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Removal of hydrogen sulfide in air using cellular concrete waste: biotic and abiotic filtrations

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Abstract

The objective of this study was to investigate the removal of hydrogen sulfide (H₂S) present in air using cellular concrete waste as the packing material. Air filtration was performed under biotic and abiotic conditions. Experiments were carried out in a laboratory-scale PVC column (internal diameter of 300 mm) filled with a volume of 70 L of cellular concrete (1 m height). The polluted air flow was generated at 4 m³ h⁻¹ corresponding to an Empty Bed Residence Time (EBRT) of 63 s. In dry conditions without biomass (abiotic conditions), cellular concrete can be an effective medium for the treatment of H₂S in air. For an H₂S concentration of 100 ppmv, the removal efficiency was around 70 % (Elimination Capacity (EC) of 5.6 g m⁻³ h⁻¹). This finding can be explained by the physicochemical reactions that can take place between H₂S and the cellular concrete components (mainly CaO, CaCO₃ and Fe₂O₃). However, interactions between cellular concrete and H₂S are not yet fully understood. Used as a packing material for H₂S biofiltration (biotic conditions), cellular concrete waste efficiently...
treated (Removal Efficiency = 100 \%) high concentrations of H$_2$S (up to 133 ppmv corresponding to an EC of up to 10.5 g m$^{-3}$ h$^{-1}$). Physicochemical and biological mechanisms explaining H$_2$S removal seem to occur simultaneously in the biofilter. At an EBRT of 63 s, the maximal elimination capacity (EC$_{\text{max}}$) was 17.8 g m$^{-3}$ h$^{-1}$. A packed bed of cellular concrete also presents a satisfactory mechanical behavior with low pressure drops.

**Keywords:** Cellular concrete; Packing material; Biofiltration; H$_2$S; Calcium oxide; Iron oxide

1 Introduction

Hydrogen sulfide is an odorous, toxic, flammable and corrosive air pollutant. It is a colorless gas with the characteristic foul odor of rotten eggs. H$_2$S can cause death immediately when concentrations are over 500 - 1000 ppmv, while exposure to lower concentrations, such as 10 - 500 ppmv, can cause various respiratory symptoms. H$_2$S may also affect the nervous, cardiovascular, and hematological systems. H$_2$S is emitted from various industries, such as petroleum refining, rendering, wastewater treatment, paper manufacturing and food processing. H$_2$S also occurs in volcanic and natural gases. Several processes are available for the treatment of hydrogen sulfide, including absorption [1–5], adsorption [6–11], and membrane separation [12–16]. These methods generally entail high energy, chemical and disposal costs. Biofiltration appears to be a convenient alternative for treating gaseous emissions containing H$_2$S. This process uses microorganisms immobilized in the biofilm attached to a packing material. The contaminated gaseous stream flows through the filter bed. H$_2$S is transferred from the gas phase to the biofilm where chemical reactions occur (Eqs. 1–2). The bacteria most used in the biofiltration of hydrogen sulfide belong to the genus *Thiobacillus*, which uses H$_2$S as an energy source for growth.
\[
\text{H}_2\text{S} + 0.5 \text{O}_2 \rightarrow \text{S}^0 + \text{H}_2\text{O} \quad (1)
\]
\[
\text{H}_2\text{S} + 2 \text{O}_2 \rightarrow 2 \text{H}^+ + \text{SO}_4^{2-} \quad (2)
\]

The selection of the packing material is a key step in a successful biofiltration operation. Organic media, such as compost, peat, and pine bark, are widely used for \( \text{H}_2\text{S} \) treatment because they contain nutrients [17–20]. Inorganic media, such as expanded schist, pozzolan and lava, are also used due to their interesting mechanical behavior [21–24]. Currently, a combination of expanded schist and UP20 (a synthetic nutrient material) can be successfully used to treat gas with a high \( \text{H}_2\text{S} \) concentration (up to 360 ppmv). However, if the biofilter is continuously overloaded by \( \text{H}_2\text{S} \), sulfate accumulation in the biofilter bed leads to a significant decrease in the process performances related to a pH decrease (pH < 1). As a result, the watering flow rate of the biofilter must be increased to avoid sulfate accumulation and maintain the pH > 1 [21]. In order to limit this fall in pH due to sulfate production, new packing materials, naturally basic and low-cost, have to be investigated for effective \( \text{H}_2\text{S} \) removal. The objective is clearly to find a material that could be used as a \( \text{H}_2\text{S} \) scavenger to treat high \( \text{H}_2\text{S} \) concentration in air as well as in biogas. In a first approach, the \( \text{H}_2\text{S} \) concentrations considered are ranged from 50 to 500 ppmv in order to compare the results obtained with data reported in the literature [17,19,25–27]. One such new material is cellular concrete waste. Cellular concrete is a material whose physical and chemical properties could be useful for the removal of \( \text{H}_2\text{S} \). Moreover, the use of waste is an interesting and economic solution for the reduction of air pollution. To the best of our knowledge, this material has never been studied for \( \text{H}_2\text{S} \) treatment. Therefore, the objective of this study was to investigate the removal of hydrogen sulfide using cellular concrete by a physical technique and a bioprocess, \textit{i.e.} biofiltration. The treatment of \( \text{H}_2\text{S} \) was first investigated by filtration of the polluted air through a packed bed of cellular concrete in the absence of biomass in dry conditions (\textit{i.e.} an abiotic filtration). Second, a classic biofiltration was tested using cellular
concrete as the porous support for biomass attachment. In the latter case, the results could be directly compared with performances obtained using expanded schist in the same operating conditions [21].

2 Materials and methods

2.1 Cellular concrete

The properties of cellular concrete depend on its microstructure and composition, which are influenced by the type of binder used, methods of pore-formation and curing. The physical, chemical, mechanical and functional characteristics of different cellular concretes are given in the review paper by Narayanan and Ramamurthy [28].

