), which is related to porosity loss. In fact, Miyajima (2012) has also demonstrated the ability of the methylene blue method to trace the clogging of Fe^0 filters (permeability loss).

Miyajima (2012) unequivocally demonstrated for the first time that admixing Fe^0 with non-expansive aggregates is a prerequisite for sustainable Fe^0 filters. Prior to 2012,

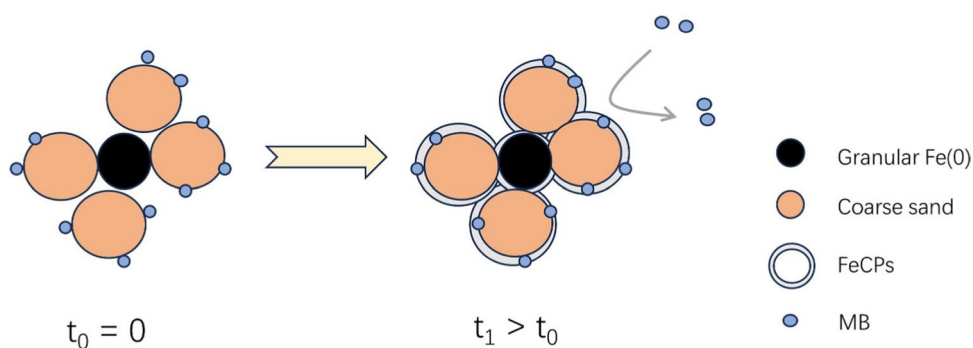


Fig. 4 The time-dependent process of methylene blue (MB) discoloration in Fe^0/sand systems. From the start of the experiment ($t_0=0$), Fe^0 corrosion produces solid iron corrosion products (FeCPs) which progressively coat the sand surface. At t_1 higher than t_0 , avail-

able sand is completely coated with FeCPs and methylene blue is no longer attracted to sand particles. From t_1 onwards, methylene blue is eliminated from the aqueous phase solely by occlusion within the matrix of precipitating FeCPs (co-precipitation)

admixing Fe^0 with other aggregates was regarded as "material dilution" with possible adverse impacts on reaction rates and contaminant removal efficiency despite a beneficial cost reduction (Kenneke and McCutcheon 2003; Kaplan and Gilmore 2004; Song et al. 2005; Bi et al. 2009). This misconception originated from the false view that Fe^0 is a reducing agent for many dissolved species (Matheson and Tratnyek 1994; O'Hannesin and Gillham 1998; Furukawa et al. 2002; Lawrinenko et al. 2023a, 2023b).

Next-generation metallic iron filters

A biased starting point

The presentation until now suggests that the Fe^0 research community has been violating the first law of thermodynamics for three decades. It has been known since 1903 that, in aqueous solutions at pH 4.5 or above (quantitative Fe precipitation), Fe^0 is corroded by H^+ (from water dissociation or a weak acid) and only by H^+ (Whitney 1903). According to Whitney (1903) and in tune with the laws of thermodynamics, O_2 reduction in the presence of

Fe^0 (Eq. 2) is mediated by electrons from Fe^{2+} (Eq. 1). Fe^0 corrosion is just accelerated as Fe^{2+} from Eq. 1 is consumed for O_2 reduction (Le Chatelier's principle). In other words, it has been a mistake to introduce Fe^0 as a stand-alone reducing agent for selected species (including O_2) in polluted waters under environmental conditions (Matheson and Tratnyek 1994; Gillham and O'Hannesin 1994; O'Hannesin and Gillham 1998). Moreover, the pioneers of the Fe^0 technology have not proved Whitney (1903) false. Instead, they were looking for a rationale "directly relevant" to "reductive dechlorination by Fe^0 " (Reynolds et al. 1990; Lee et al. 2004). However, as discussed above, reductive dechlorination occurs in the presence of Fe^0 (Eq. 4) and not by Fe^0 (Eq. 5) (Fig. 5). Clearly, the Fe^0 remediation technology was introduced with a thinking mistake. This mistake has dragged active researchers very deep into confusion (Noubactep 2015; Makota et al. 2017; Cao et al. 2022; Xiao et al. 2023), to the extent that attempts to fix some confusions are now blamed and misconstrued as "isolated misconceptions" (Lawrinenko et al. 2023a).

