ournal of Health and Safety at Work 2020; 10(1): 9-13

Received: 2019-01-19
Accepted: 2019-03-06

Removing H_2S gas from the air stream using zeolite ZSM-5 substrate impregnated with magnetite and ferric nanoparticles

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Abstract

Introduction: Hydrogen sulfide (H_2S) is a toxic gas that has adverse effects on human health and equipment. One of the methods for eliminating of H_2S gas is the use of adsorbent substrate. In this study, the effect of adding iron oxides including ferric (Fe_2O_3) and magnetite (Fe_3O_4) nanoparticles to ZSM-5 zeolite substrate was investigated on the efficiency of H_2S elimination from the air stream.

Methods: In this study, Fe_2O_3 and Fe_3O_4 nanoparticles were impregnated in ZSM-5 zeolite in two weight ratios of 3% and 5%. The structural properties of the substrate were studied using XRD, BET and SEM. Then, the efficiency of substrate in removing H_2S from air was studied while H_2S gas was injected in to a pilot setup, in concentrations of 30, 60, 90 and 120 ppm at three bed temperatures of 100, 200 and 300 ^{o}C.

Results: The accuracy of combination and the morphology of inoculated zeolite was confirmed using XRD and SEM. The BET test also showed that the loading of iron oxide nanoparticles on the substrate educed the substrate surface area. The results revealed that increasing the percentage of nanoparticles and increasing the temperature from 100 ° C to 300 ° C increase the time of breakthrough point. The maximum adsorption capacity was obtained equal to 44.449 (mgH_2S/g zeolite) for ZSM-5/$Fe_3O_{4\text{-}5\%}$ substrate at 120 ppm concentration.

Conclusion: Iron oxide nanoparticles inoculated in ZSM-5 zeolite substrate increase the capability of eliminating of H_2S gas at high temperatures and therefore can be used as a suitable method for the elimination of similar pollutants.

Keywords: ZSM-5 zeolite, Hydrogen sulfide, nanoparticles, Fe_2O_3, Fe_3O_4

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1. Introduction

Exposure to Hydrogen Sulfide, as a toxic gas, can cause symptoms such as irritation of the respiratory tract, effects on the genital system, olfactory nerve failure, and at higher concentrations of suffocation and death. In addition to the health effects, H_2S can cause corrosion and damage to the equipment. So, removal is one of the challenges in most industries involved with this gas (1). In the commercial dimension, various processes are used to remove hydrogen sulfide from the gas stream, such as alkaline/amine washing, chemical oxidation, adsorption, biofiltration, and more catalytic oxidation, most of which are costly and energy-intensive or producing secondary contamination associated with it(2). Nowadays, adsorption of gases on adsorbed substrates is one of the most common methods to remove gases. This is done both physically and chemically. Adsorbents often have valuable properties such as high specific surface area, porous structure, uniform porous and porous distribution, high-temperature resistance and selectivity (3). Zeolite substrates are one of the common adsorbents used to remove gases and vapors. Zeolites are one of the porous substrates that have long been widely used in catalytic adsorption or removal processes(4). Among the corrective actions to increase the adsorption capacity of zeolites are the addition of metal catalysts to their surface. Advances in nanotechnology in recent decades have also shown that changes in the size of metallic particles can affect their chemical, physical and catalytic properties.Most studies are available to enhance the catalytic effect of metal particles In nanometric dimensions (5). So far, many efforts have been made by researchers to remove hydrogen sulfide gas using appropriate and cost-effective solutions and The aim of the present study was to investigate the effect of modified ZSM-5 zeolite substrate surface with ferric (Fe2O3) and magnetite (Fe3O4) nanoparticles on H2S gas removal from the air stream.

2. Experimental

The present study was conducted in three phases: 1. Synthesis of hybrid substrates; 2. Determination of structural properties of synthesized substrates; 3. Determination of adsorption capacity of the substrates.