2.1.1 Composition

The cellular concrete used in this study is a recycled mineral medium, distributed by the company Florentaise in Nantes, France (http://www.florentaise.com) (Figure 1). Its composition was determined using an Energy Dispersive X-ray Fluorescence Spectrometer (EDX-800HS, Shimadzu Company) (Table 1). Cellular concrete is mainly composed of calcium (Ca) and silicon (Si). A complementary analysis was carried out using X-Ray Diffraction (XRD) (Siemens Brüker D5000). From the XRD peaks (not shown), the following phases were identified: quartz (SiO$_2$), calcium carbonate (CaCO$_3$), gypsum (CaSO$_4$, 2H$_2$O), aluminum oxide (Al$_2$O$_3$), iron oxide (Fe$_2$O$_3$), and calcium silicate hydroxide hydrate (Ca$_4$Si$_6$O$_{15}$(OH)$_3$, 2H$_2$O).

Figure 1: Picture of cellular concrete particles
Table 1. Composition (% weight) of cellular concrete and expanded schist as determined by energy dispersive X-ray (the main components are given).

<table>
<thead>
<tr>
<th>Component</th>
<th>Cellular Concrete</th>
<th>Expanded Schist</th>
</tr>
</thead>
<tbody>
<tr>
<td>Water Content</td>
<td>54%</td>
<td>56%</td>
</tr>
<tr>
<td>Mass Density</td>
<td>1248 kg m⁻³</td>
<td>1500 kg m⁻³</td>
</tr>
<tr>
<td>Specific Surface Area</td>
<td>44 ± 0.8 m² g⁻¹</td>
<td>41 m² g⁻¹</td>
</tr>
</tbody>
</table>

2.1.2 Properties

Specific surface area was determined using a Micromeritics ASAP® 2020 gas adsorption analyzer. The specific surface area \( S_{\text{BET}} \) was calculated by the Brunauer–Emmett–Teller (BET) method. Internal porosity and apparent density were measured using a mercury porosimeter, Micrometrics autopore IV 9500. The water retention capacity of a material represents the maximum mass of water retained per gram of dry material. The material was immersed for 1 h in water and then drained for 24 h. The difference in mass was used to calculate its water retention capacity. The pH of the packing material was measured with a pH electrode (Consort) connected to a multi-parameter analyzer Consort C561 (measurement accuracy 0.2 % ± 1 digit).

The specific surface area \( S_{\text{BET}} \) for cellular concrete was 44 ± 0.8 m² g⁻¹. This value is comparable to that of the synthetic material, Biosorbens™ (41 m² g⁻¹), used by Shareefdeen et al. [29] for the biofiltration of H₂S. However, other biofiltration packing materials have low surface areas (< 1 m² g⁻¹) such as sapwood, pine bark and pozzolan [24]. The density determined for cellular concrete was 547 ± 5 kg m⁻³. Some biofiltration packing materials have similar densities, such as peanut shells and bagasse (520 kg m⁻³) [30]. Other media have lower densities, such as polypropylene Pall rings (110 kg m⁻³) [31] and wood bark (96 kg m⁻³) [32], whereas others present a high density, such as expanded schist (1248 kg m⁻³; Table 3) and pozzolan (1500 kg m⁻³) [25]. The water retention capacity for cellular concrete was 56 ± 2 %. This high value can limit the watering rate in the biofilter and avoid compaction of the bed. Moreover, this value is comparable to those obtained for porous lava (47 %) [22], peat (64 %) [33] and UP20 (47 %) [34]. For the pH, cellular concrete was characterized by a value
of 9. Generally, the pH of the medium is close to neutrality, with a few exceptions such as peat (pH = 4.5) [35] and pine bark (pH = 4.5) [24]. During H₂S treatment by biofiltration, sulfuric acid is produced (Eq. 2) leading to an acidification of the packing material. Having a basic medium can be advantageous to limit the pH drop in the packed bed biofilter.

Table 2. Physical characteristics of the cellular concrete used in this study. Comparison with expanded schist [21].

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>1800 kg/m³</td>
</tr>
<tr>
<td>Moisture content</td>
<td>10%</td>
</tr>
<tr>
<td>Specific surface</td>
<td>1000 m²/kg</td>
</tr>
</tbody>
</table>

2.2 Experimental set-up

The laboratory-scale system used for the treatment of H₂S by cellular concrete is shown in Figure 2. It consisted of a PVC column with an internal diameter of 300 mm. The column was filled with cellular concrete (1 m height; volume 70 L). The air flow was generated using a regulated fan (FMV frequency controller 2107, Leroy Somer, Angouleme, France). It passed through a humidification column (if necessary) with an internal diameter of 200 mm, packed with Hiflow rings (1.50 m height). A stream of H₂S (99.7 % purity), controlled by a mass flow controller (Model 5850S, Brooks Instruments, Hatfield, USA), was diluted in the atmospheric air at the outlet of the humidification column. The polluted air was then introduced at the bottom of the biofilter. The H₂S concentration was measured along the column, which was equipped with sampling ports located at the inlet and outlet, and at 10, 30, 50, 70, 90, and 100 cm from the bottom of the column (Figure 2). The H₂S analyzer was an Onyx 5220 device (measurement accuracy ±1 %) from the Cosma Environment SA Company (Passy, France).

Leachate samples were taken periodically from the bottom of the column. Their pH values were measured by a pH electrode (Consort). Sulfate concentration was determined by the
turbidimetric method as described in Standard Methods [36] for the examination of water and wastewater.

**Figure 2.** Schematic diagram of the experimental pilot-scale column used for H₂S removal.

### 2.3 Operating conditions

The parameters used in this paper to describe the operating conditions and for the determination of the removal performances were: (i) the inlet Loading Rate (LR, g m⁻³ h⁻¹); (ii) the Elimination Capacity (EC, g m⁻³ h⁻¹); (iii) the Empty Bed Residence Time (EBRT, s); (iv) the Removal Efficiency (RE, %). All parameters are defined in Table 3. During experiments, the polluted air flow rate was constant at 4 m³ h⁻¹ corresponding to an EBRT of 63 s.

