

# Impregnating Activated Carbon with Iron Salts to Increase Hydrogen Sulfide Removal

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## Abstract

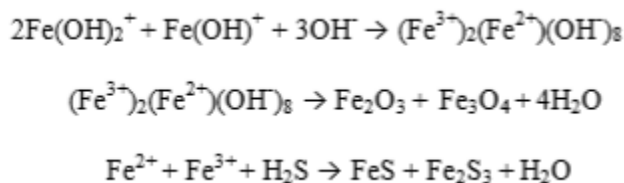
Hydrogen sulfide (H<sub>2</sub>S) emissions from landfills negatively impact surrounding communities. In addition to an unpleasant, rotten-egg odor, inhalation of the gas can cause respiratory distress and irritation. Peak emissions are released from C&D landfills through the decomposition of gypsum drywall. Current removal technologies are costly and often ineffective. The goal of this research was to determine whether heat treatments enhanced the H<sub>2</sub>S removal capacity of iron-impregnated activated carbon. Preliminary research revealed an increase in adsorption capacity through the addition of iron salts, FeCl<sub>3</sub> and FeSO<sub>4</sub>, to activated carbon at a 5:1 carbon to iron ratio. To evaluate the effect of heat treatments on iron oxide formation and subsequent adsorption capacity, samples were divided into heat treatment groups of 250°C, 450°C, and 650°C. Samples were flushed with 100 ppm of H<sub>2</sub>S under N<sub>2</sub> at 250 ml/min for two hours. Over this time duration, samples subjected to higher temperature heat treatments showed increased adsorption capacity. All heat-treated samples demonstrated higher adsorption compared to the baseline non-heat treated samples. Additional research will be conducted to assess the effect of heat treatments above 650°C.

## Introduction

Hydrogen sulfide emissions from construction and demolition (C&D) landfills are a global problem (Bandosz, Bagreev, Adib, & Turk, 2000). In the United States alone, there are over 1,908 active MSW Landfills (EPA, 2017), and 1,540 active C&D landfills (Innovation Waste Services, 2012). With respect to odor, the gas's characteristic "rotten-egg" smell is an issue for property values and human health. Studies reveal that properties adjacent to landfills are reduced in value up to 12.9% (Ready, 2005). Effects on humans vary depending on the H<sub>2</sub>S concentration at the time of exposure. At low concentrations, H<sub>2</sub>S may irritate the respiratory system, whereas at high concentrations respiratory failure may result (Ruth, 1986). Current removal methods include biological, chemical, and physical processes. Though these methods have been proven effective, their associated increased monetary costs and resources has led to research into alternative options (Cherosky & Li, 2013).

## The Role of Activated Carbon with Iron Salts

Activated carbon (AC) has been used for years for H<sub>2</sub>S removal (Boppart, 2010). The carbon adsorbent has a high surface area and microporous structure, making it suitable for capturing pollutants. Research has identified the AC surface chemistry as the predominant factor in determining H<sub>2</sub>S adsorption (Reed, Vaughan, & Jiang, 2000). Thus, modifying the AC's surface with the addition of a metal oxide increases its adsorption capacity (Rodriguez, Chaturvedi, Kuhn, & Hrbek, 1998). Previous research has shown significant H<sub>2</sub>S removal from caustic impregnated carbons (KOH, NaOH) (Boppart, 2010). However, this method is limited due to the material's inherent flammability. AC impregnated with iron salts (AC-Fe) offers a highly efficient, cost effective solution. AC plays a dual role, serving as a catalyst for direct oxidation of H<sub>2</sub>S to elemental sulfur (chemisorption), as well as physical adsorption within the carbon's porous structure (Wang, Zhang, Han, Chang, & Bao, 2013). Iron salts FeCl<sub>3</sub> and FeSO<sub>4</sub> are converted to Fe(OH)<sup>+</sup> and Fe(OH)<sup>2+</sup> through titration to a pH of 9 with NaOH. Due to their inherent thermodynamic instability, they dehydrate to ferrihydrite (Fe<sub>2</sub>O<sub>3</sub>) and magnetite (Fe<sub>3</sub>O<sub>4</sub>). To facilitate this process, the samples are heated, increasing thermodynamic destabilization and promoting the reaction. The aforementioned reaction is depicted below in Figure 1 (Faulconer, Hoogesteijn von Reitzenstein, & Mazyck, 2012).



**Figure 1.** Conversion of hydrogen sulfide via iron oxidation (Faulconer, Hoogesteijn von Reitzenstein & Mazyck, 2012)

## Methodology

### Samples

Carbon samples were developed from manufactured carbon, Calgon Carbsorb 30. The original material had a BET Surface Area of 805 m<sup>2</sup>/g, total pore volume of 0.36 cm<sup>3</sup>/g, and average pore size of 17.90 Å. This carbon was chosen as its large pore volume, large surface area, and

moderate pore size would allow for maximal adsorption of H<sub>2</sub>S. Carbon was oven-dried at 110°C for at least twenty-four hours prior to iron precipitation to remove any excess moisture.