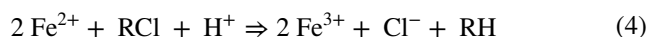
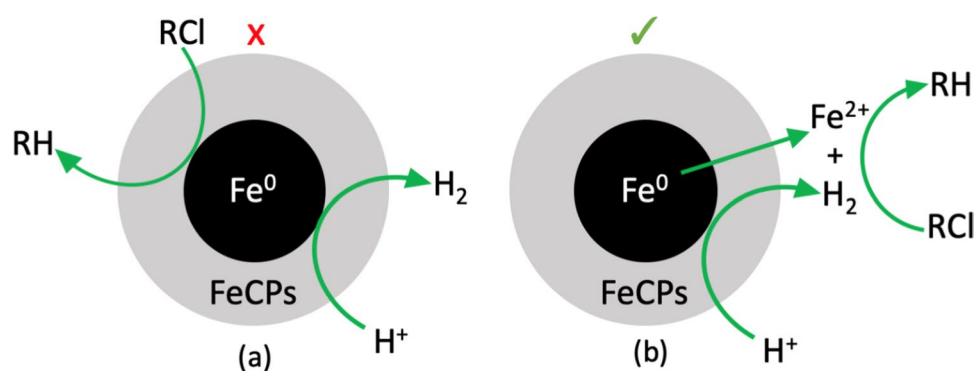
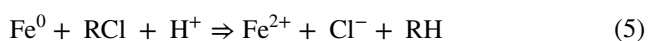


Fig. 5 The mechanism for contaminant (RCl) reduction in $\text{Fe}^0/\text{H}_2\text{O}$ systems: left: Fe^0 is oxidized by RCl and water (H^+) (Path 1), and right: Fe^0 is oxidized by water (H^+) and RCl is reduced by Fe^{II} species and H_2 (Path 2). Path 1 is not feasible because the oxide scale is a conduction barrier for electrons from Fe^0 . Path 2 is feasible and rooted on aqueous iron corrosion





In 1947, in recognition of the significance of the 1903 work of Dr. Whitney to the development of corrosion science, the National Association of Corrosion Engineers (NACE—Houston, USA) introduced the Willis Rodney Whitney Award with Dr. Whitney as the first recipient. The award is given "in recognition of significant contributions to corrosion science, such as the development or improvement of a theory that provides a fundamental understanding of corrosion phenomena and/or the prevention of corrosion." In other words, Dr. Whitney is regarded by corrosion experts as an individual who has made a major contribution to corrosion science through work that leads to a better understanding of the science of corrosion. Clearly, the consensus adopted to justify observed contaminant reduction in $\text{Fe}^0/\text{H}_2\text{O}$ systems (O'Hannesin and Gillham 1998) has disproved and even distorted well-established knowledge of corrosion scientists (Hu et al. 2021; Konadu-Amoah et al. 2022; Xiao et al. 2023). The correction shall start at the origin, where possible, with little attention to the volume of scientific articles that have been propagating the named mistakes (Chen et al. 2019; Antia 2020; Li et al. 2021; Wu et al. 2024).

It is essential to recall that some individual researchers and research groups have constantly challenged the validity of the prevailing theory (Fe^0 as a reducing agent) (Warren et al. 1995; Fiedor et al. 1998; Farrell et al. 1999; Qiu et al. 2000; Lavine et al. 2001; Lee et al. 2004; Noubactep 2007, 2008, 2015; Jiao et al. 2009; Gheju 2011, 2018; Ghauch et al. 2011). In particular, Lavine et al. (2001) investigated nitrobenzene reduction in $\text{Fe}^0/\text{H}_2\text{O}$ systems using differential pulse polarography, a technique which can simultaneously monitor the disappearance of nitrobenzene and the appearance of Fe^{2+} . Their results could not confirm any involvement of the Fe^0 surface in the processes controlling nitrobenzene reduction within the system. This result is in tune with the seminal work of Whitney (1903) and recalls that despite a prevailing wrong theory, a science-based interpretation of one's experimental observations is always possible (Vollprecht et al. 2020; Cao et al. 2021; Tao 2024; Tao et al. 2024).

Subsequent publications by several other research groups denying the named theory have been acknowledged in some overview papers like Giles et al (2011) and Sista et al. (2021). In other words, the wrong theory and the alternative have coexisted in the literature for the past 30 years. However, each individual researcher has opted for their preference (Lawrinenko et al. 2023a, 2023b; Noubactep 2023; Noubactep 2023); Xiao et al. 2023). As a result, a circular reasoning has been established, maintaining the

Fe^0 technology as a whole in the status of an innovative technology.