**Table 3.** Parameters used in this study.

#### 2.3.1 Abiotic experiments

The treatment of H₂S was first investigated by filtration of the polluted air through a packed bed of cellular concrete in the absence of biomass (Table 4). Dry material was used for phases 1 to 10. H₂S concentration was sequentially increased from 25 to 250 ppmv (phases 1 to 5). Then, for phases 6 to 10, the H₂S concentration level was changed in order to study the ability of the cellular concrete bed to respond to a significant change in the pollutant concentration, i.e. a change in the inlet loading rate (from 6.4 to 40.0 g m⁻³ h⁻¹).
Table 4. Operating conditions for the filtration of the polluted air through a bed packed with cellular concrete particles in the absence of biomass (air flow rate: 4 m$^3$ h$^{-1}$ corresponding to an EBRT of 63 s).

2.3.2 Biofiltration experiments

The treatment of H$_2$S was also considered by classic biofiltration using cellular concrete as the packing material. The bed (Figure 2) was inoculated with 5 L of a diluted solution of activated sludge (about 50 mg of dry sewage sludge per liter) from a domestic wastewater treatment plant (Tougas, Nantes, France). The cellular concrete was topped with a layer of the synthetic material UP20 (2 cm corresponding to 1.4 L) in order to provide nutrients for the biomass. UP20 contained urea phosphate (CH$_4$N$_2$O, H$_3$PO$_4$), calcium carbonate (CaCO$_3$) (C/N/P molar ratio: 100/10/5) and an organic binder (ELOTEX ST2400; 20 % in mass) from the Elotex Company (Switzerland) [34]. In order to maintain the humidity of the bed material, the biofilter was watered periodically (12 L day$^{-1}$, once a day). During biofiltration experiments, an increase in H$_2$S concentration from 40 ppmv to 360 ppmv was applied at a constant EBRT = 63 s (Table 5).

Table 5. Operating conditions for the biofiltration experiments (EBRT = 63 s).
3 Results and discussion

3.1 Treatment of H$_2$S using cellular concrete in abiotic conditions

The effect of increasing the H$_2$S concentration on process performances is highlighted in Figure 3. For phases 1 to 5, the removal efficiency ranged from 90% (25 ppmv; LR = 2.0 g m$^{-3}$ h$^{-1}$) to 50% (250 ppmv; LR = 20.0 g m$^{-3}$ h$^{-1}$). This finding indicates that a simple filtration of the polluted air through a packed bed of cellular concrete removed a significant proportion of the hydrogen sulfide. Thus, for an H$_2$S concentration of 100 ppmv, which represents a level Immediately Dangerous to Life and Health (IDLH) according to the Occupational Safety and Health Administration (OSHA) [37], the removal efficiency was around 70%, corresponding to an elimination capacity of around 5.6 g m$^{-3}$ h$^{-1}$. This result means that gas filtration through a packed bed of cellular concrete could be used either to treat the pollution directly in the case of moderately polluted air or as a primary treatment for highly polluted air loaded with H$_2$S. The removal efficiencies recorded during phases 1 to 5 were similar to or better than those obtained by biofiltration using natural packing materials, such as sapwood, pine bark or pozzolan [18,24]. In order to confirm the ability of cellular concrete to treat the polluted air and respond to a significant change in pollutant load, the H$_2$S concentration was reduced from 250 ppmv to 80 ppmv (phase 6). Surprisingly, the removal efficiency remained constant at around 50% (Figure 3). A similar procedure was tested again (phases 7 and 8). The increase in the H$_2$S concentration to 250 ppmv led to RE = 40% (against 50% for phase 5) while the return to an H$_2$S concentration of 80 ppmv led to the removal efficiency of 50% being recovered. Such results revealed that reactions occurred between the cellular concrete and H$_2$S, but they could not be strictly related to the change in the loading rate. Two significant increases in H$_2$S concentration were then applied (phases 9 and 10). Whereas a significant drop in the removal efficiency could be expected, RE remained
surprisingly constant at around 30 %. As a result, for the last phase in dry conditions corresponding to an H₂S concentration of 500 ppmv, the elimination capacity was 12 g m⁻³ h⁻¹, which confirms the possible interest of using cellular concrete waste as a primary treatment for H₂S removal.

**Figure 3.** Removal efficiency of H₂S by cellular concrete in the absence of biomass (H₂S concentrations from 25 to 500 ppmv; EBRT = 63 s).

In the light of the results described in Figure 3, the ability of cellular concrete to remove H₂S physically in the absence of biomass had to be explained. Sorption mechanisms and chemical reactions between H₂S and the cellular concrete components were considered. Due to its specific surface area (44 m² g⁻¹), adsorption tests carried out at laboratory scale indicated that cellular concrete is not a good adsorbent for H₂S removal (data not shown). Chemical reactions between H₂S and the cellular concrete components are therefore probable. Calculating the mass balance of the H₂S pollutant between the column inlet and outlet showed that 0.9 kg of H₂S was captured by the material during the 100 days of operation, which corresponded to 42 g of H₂S per kg of cellular concrete during this period (i.e. 4.2 % w/w). An analysis of the elemental composition of the material using an EDX fluorescence spectrometer confirmed that the cellular concrete reacted with H₂S, leading to an increase in the percentage weight of the sulfur component in the medium after treatment (Table 6). Between day 0 and day 100, the amount of sulfur component doubled. Taking the volume of the packing material in the column, the porosity of the bed material and the density of the cellular concrete into account, it can be calculated that the increase in the percentage weight
of the sulfur component was consistent with the amount of \( \text{H}_2\text{S} \) calculated from the mass balance.