Phase 1. Synthesis of hybrid substrates

In the hybrid substrates synthesis phase, ZSM-5 type zeolites with a diameter of 0.5-1.5 mm which were used in three raw forms, loaded with Fe_2O_3 nanoparticle and loaded with Fe_3O_4 nanoparticle.

Iron nanoparticles were loaded onto the ZSM-5 zeolite substrate according to previous studies with 3 and 5% weight ratios. The five substrates obtained were named With abbreviations $ZSM\text{-}5_{raw}$ ‘$ZSM\text{-}5_{Fe3O4\text{-}3\%}$ ‘$ZSM\text{-}5_{Fe3O4\text{-}5\%}$ ‘$ZSM\text{-}5_{Fe2O3\text{-}3\%}$‘ $ZSM\text{-}5_{Fe2O3\text{-}5\%}$.

Phase 2. Determination of structural characterization of synthesized substrates

The XRD was used to characterize the substrate and confirm the presence of nanoparticles on the substrate, and to investigate the adsorption and desorption isotherm of the nitrogen, specific surface area and pore size of the substrate using BET. SEM was also used to observe the surface and porosity of the substrates.

Phase 3. Determination of adsorption capacity of the substrates

In order to compare the performance of the hybrid substrates synthesized in the removal of hydrogen sulfide gas, a laboratory pilot system under the laboratory hood was used (Figure 1). Concentrations of 30, 60, 90, and 120 ppm of hydrogen sulfide were prepared in the mixing chamber by dilution with clean air. The prepared atmosphere was injected into the reactor at different temperatures containing the substrate at the 1 lit/min flow rate. The adsorption capacity of the synthesized substrates was then investigated by removing the concentrations of 30, 60, 90, 120 ppm H_2S gas at three temperatures of 100, 200 and 300 ^oC. The concentration of H_2S gas in the reactor outlet was monitored until it reached the sorbent breakthrough point (equivalent to 5% of the pollutant inlet concentration from the

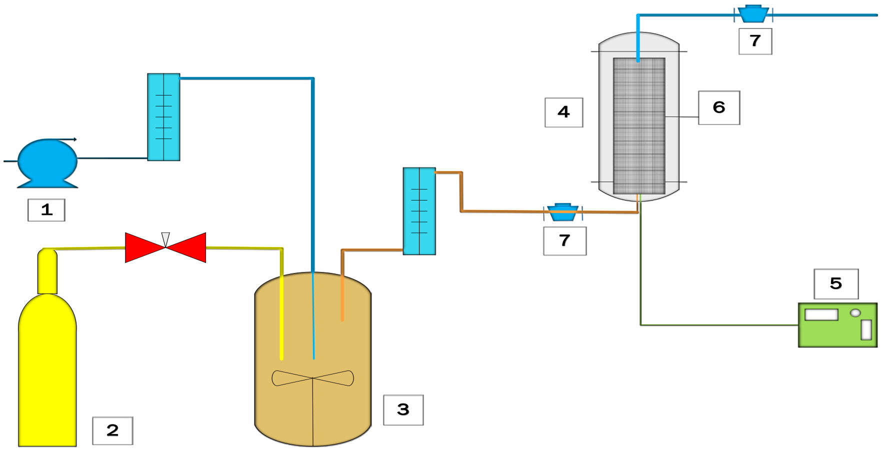

Fig. 1. Laboratory pilot: 1) air blower pump 2) hydrogen sulfide cylinder 3) mixing chamber 4) heater 5) thermostat and temperature control system 6) reactor 7) sampling ports.

reactor outlet). Then, the adsorption capacity of the studied substrates was estimated at the sorbent breakthrough point.