Roadmap for sustainable filters

The misconception that Fe^0 is an environmental reducing agent created unprecedented confusion within the environmental research community (Furukawa et al. 2002; Makota et al. 2017; Vollprecht et al. 2018; Hu et al. 2021; Lawrinenko et al. 2023a, b). The realization of this confusion around 2005 has prompted the exploration of new avenues of investigation (Noubactep 2007, 2012; Makota et al. 2017; Yang et al. 2022). A critical evaluation of the literature revealed three main pervasive issues in research methodology: (1) the lack of standardized procedures to characterize the $\text{Fe}^0/\text{H}_2\text{O}$ system (Noubactep et al. 2006; Ghauch et al. 2011), (2) the lack of organized and reliable data (Noubactep 2023, 2005a), and (3) the lack of unified tools for Fe^0 characterization, including the lack of a reference Fe^0 material (Noubactep et al. 2005b; Reardon 2005). Moreover, past and current shortcomings in research methodology highlighted the need for a multifaceted approach to tackle the named confusion. Thus, from 2008 onwards, our research group set out to explore and develop affordable and applicable strategies to advance shared efforts in optimizing the application of Fe^0 within the water treatment industry. Table 1 summarizes some milestones of this research effort.

One key phenomenon characterizing the $\text{Fe}^0/\text{H}_2\text{O}$ system is the occurrence of some chemical reactions between dissolved Fe^{2+} from Eq. 1 and water constituents, including inorganic salts and molecular O_2 (Wang et al. 2018; Tao et al. 2023, 2024). These interactions culminate in the coverage of the Fe^0 surface by a film of iron corrosion products (e.g. oxide scale) (Nesic 2007). Thus, Fe^0 oxidative dissolution (anodic dissolution) and contaminant removal (e.g. physical adsorption) and/or transformation (e.g. chemical reduction) occur at a different timescale (Schreier and Reinhard 1994; Hao et al. 2005; Naseri et al. 2017). This was one of the first experimental observations in investigating the $\text{Fe}^0/\text{H}_2\text{O}$ system (Schreier and Reinhard 1994) and a clear proof that Fe^0 dissolution and contaminant reduction are not simultaneous reactions (Lipczynska-Kochany et al. 1994; Lavine et al. 2001; Lan et al. 2023). This means that reducing electrons for any dissolved species are not from Fe^0 or that contaminant reduction is not the cathodic reaction coupled to Fe^0 oxidation (Noubactep 2016a; Hu et al. 2021; Xiao et al. 2023). This corresponds to the report of Whitney (1903) and disproves the theory introduced by the pioneers of the Fe^0 remediation technology in the early 1990s. The question arises, how can this evidence be denied for so many years? The answer lies in the suitability of the used methodologies to investigate the $\text{Fe}^0/\text{H}_2\text{O}$ system in both batch and

Table 1 Milestones in the development of Fe⁰ filters

Anno	Key finding	References
2007	Fe ⁰ is not an environmental reducing agent	Noubactep (2007)
2009	The first holistic analysis of the Fe ⁰ /H ₂ O is presented	Noubactep (2009)
2009	Only quiescent batch experiments are suitable to investigate the Fe ⁰ /H ₂ O system	Noubactep et al. (2009a)
2009	A concept is presented to use Fe ⁰ -based filters for universal access to safe drinking water	Noubactep et al. (2009b)
2010	Admixing non-expansive aggregates to Fe ⁰ sustains the efficiency of Fe ⁰ -based systems: Only hybrid Fe ⁰ /aggregate filters are durable	Noubactep and Caré (2010a)
2011	Reduction is not a relevant mechanism for decontamination in Fe ⁰ /H ₂ O systems	Noubactep (2011)
2012	Presentation of the methylene blue method (MB method)	Miyajima (2012)
2012	Methylene blue method demonstrated the sustainability of hybrid Fe ⁰ filters	Miyajima (2012)
2013	Methylene blue method is used to characterize the reactivity of Fe ⁰ materials. The paucity of efforts to characterize Fe ⁰ materials is outlined	Batkeu-K et al. (2013)
2015	Methylene blue method elucidated the operating mode of Fe ⁰ /MnO ₂ systems	Batkeu-K (2015)
2015	Methylene blue method demonstrated that Fe ⁰ /H ₂ O systems are ion-selective	Phukan (2015)
2015	The Fe ⁰ remediation community is overlooking alternative views	Noubactep (2015)
2017	A framework to investigate the Fe ⁰ /H ₂ O system is presented	Naseri et al. (2017)
2019	1,10-Phenanthroline (Phen) is used to characterize the intrinsic reactivity of Fe ⁰ materials (Phen method)	Lufingo et al. (2019)
2019	Methylene blue method elucidated the importance of experimental duration in investigating the Fe ⁰ /H ₂ O system	Alyoussef (2019)
2019	A sustainable household water filter using steel wool is piloted in a peri-urban area in Douala (Cameroon)	Tepong-Tsindé et al. (2019)
2021	The kinetic of Fe ⁰ corrosion has not been investigated	Yang et al. (2021)
2023	Methylene blue method elucidated the mechanism of phosphate removal in Fe ⁰ /H ₂ O systems	Konadu-Amoah (2023)
2023	Ascorbic acid (AA) is used to characterize the intrinsic reactivity of Fe ⁰ materials (AA method)	Cui et al. (2023)
2024	Methylene blue method elucidated the influences of inorganic ligands on the efficiency of Fe ⁰ /H ₂ O systems	Tao (2024)
2024	The methylene blue method is independently presented using a granitic residual soil (Gr) as dispersant for nano-nFe ⁰ : Fe ⁰ impedes MB removal by Gr	Zarime et al. (2024)