**Table 6.** Influence of \( \text{H}_2\text{S} \) treatment on cellular concrete composition.

The corrosion of concrete due to the presence of \( \text{H}_2\text{S} \) is well documented in the literature, especially in sewer systems [38]. However, there are no data suggesting a single-step reaction between \( \text{H}_2\text{S} \) and concrete [39]. There are indications of multi-step reactions leading to the formation of sulfate, gypsum, ettringite and pyrite [38,40,41]. Moreover, the interactions between \( \text{H}_2\text{S} \) and concrete depend on its composition, which is mainly influenced by the type of binder used. Considering the initial composition of the cellular concrete (Table 1), the removal of \( \text{H}_2\text{S} \) can be attributed to the following reactions:

\[
\begin{align*}
\text{Fe}_2\text{O}_3 + 2 \text{H}_2\text{S} + \text{H}_2 & \rightarrow 2 \text{FeS} + 3 \text{H}_2\text{O} \\
\text{FeS} + \text{H}_2\text{S} & \rightarrow \text{FeS}_2 + \text{H}_2 \\
\text{CaCO}_3 + \text{H}_2\text{S} & \rightarrow \text{CaS} + \text{H}_2\text{O} + \text{CO}_2 \\
\text{CaO} + \text{H}_2\text{S} & \rightarrow \text{CaS} + \text{H}_2\text{O} \\
\text{CaS} + 2 \text{CO}_2 + 2 \text{H}_2\text{O} & \rightarrow \text{CaSO}_4 \cdot 2\text{H}_2\text{O} + 2 \text{C}
\end{align*}
\]

The influence of iron was investigated because the addition of iron salts is widely used to control \( \text{H}_2\text{S} \) emissions in sewer systems [38] and several materials containing iron oxide, like sewage sludge, red mud, bottom ashes or steel slags, have been identified as possible iron sponges for \( \text{H}_2\text{S} \) removal [42]. An analysis by X-ray diffraction (XRD) was carried out before and after treatment. The appearance of two new phases (iron (II) sulfide (FeS) and FeS\(_2\) (pyrite)) on the material after treatment was evidenced, confirming chemical reactions between iron and \( \text{H}_2\text{S} \) (Eqs. 3-4). A black precipitate of FeS formed on the cellular concrete.
bed according to Eq. (3). However, the rate of FeS$_2$ formation is slow relative to the rate of dissociation of FeS [43]. Thus, FeS acts as a continuous source for pyrite formation (Eq. 4). These results are similar to those found by Sahu et al. [43] who reported the formation of FeS$_2$ and FeS during the treatment of H$_2$S using red mud. If the ability of cellular concrete to remove H$_2$S was mainly due to the presence of iron and taking into account the initial amount of Fe$_2$O$_3$ in the cellular concrete (1.3 % in weight; Table 1), it can be calculated that the amount of H$_2$S that could be treated by the whole packed bed is around 0.25 kg, i.e. around a quarter of the amount of H$_2$S removed from the air. In other words, even if the total amount of iron present in the packed bed probably reacted with H$_2$S, other reactions (Eq. 5-7) must be considered inside the packing material to satisfy the mass balance of the H$_2$S pollutant between the column inlet and outlet. It should be noted that the possible production of SO$_2$ due to H$_2$S oxidation was taken into consideration in the mass balance of H$_2$S. It is also possible that cellular concrete acts as an iron sponge, allowing the regeneration of iron and the production of elemental sulfur S$^0$. The use of iron oxide for gas desulfurization is a well-known technology. The hydrated iron oxide reacts with H$_2$S forming iron sulfide, thus removing H$_2$S from the gas [44]. Commercial products, such as Sulfamaster™, Sulfur-Rite™, Media-G2™ and Sulfatreat™, are major iron sponge systems in which iron oxides are coated onto different supports [45]. Iron oxides can remove H$_2$S by forming insoluble iron sulfides, which can be regenerated by oxidation with air to give elemental sulfur:

$$Fe_2O_3 + 3 \text{H}_2\text{S} \rightarrow Fe_2S_3 + 3 \text{H}_2\text{O} \quad (8)$$

$$Fe_2S_3 + 3/2 \text{O}_2 \rightarrow Fe_2O_3 + 3 S^0 \quad (9)$$

In such conditions, the packed bed can become clogged by the accumulation of elemental sulfur. However, the regeneration of iron oxide according to Eq. (9) was not evidenced and remains to be demonstrated.
The conversion of concrete to gypsum and ettringite could also explain the ability of this material to remove H$_2$S. According to Eqs. (5-6), calcium carbonate (CaCO$_3$) and calcium oxide (CaO) can also react with H$_2$S to form calcium sulfide (CaS) while according to Eq. (7), calcium sulfide could lead to the production of calcium sulfate (gypsum). The calcium sulfate formed can subsequently react, usually via the formation of monosulfoaluminate, to form ettringite [46]. Ettringite is known to be an expansive material that can cause the disintegration of concrete [39]. In the present case, this potential disintegration is not a problem because the purpose of the study is to use cellular concrete waste for gas treatment. Besides, as cellular concrete is mainly composed of calcium oxide, one can assume that large amounts of H$_2$S will be removed before its complete destruction. To date, and although interactions between cellular concrete and H$_2$S are not fully understood, it can be considered that each gram of the cellular concrete used in this study could remove at least 42 mg of H$_2$S. Such a finding will be useful to design a column filled with cellular concrete for the treatment of gas polluted by H$_2$S. For this purpose, further investigations will be needed to study the behavior of cellular concrete in order to treat H$_2$S in abiotic conditions over a long period.

3.2 Biofiltration of H$_2$S using cellular concrete as the packing material

As cellular concrete can remove H$_2$S in abiotic conditions, it can be expected to be an effective support for biofiltration. To check this assumption, the column was inoculated with 5 L of a diluted solution of activated sludge from a domestic wastewater treatment plant (Tougas, Nantes, France). Consequently, the cellular concrete was not changed between the abiotic and biotic experiments.