3. Results and Discussion

SEM and XRD images showed that the ZSM-5 zeolite substrate has a spherical and crystalline structure (Fig. 2). The surface area of the studied zeolite substrate was also obtained using the BET instrument and the results showed that the loading of magnetite iron oxide nanoparticles on the surface reduced the specific surface area, volume, and diameter of the substrate cavities. The presence of magnetite and ferric oxide nanoparticles in XRD test results indicated the proper loading of iron oxide nanoparticles on the raw zeolite substrate(6)

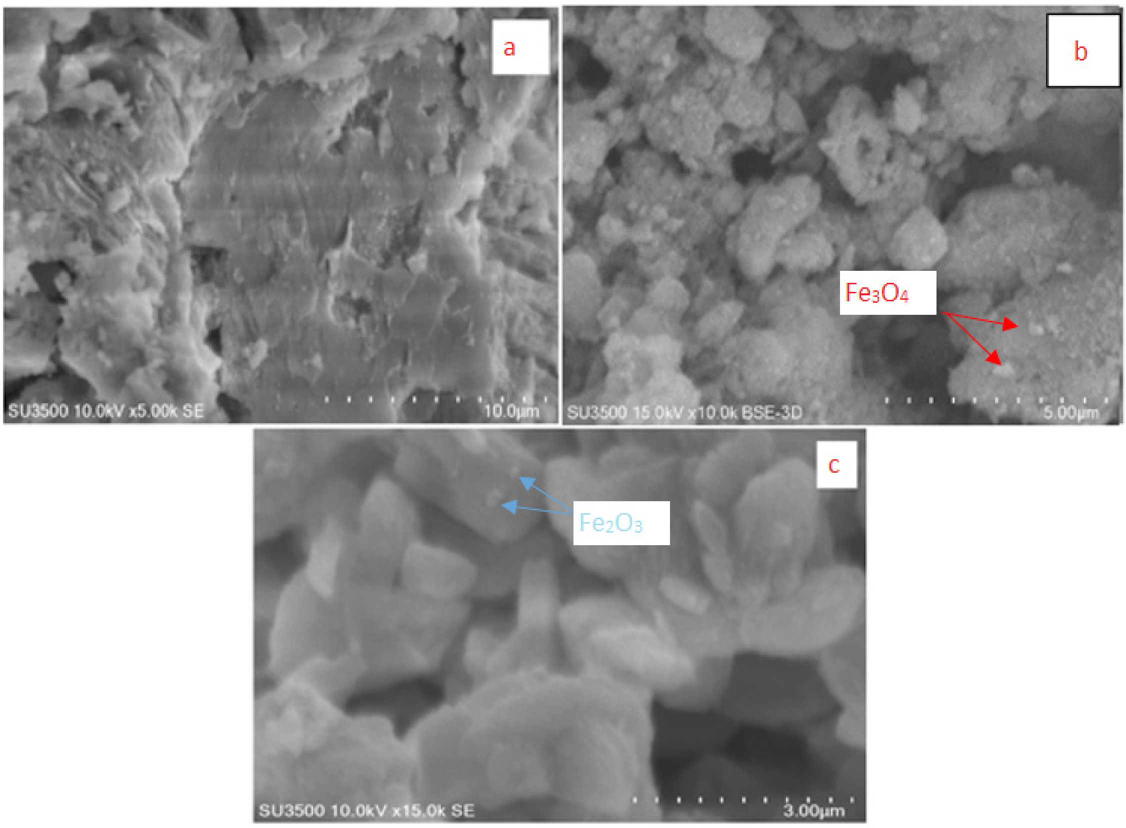

Fig. 2. Pictures SEM: (a) ZSM-5 (b) ZSM-5/ 5% Fe_3O_4 (c) ZSM-5/ 5% Fe_2O_3

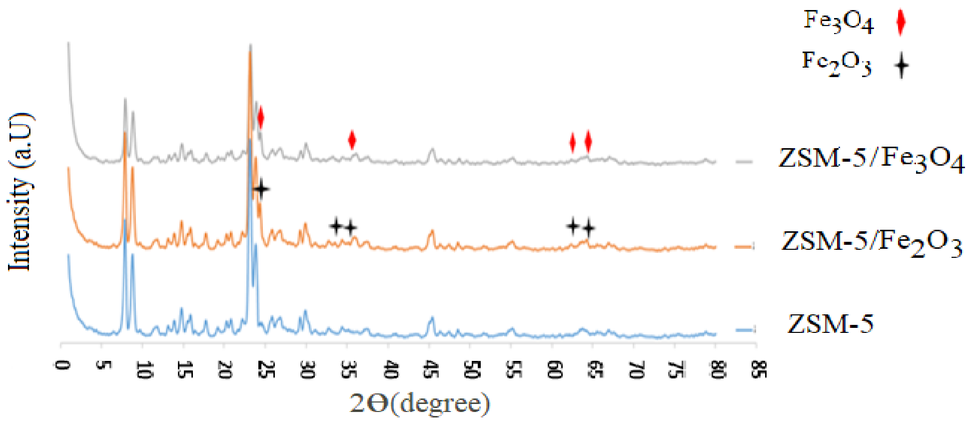

Fig. 4. XRD pattern for ZSM-5 raw substrate and catalyst substrate impregnated with iron oxide nanoparticles (Fe_2O_3-5% and Fe_3O_4- 5%).

. The crystal structure of the substrate after loading of iron oxide nanoparticles was also confirmed by XRD test results (Figure 4). Researchers have also noted in their studies that the specific area of zeolite substrates decreases when nanoparticles are loaded on them due to the ability of nanoparticles to block the zeolite cavities (7), which is consistent with the present study. The results showed that increasing the percentage of loading of iron oxide nanoparticles on the substrate from 0 to 2% for both types of nanoparticles increased the adsorption capacity of the substrate and consequently increased the time to reach the breakthrough point (Fig. 5 - A) and hence two types of substrate $ZSM\text{-}5_{Fe2O3\text{-}5\%}$ and $ZSM\text{-}5_{Fe3O4\text{-}5\%}$ were selected as optimum