### **Iron Precipitation**

The method used was a modified form of the procedure utilized by Faulconer, Hoogesteijn von Reitzenstein and Mazyck (2012) for mercury removal via iron-impregnated activated carbon. The iron salt solution consisted of ferric chloride hexahydrate (FeCl<sub>3</sub> · 6H<sub>2</sub>O, 99%, pure, granulated) from Acros Organics and ferrous sulfate heptahydrate (FeSO<sub>4</sub> · 7H<sub>2</sub>O) from Fisher Scientific. All samples were impregnated with iron via a 5:1 carbon to iron ratio. This ratio was determined in previous testing, when different ratios of iron to carbon (50:1, 20:1, 10:1, and 5:1) were tested for adsorption capacity (Liu, 2017). To achieve the desired AC:Fe, 7.7452 grams of FeCl<sub>3</sub> and 6.6352 grams of FeSO<sub>4</sub> were added to 100 mL of DI water. The baseline pH of the iron solution was recorded. From there, 20 grams of activated carbon was added. The AC-Fe solution was magnetically stirred for 20 minutes to allow for adequate contact time. The pre-titration pH of the AC-Fe solution was recorded. Once the solution was thoroughly mixed, 10M NaOH was carefully added until a pH of 9 was achieved. The solution was allowed to sit for 60 minutes following titration to ensure pH stabilization. Following this, the solution was filtered and placed in the oven at 110°C for 24 hours prior to heat treatment. Samples were then measured for mass loss or gain.

### **Heat Treatments**

Samples were subjected to one of four heat treatment groups; none (control), 250°C, 450°C, and 650°C. All samples were heated in a furnace under N<sub>2</sub> at 300 ml/min at their respective temperature for four hours. Heat treated samples were stored in the oven at 110°C. Five samples of each treatment were tested for a total of 20 samples.

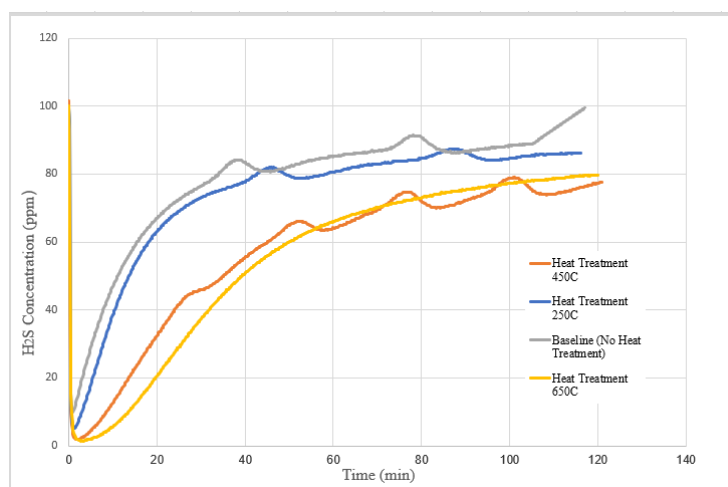
### **H<sub>2</sub>S Test Stand**

One gram of each sample was placed in an H<sub>2</sub>S test bed. A hydrogen sulfide concentration of 100 ppm was assured by establishing a base on a blank line for 20 minutes. After the baseline of 100 ppm was achieved, flow was switched to the H<sub>2</sub>S test bed. This procedure is commonly known as the dynamic test. Thus, for this specific experiment, 0.01% vol (100 ppm) of H<sub>2</sub>S was passed through a column of carbon (diameter 1.27 cm, bed height 0.5 cm) at a flow rate of 250 ml/min. The experiments were carried out at room temperature. The test was stopped after 120 minutes or once the breakthrough concentration (100ppm) was reached. Monitoring of the H<sub>2</sub>S

concentration was accomplished through the Interscan Corporation RM17-500m continuous monitoring system. Adsorption capacities were calculated by integrating the area above the breakthrough curves and taking into account the H<sub>2</sub>S concentration in the inlet gas, flow rate, breakthrough time, and mass of adsorbents.

## Results

The corresponding breakthrough curves are shown in Figure 2. As indicated by the slope of the curve, the maximum H<sub>2</sub>S adsorption occurs within the first 40 minutes. Following this period, removal continues but at a slower rate until it eventually levels off or achieves breakthrough. Figure 2 shows that the 650°C heat treatment group performed the best, removing the most H<sub>2</sub>S within the first 40 minutes and continuing to steadily remove over time. However, at around 55 minutes, the 450°C heat treatment group curve intersects that of the 650°C group, indicating that the former temperature may outperform the 650°C treatment group over longer durations.



**Figure 2.** Hydrogen sulfide breakthrough curves for different heat treatments. A steep slope indicates poor adsorption capacity.

The results of adsorption capacity calculation are depicted below in Table 1. As expected, the baseline samples (no heat treatment) showed the lowest average adsorption capacity at 0.612 mg H<sub>2</sub>S/g AC. This lower value can be explained by the absence of heat, which facilitates the formation of iron oxide groups. The 250°C heat treatment group shows a slightly improved adsorption capacity compared to the baseline. Adsorption capacity is highest in the 450°C group. However, the difference between the 450°C and 650°C groups is small enough to warrant further studies.

**Table 1.** Adsorption Capacity of Heat-Treated Fe:AC Carbons

Heat Treatment (°C)	Average Adsorption Capacity (mg H <sub>2</sub> S / g AC)
None	.612
250	.763
450	1.14
650	1.05

### Discussion/Conclusion

Iron coated activated carbon samples have been modified via different heat treatments to specifically target hydrogen sulfide removal. Current results show an improved adsorption capacity with increasing temperatures. However, the small difference between heat treatment groups 450°C and 650°C warrant further investigation.

Overall, this research aims to use the iron coated activated carbon material in a mat for H<sub>2</sub>S removal from landfills. These findings will aid in developing an iron coated activated carbon that performs optimally for this application. Once the mat has been manufactured, preliminary testing should be conducted at landfill sites to address any environmental effects (weather, temperature, humidity, etc.). Future work will focus on performing additional 450°C /650°C heat treatments, as well as adding an 850°C treatment group. Previous studies have shown temperatures exceeding 850°C to yield higher H<sub>2</sub>S adsorption capacities for nitrogen enhanced activated carbon (Bagreev, Menendez, Dukhno, Tarasenko, & Bandosz, 2004). It will be interesting to see if these findings extend to iron impregnated activated carbon.

### Acknowledgements

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