column experiments (Naseri et al. 2017; Bilardi et al. 2023; Tao et al. 2023, 2024; Xiao et al. 2023).

There have been a large number of systematic studies designed to elucidate the mechanisms of contaminant removal in Fe⁰/H₂O systems and the effects of many operational parameters, e.g. Cl⁻, Fe⁰ size, Fe⁰ treatment, Fe⁰ type, HCO₃⁻, mixing intensity, O₂, SO₄²⁻, on related processes (Sun et al. 2016; Abd El-Monaem et al. 2024; Tao et al. 2024). However, controversial results have been typically reported, even for the same probe contaminant and the same Fe⁰ material (Matheson and Tratnyek 1994; Jiao et al. 2009). This is usually justified by the evidence that varying experimental procedures are employed for the collection of data (Sun et al. 2016; Tao et al. 2023, 2024). For batch experiments, these procedures differ in Fe⁰ pre-treatment, Fe⁰ particle size (e.g. granular, powder, nanoscale), admixing agents used (e.g. FeS₂, MnO₂, pumice, sand), volume of the experimental vessels (mL), volume of solution added (mL), equilibration time allowed, and ambient temperature (Vidic et al. 1990; Polasek 2007; Naseri et al. 2017).

The reasoning in sections "[Metallic iron corrosion in porous media](#)" and "[Fundamental flaws in designing metallic](#)

[iron filters](#)" suggests that the major pitfall in conventional batch experiments has been the disturbance of the "natural" layering sequence in Fe⁰/oxide/H₂O in which the Fe⁰ surface is permanently shielded by an oxide scale. In fact, while performing batch experiments, mixing intensities of up to more than 400 rpm are used to explicitly keep all particles in suspension and avoid mass transfer limitations (Devlin et al. 1998; Miyajima and Noubactep 2012; Noubactep et al. 2012a). Such procedures certainly provide reproducible results, but without practical significance for filtration systems. In Fe⁰ filters, transport in the vicinity of Fe⁰ is diffusion-driven (Matheson and Tratnyek 1994; Nesic 2007). For this reason, quiescent batch experiments are recommended for treatability studies (Miyajima and Noubactep 2012; Noubactep 2012; Naseri et al. 2017). Our research group has been using exclusively quiescent batch experiments (mostly Fe⁰/sand mixtures) for the past 15 years (since 2009) for experimental durations of up to 90 days (3 months). A major result has been that expressing the performance of Fe⁰/H₂O systems in terms of adsorption capacity (e.g. mg of contaminant per g of Fe⁰) is not acceptable (Naseri et al. 2017). This is because: (1) Fe⁰ is not depleted, (2) its corrosion rate is

not known, and (3) no standard experimental protocol has been established.