3.2.1 Effect of increasing concentrations on H$_2$S removal

The influence of an increasing concentration of H$_2$S on the performances of the process is
shown in Figure 4. Hydrogen sulfide was totally eliminated from the 5\textsuperscript{th} day. A high removal efficiency of H\textsubscript{2}S (> 99 \%) was observed for concentrations up to 133 ppmv. The performances of the process started to decrease from the 43\textsuperscript{rd} day, and this decrease was clearly highlighted from the 60\textsuperscript{th} day when the H\textsubscript{2}S concentration increased from 250 to 360 ppmv (LR from 20.0 to 28.8 g m\textsuperscript{3} h\textsuperscript{-1}). For an EBRT of 63 s, the maximal elimination capacity (EC\textsubscript{max}) obtained with cellular concrete was 17.8 g m\textsuperscript{3} h\textsuperscript{-1}. This value is higher than some data reported with other packing materials used for H\textsubscript{2}S biofiltration. For instance, at an EBRT of 57 s, a maximal elimination capacity of 8 g m\textsuperscript{-3} h\textsuperscript{-1} was achieved in a biofilter filled with sapwood [24]. At EBRT = 51 s, an EC\textsubscript{max} of 8 g m\textsuperscript{-3} h\textsuperscript{-1} was obtained with Pall rings by Kim \textit{et al.} [47]. At an EBRT of 30 s, Shareefdeen \textit{et al.} did not exceed an EC\textsubscript{max} of 8 g m\textsuperscript{-3} h\textsuperscript{-1} using a synthetic medium BIOSORBENS\textsuperscript{TM} as packing material [29]. The comparison with recent biofiltration results reported in the literature at an EBRT close to 63 s (Table 7) indicates that the removal performances of cellular concrete, although less than those of packing materials such as peat or polyurethane foam, are satisfactory. As a result, cellular concrete waste could be an effective and cheap material for the treatment of gas polluted by H\textsubscript{2}S, especially as the mechanical behavior of the packed bed is suitable as shown below.

| Table 7. Examples of recent biofiltration results reported in the literature on the treatment of gas polluted by H\textsubscript{2}S at an EBRT close to 63 s. |

Increasing H\textsubscript{2}S concentrations led to a decrease in pH (Figure 4). This can be explained by the accumulation of sulfuric acid as a by-product of the biological oxidation of H\textsubscript{2}S (Eq. 2). However, even for a high H\textsubscript{2}S concentration (360 ppmv), the pH remained greater than 2, whereas the use of expanded schist as the packing material to remove the same H\textsubscript{2}S
concentration involved a decrease in pH to values lower than 1, leading to a significant fall in the biofilter performances [21]. The initial pH of cellular concrete (pH = 9) highlights the potential positive effect of this new medium as a packing material for H$_2$S biofiltration.

![Figure 4. Removal efficiency of H$_2$S and pH changes in a biofilter packed with cellular concrete.](concentrations of H$_2$S from 40 to 360 ppmv; EBRT = 63 s).](#)

Figure 5 shows a picture of the column filled with cellular concrete some days after the end of the biofiltration experiment. One can observe that the column was stratified into two different layers. At the bottom, in a layer of around 20 cm, the cellular concrete kept its original color but a slight orange color also appeared, whereas above, the whole packing material became black. Such coloration underlines the probable presence of ferric oxide Fe$^{3+}$ and iron (II) sulfide FeS. The presence of Fe$^{3+}$ can be related to the original composition of the cellular concrete (presence of Fe$_2$O$_3$, Table 1) and the regeneration of Fe$^{3+}$ in the biofilter can be explained by the following reactions:

\[ \text{H}_2\text{S} + 2 \text{Fe}^{3+} + 2 \text{OH}^- \rightarrow \text{S}^0 + 2 \text{Fe}^{2+} + 2 \text{H}_2\text{O} \quad (10) \]

\[ 2 \text{Fe}^{2+} + \text{H}_2\text{O} + 0.5 \text{O}_2 \rightarrow 2 \text{Fe}^{3+} + 2 \text{OH}^- \quad (11) \]

According to Eq. (10), Fe$^{3+}$ reacts with H$_2$S to form elemental sulfur. Then, the Fe$^{2+}$ produced can be converted into Fe$^{3+}$ by oxidation with air (Eq. 11). Fe$^{2+}$ can also be biologically oxidized into Fe$^{3+}$ using *Thiobacillus ferrooxidans*. According to Pagella and De Faveri [48], the optimum pH for the growth of *T. ferrooxidans* is around 2.2, which corresponds to the pH values recorded at the end of the experiment (Figure 4). At these low pH values, ferric ion precipitation is avoided. It should be noted that the combined action of a chemical reaction
step (Eq. 10) and a biological oxidation step exploiting the ability of *T. ferrooxidans* was considered by Pagella and De Faveri [48] for H$_2$S gas treatment using two distinct columns. This coupled process was first studied under the name of BIO-SR [49] and is close to the commercial SulFerox® process (a Shell Iron Redox process), in which Fe$^{2+}$ is converted to Fe$^{3+}$ by oxidation with air. It is interesting to note that, in the presence of biomass, cellular concrete can probably regenerate ferric ion. Finally, the stratification shows that different removal mechanisms (physicochemical and biological) occurred simultaneously in the biofilter. The change in pH along the height of the column could explain this stratification. Further experiments are necessary to confirm this interpretation.

![Figure 5. Picture of the column filled with cellular concrete at the end of the biofiltration experiment.](image)

### 3.2.2 Effect of sulfate accumulation on H$_2$S removal

Sulfuric acid is produced during H$_2$S treatment by biofiltration (Eq. 2). Figure 6 illustrates the influence of sulfate accumulation on the performances of the process. For a sulfate concentration lower than 21 mg S-sulfate/g$_{\text{dry medium}}$, H$_2$S was completely removed in the biofilter. These results suggest that a sulfate content of around 21 mg S-SO$_4^{2-}$/g is a critical level for the removal of the pollutant. Above this concentration, the removal efficiency decreased. Thus, a significant drop in the removal efficiency, up to 60%, was observed for a concentration of 360 ppmv corresponding to a sulfate concentration of 30 mg S-sulfate/g$_{\text{dry medium}}$ (Figure 6). Such a decrease could be due to a drop in the microbial activity in relation to a biomass inhibition as well as an H$_2$S mass transfer limitation related to the low pH. Therefore, to maintain a high H$_2$S removal efficiency, it is preferable to work at sulfate concentrations lower than 21 mg S-sulfate/g$_{\text{dry medium}}$. This critical value is close to that found by Yang and Allen [20] (25 mg S-sulfate/g$_{\text{dry medium}}$).
and greater than those found for expanded schist (12 mg S\textsubscript{sulfate}/g\textsubscript{dry medium}) [21] and compost (12 mg S\textsubscript{sulfate}/g\textsubscript{dry medium}) [50]. To avoid sulfate accumulation in the biofilter, Ramirez-Saenz et al. [51] suggested a periodical recirculation of water in the packed bed to limit the concentration to about 8 mg S\textsubscript{sulfate}/g\textsubscript{dry medium}.