substrates to investigate the effect of temperature and concentration on hydrogen sulfide gas removal process. The results also showed (Fig. 5 - B) that the time to reach the breakthrough point increased with increasing temperature from 100 °C to 300 °C (P ≤05/0). Therefore, the temperature of 300 °C was selected as the optimum temperature. Comparison of adsorption capacity for ZSM-5Fe3O4-5% and ZSM-5Fe2O3-5% substrates at different concentrations of hydrogen sulfide gas at optimum temperature of 300 °C also showed that substrates coated with magnetite oxide nanoparticles had higher efficiency in sulfide removal. And its highest adsorption capacity was obtained in 120 ppm of H_2S gas concentration (Fig. 5 - C). Studies show that at high concentrations, the thrust force due to partial pressure of the pollutant in the gas phase increases, resulting in an increased probability of distribution of the contaminant between the adsorbent pores and the adsorption rate (8,9).

4. Conclusions

In the present study, the process of removal of different concentrations of hydrogen sulfide by ZSM-5 zeolite loaded with magnetite and ferric oxide nanoparticles was investigated. It can be a good way to remove hydrogen sulfide at high temperatures. Also, according to the results of the study, increasing the nanoparticle loading percentage from 2 to 5% due to the increase of catalytic sites can increase the desulfurization activity. Therefore, it is suggested in future studies to investigate the efficiency of zeolite substrates at different loading rates of different metal nanoparticles to remove hydrogen sulfide or other contaminants present in similar processes.