The literature reports difficulties in understanding the long-term permeability of Fe⁰ filters (Domga et al. 2015; Moraci et al. 2016; Bilardi et al. 2023). Our research group has established around 2010 that sustainable Fe⁰ filters are hybrid in nature, wherein Fe⁰ is admixed with non-expansive aggregates (e.g. Fe⁰/MnO₂, Fe⁰/pyrite, Fe⁰/sand) (Noubactep and Caré 2010a, b; Noubactep et al. 2011, 2012b). It is established that factors influencing the sustainability of Fe⁰ filters include: (1) reactive Fe⁰ (intrinsic reactivity), (2) Fe⁰ shape and size, (3) nature and extent of contamination, (4) biogeochemical and hydrogeological characteristics of the aquifer, and (5) groundwater flow velocity (Tao et al. 2022, 2023, 2024). Bilardi et al. (2023) analysed the relative importance of these parameters and showed that "admixing Fe⁰ with non-expansive granular materials" is the most suitable tool to design "long-term hydraulically efficient" Fe⁰ filters. This corresponds to our findings from 2010 (Noubactep and Caré 2010a, b), showing that despite hundreds of new peer-reviewed scientific publications, little progress has been made. The reason for this sad situation is the proliferation of pragmatic, but independent investigations without any opportunity to compare achieved results. In particular, concerning the impact of Fe⁰/sand ratio, the rationale for working with 50% mixing ratio (w/w or v/v) is rarely specified, while (1) the Fe⁰ intrinsic reactivity is not characterized (Cui et al. 2023), (2) evidence exists that far lesser Fe⁰/sand ratios are efficient (Tepong-Tsindé 2021), and (3) experiments are performed for too short experimental durations (Alyoussef 2019; Yang 2022; Yang et al. 2022; Xiao et al. 2023; Tao et al. 2024).

This study recalled the following: (1) intensively homogenizing the experimental vessels has a significant influence on the decontamination efficiency of Fe⁰ while disturbing the real reaction mechanisms, (2) there have been no unified tools to characterize the intrinsic reactivity of Fe⁰ materials used, and (3) treatability studies have been performed for too short experimental durations. These three factors together explain differences in decontamination efficiencies reported in the literature for the same Fe⁰/contaminant pair, for example Fe⁰/CCl₄ for which Matheson and Tratnyek (1994) favoured an electrochemical mechanism while Jiao et al. (2009) explicitly rule out any electron transfer from Fe⁰. Additionally, the Fe⁰ remediation technology involves immersing small Fe⁰ particles (less than 5 mm thick) in water-saturated zones for long times. This is a novel situation for environmental scientists since there are no comparable data in the scientific literature, for example, on the long-term corrosion kinetics (Yang et al. 2022).

Therefore, a new experimental procedure (long-term quiescent batch experiments) is recommended for obtaining better data reflecting the true Fe⁰ corrosion

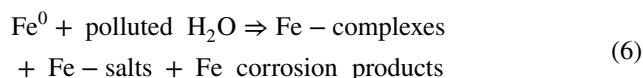
kinetics. Alternatively, the experimental vessels can be very gently homogenized, e.g. stirred at 75 rpm, such that the generation of an oxide scale on the Fe⁰ surface is not disturbed (Miyajima and Noubactep 2012). Universally employing this experimental procedure will enable the acquisition of reliable and comparable data to design and model better Fe⁰ filters for the water treatment industry. Since a parameter like an "adsorption capacity" for Fe⁰ materials cannot be defined, it is urgent to develop some standard experimental protocols to enable the comparison of results from independent investigations. The starting point is certainly the characterization of the intrinsic reactivity of available Fe⁰ materials, e.g. iPutec GmbH & Co. KG in Germany, Connelly-GPM in the USA or Högnäs in Sweden. Such Fe⁰ materials can be intensively characterized for their long-term ability to: (1) generate H₂ under well-defined conditions, e.g. tap water, spring water and saline water, (2) release Fe²⁺ in selected solutions, e.g. ascorbic acid, ethylenediaminetetraacetic acid—EDTA, and (3) remove some selected probe contaminants under well-defined operational conditions.

The discussion of a large number of data, obtained under such controlled experimental conditions (e.g. items (1) to (3)), would reveal some common underlying trends for interactions within Fe⁰/H₂O systems that provide confidence for a design that is non-material-specific. Accordingly, when a significant body of data exists on removal rates for selected contaminants by Fe⁰ materials depicting certain characteristics, site-specific treatability studies may only be required to fine-tune the design criteria for the optimal performance of Fe⁰ filters. Such data would provide a good starting point for the design of future laboratory-, pilot- and field-scale studies for investigating the remediation Fe⁰/H₂O system (McGeough et al. 2007; Naseri et al. 2017; Xiao et al. 2023; Tao 2024).

The presentation herein has recalled that Fe⁰ is corroded by water (H⁺) and not by any contaminant. This means that polluted water contains one or several species that can enhance or impede Fe⁰ corrosion (Sun et al. 2016; Naseri et al. 2017; Xiao et al. 2023; Tao 2024; Tao et al. 2024). The question is: how are individual contaminants removed during Fe⁰ corrosion in the presence of polluted water? This approach is supported by reports on Fe⁰ corrosion in deionized water (Luo et al. 2013; Xin et al. 2016) and the removal of several species without redox reactivity in Fe⁰/H₂O, such as methylene blue (Miyajima 2012; Btatkeu et al. 2016; Abd El-Monaem et al. 2024) pathogens (Bojić et al. 2001; You et al. 2005), phosphates (James et al. 1992; Erickson et al. 2012), triazoles (Jia et al. 2007a, b) and zinc (Bojic et al. 2009; Kishimoto et al. 2011).