**Figure 6.** Effect of sulfate accumulation on H\textsubscript{2}S degradation.

### 3.2.3 Pressure drops

The pressure drops (\(\Delta P\)) were measured between the ports located at 10 and 100 cm from the bottom of the biofilter. Pressure drops in biofilters depend mainly on the superficial gas velocity and particle size [52]. At the beginning of the operation, \(\Delta P\) varied between 2 Pa m\(^{-1}\) and 62 Pa m\(^{-1}\) for gas velocities varying between 56 and 565 m h\(^{-1}\) (Figure 7). After 110 days of operation, the pressure drops slightly increased, to reach values between 2 and 74 Pa m\(^{-1}\) for the same range of gas velocities. This increase in pressure drop (around 20 \%) during H\textsubscript{2}S biofiltration can be explained by: (i) the growth of the biofilm; (ii) a possible deposit of elemental sulfur [52–54]; and (iii) a possible formation of gypsum and ettringite (as described in Section 3.1) leading to a degradation of the cellular concrete. In this case, monitoring the \(\Delta P\) change over a long period will give useful information about the change in the mechanical behavior of the packed bed. Nonetheless, it should be highlighted that these \(\Delta P\) values are mostly lower than those found using other packing materials in different studies, which confirms the interest of using cellular concrete for biofiltration. For instance, using pine bark, \(\Delta P\) varied from 15 to 370 Pa m\(^{-1}\) at gas velocities varying between 65 and 520 m h\(^{-1}\) [25] while for pig manure and sawdust, Elias et al. [54] measured pressure drops between 15 and 460 Pa m\(^{-1}\) at gas velocities ranging between 100 and 200 m h\(^{-1}\). Moreover, the \(\Delta P\) values
recorded for the cellular concrete bed are even lower than those found using expanded schist whose mechanical behavior has been identified as excellent for long operation periods (no attrition, no bed compaction) [21]. In fact, for the same range of gas velocities, ΔP varied from 4 to 105 Pa m\(^{-1}\) (Figure 7), namely 40 % more for a gas velocity of 565 m h\(^{-1}\).

![Figure 7. Pressure drop measurements in the biofilter for gas velocities varying between 56 and 565 m h\(^{-1}\) (symbols: experimental data; dashed line: Ergun’s model [55]). Comparison with data recorded in the same biofilter filled with expanded schist [21].](image)

3.2.4 Comparative study: cellular concrete versus expanded schist particles

The comparison between cellular concrete and expanded schist particles is useful because the latter is recognized as an excellent material for H\(_2\)S removal in terms of removal efficiency and mechanical behavior [23,26]. As indicated above, the pressure drops in the biofilter filled with cellular concrete were lower than those obtained with expanded schist during 110 days of operation at a gas velocity of 565 m h\(^{-1}\) (Figure 7). Nonetheless, this interesting finding remains to be confirmed by studying the behavior of cellular concrete over a long period (> 1 year in operating conditions). By comparing the removal efficiencies of H\(_2\)S obtained by cellular concrete to those reported in Ben Jaber et al. [21] using expanded schist, one can observe that expanded schist showed better performances (Figure 8). Although both materials are efficient for concentrations lower than 133 ppmv (LR = 10.6 g m\(^{-3}\) h\(^{-1}\)), differences can be observed for concentrations higher than 250 ppmv (corresponding to LR > 20.0 g m\(^{-3}\) h\(^{-1}\)). For a loading rate of 28.8 g m\(^{-3}\) h\(^{-1}\) (360 ppmv), the removal efficiencies obtained for expanded schist and cellular concrete were 87 % and 63 %, respectively (Figure 8). The physical
characteristics of the two materials given in Table 1 could explain this difference. For example, the amount of iron oxide Fe$_2$O$_3$ in expanded schist is ten times higher than in cellular concrete. Moreover, the amount of calcium oxide CaO in expanded schist is very low whereas it is a major component of cellular concrete. Assuming that the overall H$_2$S removal was due to both a physical removal caused by the presence of iron oxide and a biodegradation by the biomass, expanded schist should have better properties for H$_2$S treatment than cellular concrete. A packed bed of expanded schist was previously tested for H$_2$S removal in abiotic conditions; the results are extensively described in Dumont et al. [53]. Removal efficiencies from 30 to 50 % were recorded. However, the experiments were not carried out in dry conditions. Moreover, the EBRTs applied (14 to 35 s) were significantly lower than that used in the present study, which prevents a direct comparison. As a result, further experiments need to be carried out to evaluate and compare the performances of both packing materials to remove H$_2$S without biomass in dry conditions. Such an investigation should provide valuable information about the possible mechanisms of H$_2$S removal due to the presence of calcium and iron in both materials, respectively.

**Figure 8.** Comparison of the removal efficiencies of H$_2$S using cellular concrete and expanded schist as packing materials (EBRT = 63 s; pH > 1).

### 4 Conclusion

The removal of H$_2$S by cellular concrete waste as a new packing material was evaluated. At a constant EBRT of 63 s, the results are promising in terms of removal efficiency and pressure drops. This packing material thus presents several advantages, summarized below.
In dry conditions without biomass, cellular concrete can be an effective medium for the treatment of H$_2$S in air. This finding can be explained by chemical reactions that can take place between H$_2$S, CaO, CaCO$_3$ and Fe$_2$O$_3$. The large amount of H$_2$S removed suggests that multiple reactions occur in cellular concrete. Gypsum and ettringite are probably formed. Moreover, cellular concrete could act as an iron sponge with iron regeneration. However, the interactions between cellular concrete components and H$_2$S are not yet fully understood. Consequently, further studies are needed to identify the chemical mechanisms between H$_2$S and this material.