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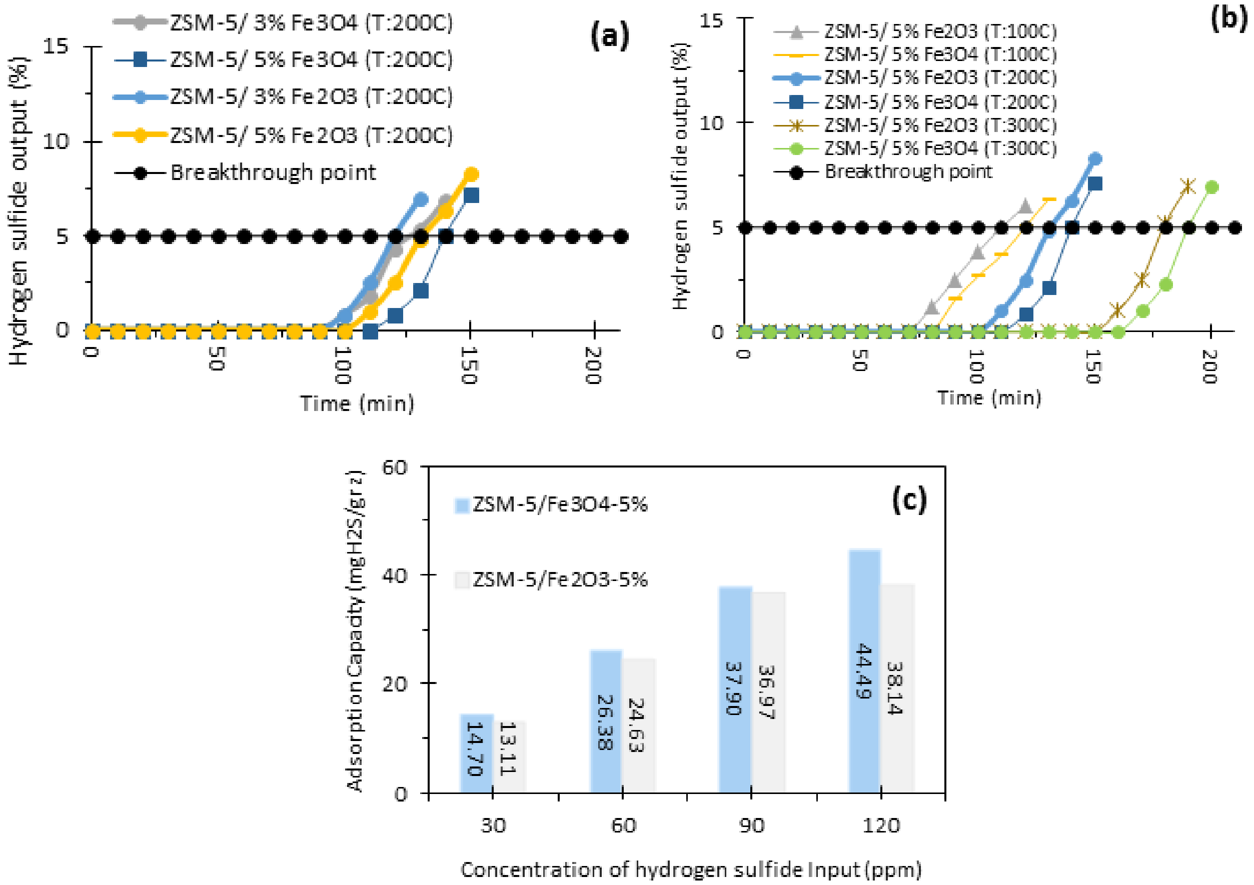

Fig. 5. **(a)** Catalytic substrate breakthrough curve with different loading percentages of iron nanoparticles (Temperature:200 °C – Flow 1 lit/min – concentration 60 ppm). **(b)** The breakthrough curve of the catalyst substrates at different reactor temperatures (Temperature:100, 200, 300 °C – Flow 1 lit/min – concentration 60 ppm). **(C)** Adsorption capacity of catalytic substrates with optimum iron nanoparticle loading percentage and optimum temperature nanoparticles (Temperature:300 °C – Flow 1 lit/min – concentration 30, 60, 90, 120 ppm).

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