Perspective

The Fe^0 remediation research community has been working with the wrong concept for the past three decades. Thus, this presentation seeks to motivate colleagues to use the new information to design better remediation $\text{Fe}^0/\text{H}_2\text{O}$ systems. In particular, the presentation has insisted on the relatively low significance of contaminants for the general design of such systems. In fact, the simplified dynamic $\text{Fe}^0/\text{H}_2\text{O}$ system can be summarized in a surrogate reaction after Eq. 6:



The rationale for Eq. 6 is recalled in Sect. "Metallic iron corrosion in porous media" as resulting from the seminal work of Whitney (1903). Polluted water may be natural water or wastewaters from agricultural, domestic and industrial origins. Each water or wastewater is characterized by its chemical composition, including the concentration of protons (pH value) (Sun et al. 2016; Zhu et al. 2022; Li et al. 2023). The variability of water or wastewater sources makes the design of remediation $\text{Fe}^0/\text{H}_2\text{O}$ systems highly site-specific. The presence of Fe complexes and Fe salts in Eq. 6 delays the availability of iron corrosion products (FeCPs), which are the contaminant scavengers (Fig. 6) (Tao et al. 2024). Once FeCPs are available, inorganic ligands, natural organic matter and polluting contaminants, e.g. arsenic, compete with each other to influence their further transformation, including the adsorptive removal of polluting contaminants (Tao 2024; Tao et al. 2024).

In other words, Tao (2024) has re-demonstrated that investigating the remediation $\text{Fe}^0/\text{H}_2\text{O}$ system can be summarized by characterizing the process of Fe^0 corrosion in the polluted water of concern (Eq. 6), while keeping in mind that each Fe^0 reacts with its own intrinsic kinetics (corrosion rate) and that this kinetics declines with the

time (reactivity loss) (Tao et al. 2024). The uniqueness of each reactive Fe^0 material suggests that it is possible to manufacture and select appropriate Fe^0 materials for specific applications. Two examples of such applications are safe drinking water supply and food fortification.

Safe drinking water supply

People throughout the world are afflicted by inadequate access to clean potable water, and this problem is expected to get worse in certain low-income regions. Since the early 2000s, a tremendous amount of research has been conducted to make Fe^0 -based filters affordable, efficient and robust methods of water treatment at domestic and community scales (Baumgartner et al. 2007; Shannon et al. 2008; Singh et al. 2014; Banerji and Chaudhari 2017; Kundu et al. 2018; Noubactep 2018, 2024; Ghosh et al. 2024). In other words, the affordability of such systems and their acceptability have already been largely discussed (Murcott 2007; Neumann et al. 2013; Kundu et al. 2018; Nguyen and Mulligan 2023; Ghosh et al. 2024). Here, we highlight how existing devices can be made more efficient based on the science of the system (Eq. 6, Fig. 2).

Two main types of Fe^0 filters were developed for household water filters: (1) the Kanchan filter (Ngai et al. 2006, 2007) and (2) the Sono filter (Hussam and Munir 2007). Both systems were primarily developed for arsenic removal. However, Eq. 6 and field results demonstrated that far more species than arsenic were quantitatively removed from polluted waters (Naseri et al. 2017; Kundu et al. 2018; Huang et al. 2021a, b; Ndé-Tchoupé et al. 2022). A Sono filter uses a proprietary material and is regarded as one of the best available household water filters (Naseri et al. 2017), but has not been disseminated to the expected extent (Neumann et al. 2013; Kundu et al. 2016; Edward et al. 2023). As concerning the Kanchan filter, it has received continuous attention since 2007 (Singh et al. 2014; Wenk et al. 2014;