Used as a packing material for H$_2$S biofiltration, cellular concrete waste efficiently treated (RE = 100 %) high concentrations of H$_2$S in air up to 133 ppmv (loading rate up to 10.5 g m$^{-3}$ h$^{-1}$). Physicochemical and biological mechanisms explaining H$_2$S removal seem to occur simultaneously in the biofilter. At an EBRT of 63 s, the maximal elimination capacity (EC$_{\text{max}}$) calculated was 17.8 g m$^{-3}$ h$^{-1}$. The packed bed of cellular concrete also presents a satisfactory mechanical behavior with low pressure drops (30 % lower than those found with expanded schist in the same conditions).

This study is the first experimental evidence that the gaseous pollutant H$_2$S can be removed using cellular concrete waste. Further investigations are necessary (i) to identify the precise chemical and biological mechanisms involved in both abiotic and biofiltration conditions; (ii) to determine the ability of the material to be used over a long period. Investigations could also be performed for the treatment of H$_2$S in biogas with high values of EBRT.

Acknowledgement
The authors would like to thank Franck ROUXEL and the TC-PLASTIC Company for their financial support.

References


Table 4. Composition (% weight) of cellular concrete and expanded schist as determined by energy dispersive X-ray (the main components are given).

<table>
<thead>
<tr>
<th>Elemental composition of cellular concrete (%)</th>
<th>Comparison of composition of cellular concrete and expanded schist</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Composition (%)</td>
</tr>
<tr>
<td>Ca</td>
<td>44.8</td>
</tr>
<tr>
<td>Si</td>
<td>41.8</td>
</tr>
<tr>
<td>S</td>
<td>6.8</td>
</tr>
<tr>
<td>Fe</td>
<td>2.7</td>
</tr>
<tr>
<td>Al</td>
<td>2.0</td>
</tr>
<tr>
<td>P</td>
<td>1.3</td>
</tr>
<tr>
<td>K</td>
<td>0.4</td>
</tr>
</tbody>
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Table 5. Physical characteristics of the cellular concrete used in this study. Comparison with expanded schist [13].

<table>
<thead>
<tr>
<th>Physical characteristics</th>
<th>Cellular concrete</th>
<th>Expanded schist</th>
</tr>
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<tbody>
<tr>
<td>Density (kg m$^{-3}$)</td>
<td>547</td>
<td>1248</td>
</tr>
<tr>
<td>Median diameter (mm)</td>
<td>11</td>
<td>12</td>
</tr>
<tr>
<td>Specific surface area $S_{BET}$ (m$^2$ g$^{-1}$)</td>
<td>44</td>
<td>-</td>
</tr>
<tr>
<td>Internal porosity (%)</td>
<td>64</td>
<td>47</td>
</tr>
<tr>
<td>Initial pH</td>
<td>9</td>
<td>7</td>
</tr>
<tr>
<td>Water retention capacity (%)</td>
<td>56 %</td>
<td>-</td>
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Table 6. Parameters used in this study.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Definition</th>
<th>Nomenclature</th>
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</thead>
<tbody>
<tr>
<td>Loading Rate (LR)</td>
<td>$\text{LR} \left( \text{g m}^{-3} \text{ h}^{-1} \right) = \frac{Q}{V} C_G^{\text{in}}$</td>
<td>$C_G^{\text{in}}$: inlet concentration (g m^{-3})</td>
</tr>
<tr>
<td>Elimination Capacity (EC)</td>
<td>$\text{EC} \left( \text{g m}^{-3} \text{ h}^{-1} \right) = \left( C_G^{\text{in}} - C_G^{\text{out}} \right) \frac{Q}{V}$</td>
<td>$C_G^{\text{out}}$: outlet concentration (g m^{-3})</td>
</tr>
<tr>
<td>Empty Bed Residence Time (EBRT)</td>
<td>$\text{EBRT} (s) = \frac{V}{Q}$</td>
<td>$Q$: gas flow rate (m$^3$ h$^{-1}$)</td>
</tr>
<tr>
<td>Removal Efficiency (RE)</td>
<td>$\text{RE} (%) = \left( \frac{C_G^{\text{in}} - C_G^{\text{out}}}{C_G^{\text{in}}} \right) \times 100$</td>
<td>$V$: bed volume (m$^3$)</td>
</tr>
</tbody>
</table>
Table 4. Operating conditions for the filtration of the polluted air through a bed packed with cellular concrete particles in the absence of biomass (air flow rate: 4 m$^3$ h$^{-1}$ corresponding to an EBRT of 63 s; [H$_2$S] concentrations ± 1%; LR ± 5%).

<table>
<thead>
<tr>
<th>Phase</th>
<th>Duration (days)</th>
<th>[H$_2$S] (ppmv)</th>
<th>LR (g m$^{-3}$ h$^{-1}$)</th>
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<tr>
<td></td>
<td>Dry conditions</td>
<td></td>
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</tr>
<tr>
<td>1</td>
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<td>25</td>
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<td>2</td>
<td>13</td>
<td>50</td>
<td>4.0</td>
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<td>3</td>
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<td>100</td>
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<td>4</td>
<td>12</td>
<td>150</td>
<td>12.0</td>
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<td>5</td>
<td>10</td>
<td>250</td>
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<td>6</td>
<td>5</td>
<td>80</td>
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<td>6.4</td>
</tr>
<tr>
<td>9</td>
<td>5</td>
<td>350</td>
<td>28.0</td>
</tr>
<tr>
<td>10</td>
<td>6</td>
<td>500</td>
<td>40.0</td>
</tr>
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Table 5. Operating conditions for the biofiltration experiments (EBRT = 63 s; $[\text{H}_2\text{S}]$ concentrations ± 1 %; LR ± 5 %).