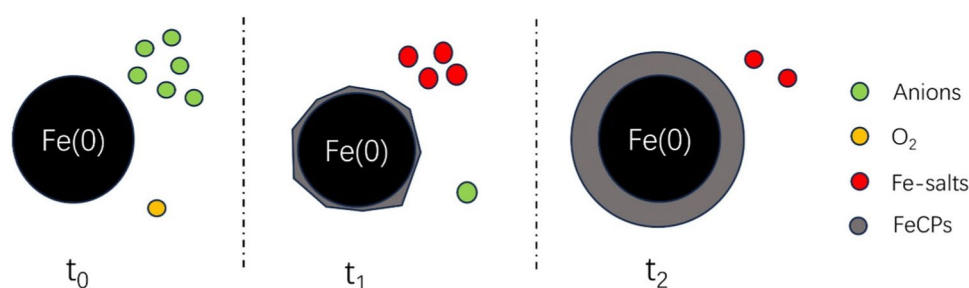


Fig. 6 Iron corrosion in $\text{Fe}^0/\text{H}_2\text{O}$ systems as influenced by the presence of diverse solutes, e.g. anions and O_2 . Fe^0 is oxidized by water (H^+), and O_2 is eventually reduced by Fe^{II} species and H_2 . However, all solutes influence the process of iron oxidation and the subsequent

precipitation of solid iron corrosion products (FeCPs) (co-precipitation). Once FeCPs are available, all solutes, including pollutants compete for adsorption at their surface

Ogata et al. 2020; Mueller et al. 2021; Mueller et al. 2023; Nguyen and Mulligan 2023; Noubactep 2024), but is still not satisfactorily implemented. According to Huang et al. (2021a) and Ndé-Tchoupé et al. (2022), the original Kanchan filter was designed with a thinking mistake. Other researchers have rightly concluded that Sono filters are better than Kanchan ones (Singh et al. 2014; Wenk et al. 2014; Noubactep 2024). Equation 6 shows that where Kanchan filters were successful in supplying safe drinking water, the combination of "water quality, Fe^0 reactivity, Fe^0 mass, water flow velocity" was optimal to treat polluted water to drinking standards. At some other sites, the water chemistry was also responsible for the observed failures. However, the actual mistake is to design "one filter for all situations." Clearly, despite a design mistake (not addressed herein), a filter can be efficient but may be less or not sustainable. Community-scale filters, e.g. IIT Bombay filter, built on the principle of the Kanchan filter have been very efficient for the past 15 years (Banerji and Chaudhari 2017) and could serve communities of up to 8,000 people (Etmanski and Darton 2014; Ghosh et al. 2024).

Altogether, using the Kanchan filter and the IIT Bombay filter as reference systems, the whole world can design modified decentralized water treatment systems to achieve Goal 6.1 of the United Nations Sustainable Development Goals (SDGs) within the remaining six years to 2030. Since 2009, our research group has been actualizing related concepts almost on a yearly basis (Noubactep et al. 2009b; Noubactep 2010, 2013, 2018, 2020, 2024; Yang et al. 2020). However, more holistic research is needed, mainly at the pilot scale.

Metallic iron for the control of anaemia

Low-income countries are still suffering from iron deficiency anaemia (IDA). This is because iron fortification of food (e.g. table salt, wheat flour) has been the most important tool to control and defeat the IDA in the developed world where food is systematically processed (Hurrell 2021; Kumari and Chauhan 2022). Processed food is not often available in developing countries as a rule, and when it is available, it is not affordable for the poor (Charles 2012; Noubactep et al. 2023a; b). For these reasons, alternative iron sources have been sought for low-income countries. Relevant alternative solutions include: (1) cooking with iron pots (Arcanjo et al. 2020), (2) cooking in pots of other materials with iron ingots in the meat (Rodríguez-Vivaldi and Beerman 2018; Massey et al. 2023) and (3) fortifying potable water with Fe^{II} salts and ascorbic acid (Dutra-de-Oliveira et al. 2011).

Equation 6 suggests that all tools using iron metal (Fe^0) to combat IDA are not necessarily reliable because of the unknown corrosion rates (Yang et al. 2020). On the other hand, the use of Fe^{II} salts is expensive. These two facts

have motivated the development of a new approach to water fortification. The approach entails leaching iron from Fe^0 with a solution of ascorbic acid (AA) and obtaining a concentrated Fe^{II} -AA solution (Solution 1). Solution 1 is then diluted to obtain drinking water with the desired Fe concentration, allowing the prevention or the healing of IDA. For details on this approach, interested readers are referred to Cui et al. (2023), Noubactep (2023), and Noubactep et al. (2023a; b). Clearly, the understanding of the operating mode of Fe^0 -based filters has inspired an innovation that can help the developing countries out of "the valley of tears" regarding the tragedy of IDA. In fact, the scientific community seems to be helpless in finding an affordable and universally applicable solution to control and defeat IDA (Rai et al. 2018; Pasupathy et al. 2023). More multidisciplinary research is needed to realize this concept for the benefit of the whole world particularly poor communities in low-income countries.

Coping with a reactive material

Understanding the corrosion process is crucial for designing novel Fe^0 -based technologies such as those for water treatment ("Safe drinking water supply" section) and combatting anaemia ("Metallic iron for the control of anaemia" section). These new technologies striving at "putting corrosion to use" (Tratnyek 1996) cannot just translate available results from research in corrosion science (Landolt 2007; Nescic 2007; El Ibrahimy et al. 2021; Li et al. 2024). This is because past research had mostly strived to avoid or mitigate corrosion (Holmes and Meadowcroft 1977; Nescic 2007; Balko et al. 2012; Chekli et al. 2016; El Ibrahimy et al. 2021). These efforts have already established the complexity of involved processes with at least two phases, for instance, solid (e.g. Fe^0 , FeCPs) and liquid (H_2O); and solid and gas (e.g. H_2). Therefore, corrosion research is a highly multidisciplinary task demanding the combination of several experimental methods to adequately monitor the corrosion process (Lavine et al. 2001; Balko et al. 2012; Wielinski et al. 2022). Traditionally, corrosion research is performed mainly by chemists, electrochemists, material scientists and physicists (Holmes and Meadowcroft 1977; Landolt 2007). These scientists express the corrosion rate in many different ways, including corroded depth per unit of time, current density, mass loss per unit time, number of moles transformed per unit of time, and volume of H_2 generated per unit of time. However, for water treatment, the corrosion rate has to be correlated with the extent of contaminant removal (Lavine et al. 2001; Balko et al. 2012; Xiao et al. 2023; Tao et al. 2024).

The challenge of using Fe^0 for water treatment entails considering two groups of phenomena and their

uncertainties: (1) the Fe⁰ corrosion rate and the amount of FeCPs generated in-situ, and (2) the mineralogical constitution of FeCPs with their adsorptive affinities to the contaminants of concern. The question of how the whole process is influenced by the solution chemistry is still not properly addressed (Sun et al. 2016; Tao et al. 2024). The presentation above has demonstrated that the Fe⁰ reactivity is yet to be properly considered while the recent literature reports on fundamental flaws in designing and interpreting adsorption experiments (Tien 2007; Tran et al. 2017, 2023). In other words, while non-considering the amounts of contaminant scavengers generated in their systems, scientists have developed new models that are necessarily false. The viability of the Fe⁰ remediation as technology depends largely on the quality of the work produced by its investigators. Based on the fact that the problems exposed herein have been revealed since 2007 (Noubactep 2007, 2008), it is fair to state that, within the Fe⁰ research community, the majority of the engaged individuals are without adequate preparations or readiness to consider alternative views or theories. This must be a concern to the whole environmental research community. This issue cannot be resolved by a few individuals or research groups (Tien 2007; Ghanch et al 2011; Hu et al. 2021; Gheju and Balcu 2023; Lan et al. 2023). This paper was written to make the problem better known to colleagues and the general public.

Conclusion

An analysis of the Fe⁰/H₂O system reveals that, for some 30 years, the Fe⁰ research community has misinterpreted the good experimental observation that some pollutants are chemically reduced in the presence of Fe⁰ as an electrochemical process (electrons from Fe⁰). In other words, inaccurate calculations, conclusions and discussions were drawn by scientists over a period equivalent to some 7 to 10 generations of PhD students. According to the viewpoints discussed above, the following key conclusions and perspectives are put forward:

- Fe⁰ is not a stand-alone reducing agent under environmental conditions, at pH higher than 4.5;
- The removal performance of Fe⁰ materials should not be expressed in mass per mass basis (e.g. mg pollutant per g Fe⁰), unless Fe⁰ is exhausted. This is because appropriate alternative parameters suitable for Fe⁰ are yet to be developed;
- Permeability loss and reactivity loss are two inherent and related properties of Fe⁰ in a porous system. The occurrence of both processes is intimately related to the

intrinsic reactivity of the materials used and the pore space available;

- An in-depth characterization of the long-term reactivity of selected Fe⁰ materials under relevant operational conditions is regarded as a key step in designing the next-generation Fe⁰ filters.
- In discussing experimental observations, more attention should be paid to understanding the unique properties of Fe⁰ corrosion products and target pollutants to avoid mistakes made when explaining removal mechanisms.

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Data availability Not applicable.

Code availability Not applicable.

Declarations

Conflict of interest The authors declare no conflicts of interest.

Ethics approval Not applicable.

Consent to participate Not applicable.

Consent for publication All authors have given consent for publication.

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