<table>
<thead>
<tr>
<th>Phase</th>
<th>Duration (days)</th>
<th>$[\text{H}_2\text{S}]$ (ppmv)</th>
<th>LR (g m$^{-3}$ h$^{-1}$)</th>
</tr>
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<tr>
<td>1</td>
<td>7</td>
<td>40</td>
<td>3.2</td>
</tr>
<tr>
<td>2</td>
<td>7</td>
<td>60</td>
<td>4.8</td>
</tr>
<tr>
<td>3</td>
<td>13</td>
<td>80</td>
<td>6.4</td>
</tr>
<tr>
<td>4</td>
<td>16</td>
<td>133</td>
<td>10.6</td>
</tr>
<tr>
<td>5</td>
<td>16</td>
<td>250</td>
<td>20.0</td>
</tr>
<tr>
<td>6</td>
<td>15</td>
<td>360</td>
<td>28.8</td>
</tr>
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Table 6. Influence of H$_2$S treatment on cellular concrete composition.

<table>
<thead>
<tr>
<th>Composition</th>
<th>Before H$_2$S treatment (% weight)</th>
<th>After 100 days of H$_2$S treatment (% weight)</th>
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<tbody>
<tr>
<td>Ca</td>
<td>44.8</td>
<td>42.0</td>
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<tr>
<td>Si</td>
<td>41.8</td>
<td>36.5</td>
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<tr>
<td>S</td>
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<td>15.6</td>
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<tr>
<td>Fe</td>
<td>2.7</td>
<td>2.5</td>
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<tr>
<td>Al</td>
<td>2.0</td>
<td>1.6</td>
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<td>P</td>
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<td>1.2</td>
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<tr>
<td>K</td>
<td>0.4</td>
<td>0.4</td>
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**Table 7.** Examples of recent biofiltration results reported in the literature on the treatment of gas polluted by H$_2$S at an EBRT close to 63 s.

<table>
<thead>
<tr>
<th>Packing material</th>
<th>EBRT (s)</th>
<th>Elimination Capacity EC (g m$^{-3}$ h$^{-1}$)</th>
<th>Removal Efficiency RE (%)</th>
<th>Reference</th>
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<tr>
<td>Peat</td>
<td>60</td>
<td>65.9</td>
<td>90</td>
<td>[38]</td>
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<tr>
<td>Peat</td>
<td>57</td>
<td>25.5</td>
<td>50</td>
<td>[16]</td>
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<tr>
<td>Sugarcane bagasse</td>
<td>49</td>
<td>73</td>
<td></td>
<td>[39]</td>
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<tr>
<td>Coconut fiber</td>
<td>49</td>
<td>68</td>
<td></td>
<td>[39]</td>
</tr>
<tr>
<td>Pine bark</td>
<td>57</td>
<td>10</td>
<td>69</td>
<td>[22]</td>
</tr>
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<td>Sapwood</td>
<td>57</td>
<td>8</td>
<td>50</td>
<td>[16]</td>
</tr>
<tr>
<td>Synthetic medium (UP20)</td>
<td>57</td>
<td>10</td>
<td>93</td>
<td>[22]</td>
</tr>
<tr>
<td>Peat + UP20 (mixed)</td>
<td>57</td>
<td>25.5</td>
<td>80</td>
<td>[16]</td>
</tr>
<tr>
<td>Pozzolan + UP20 (layers)</td>
<td>57</td>
<td>10</td>
<td>74</td>
<td>[22]</td>
</tr>
<tr>
<td>Polyurethane foam</td>
<td>80</td>
<td>56.6</td>
<td>95</td>
<td>[40]</td>
</tr>
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<td>Polyurethane foam</td>
<td>49</td>
<td>66</td>
<td></td>
<td>[39]</td>
</tr>
<tr>
<td>Biomedical encapsulated by Na-alginate and polyvinyl alcohol</td>
<td>51</td>
<td>6</td>
<td>99</td>
<td>[37]</td>
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<tr>
<td>Cellular concrete</td>
<td>63</td>
<td>10.5</td>
<td>100</td>
<td>This study</td>
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</tbody>
</table>

This study
Figure 2: Picture of cellular concrete particles

Figure 2. Schematic diagram of the experimental pilot-scale column used for H₂S removal.

Figure 3. Removal efficiency of H₂S by cellular concrete in the absence of biomass (H₂S concentrations from 25 to 500 ppmv; EBRT = 63 s; RE values ± 2%).

Figure 4. Removal efficiency of H₂S and pH changes in a biofilter packed with cellular concrete (concentrations of H₂S from 40 to 360 ppmv; EBRT = 63 s; RE values ± 2%; pH values ± 0.2).

Figure 5. Picture of the column filled with cellular concrete at the end of the biofiltration experiment.

Figure 6. Effect of sulfate accumulation on H₂S degradation (RE values ± 2%; pH values ± 0.2).

Figure 7. Pressure drop measurements in the biofilter for gas velocities varying between 56 and 565 m h⁻¹ (symbols: experimental data; dashed line: Ergun’s model [48]). Comparison with data recorded in the same biofilter filled with expanded schist [13].
Figure 8. Comparison of the removal efficiencies of \( \text{H}_2\text{S} \) using cellular concrete and expanded schist as packing materials (EBRT = 63 s; pH > 1).
The diagram shows the relationship between relative error (RE) and linear regularity (LR) for two materials: Cellular concrete and Expanded schist. The x-axis represents LR (g m⁻³ h⁻¹), while the y-axis represents RE (%). Each point on the graph corresponds to a specific LR value, with error bars indicating variability. Cellular concrete is represented by red diamonds, and Expanded schist by green squares.
Graphical abstract
Highlights

H₂S removal using cellular concrete waste was investigated

Cellular concrete is efficient for removing H₂S under dry conditions without biomass

Under abiotic conditions, each gram of concrete could remove at least 42 mg of H₂S

Cellular concrete waste is also efficient as a packing material for biofiltration

At EBRT = 63 s, the ECₘₐₓ of the biofilter was found to be 17.8 g m⁻³ h⁻¹