Investigation of performance of desulphurization process from liquefied gas

Mohammad Reza Memar Kazerooni¹,², Farshad Farahbod³*

¹Department of Chemical Engineering, Science and Research branch, Islamic Azad University, Sirjan, (IRAN)
²Department of Chemical Engineering, Sirjan branch, Islamic Azad University, Sirjan, (IRAN)
³Department of Chemical Engineering, Firoozabad Branch, Islamic Azad University, Firoozabad, (IRAN)
E-mail: mf_fche@iauf.ac.ir

ABSTRACT

Nano catalytic sulphur removal process is surveyed in this paper, experimentally. Qualification of Sulphur removal from liquefied natural gas process is investigated due to catalytic bed geometries which contains nano ferrite oxide and operation conditions. The purpose of experiments is finding the conditions which lead to the lowest amount of sulphur content in the outlet. Fraction of sulphur content in the outlet to the amount of sulphur in the inlet is considered as the operation criteria, in this work. The effects of operating conditions such as operating temperature and pressure, the amount of sulphur concentration in feed stream, size of nano catalyst, the bed diameter and bed height are investigated. © 2016 Trade Science Inc. - INDIA

INTRODUCTION

The earliest, widespread description of nanotechnology has been referred to the particular technological goal of precisely manipulating atoms and molecules for fabrication of macro scale products, also now is referred to as molecular nanotechnology[1]. A more generalized description of nanotechnology was subsequently established by the National Nanotechnology Initiative, which defines nanotechnology as the study and application of fine particles which are sized from 1 to 100 nanometres in all of the science fields[2].

Sulphur compounds in fuels such as liquefied petroleum gas cause problems on two fronts: they release toxic gases during combustion process, and they damage metals and catalysts in engines and fuel cells[3]. They usually are removed using a liquid treatment that adsorbs the sulphur compounds from the liquefied petroleum gas, but the process is cumber some and requires that the hydrocarbon be cooled and reheated, making the fuel less energy efficient[4]. To solve these problems, researchers have turned to solid metal oxide adsorbents, but those have their own sets of challenges[5]. While they work at high temperatures, eliminating the need to cool and re-heat the fuel, their performance is limited by stability issues. They lose their activity after only a few cycles of use[6]. Sulphur compounds and sulphurs in hydrocarbon and petroleum cuts lead to environmental pollution and corrosion problems in pipelines and storage tanks. So it is necessary to decrease sulphur and sulphurs concentrations in hydrocarbon cuts to international standard levels in petroleum industries. For this purpose, DMD process has been developed as a desulphurization process providing the possibility of desulphurization of different hydrocarbon cuts even crude oil as a feed. Previous studies found that sulphur compounds adsorption works best at the surface of solid metal oxides[7]. So, the authors set out to create a material with maximum surface area. The solution seems
to be tiny grains of ferrite oxide nano particles, uniting high surface area, high reactivity and structural integrity in a high-performance sulphur adsorbent\[8\]. Ferrite oxide has been numerous used for removing of sulphide compounds from liquefied petroleum gas streams in processes like reforming\[9,10\], integrated gasification combined cycle\[10\] and fuel cell\[10\]. Although, Fe2O3 has been well evaluated with sulphide compounds feed stocks, the performance of ferrite oxide nano structure with different operating conditions and structural characteristics in sulphur removal has not been specially evaluated in details. This work is devoted to using experimental design methodology to identify the optimum conditions for sulphur removal by nano ferrite oxide catalysts. Clearly, the nano-sized Fe2O3 is more reactive than the same material in bulk form, enabling complete sulphur removal with less material, allowing for a smaller reactor. The nano particles stay stable and active after several cycles\[9\]. Thermal Swing Regeneration is a common industry process used for desulphurization process. In that process, chemical sponges called sorbents remove toxic and flammable gases, such as rotten-egg smelling hydrogen sulphide from natural gas. The liquefied petroleum gas must first be treated with a solution of chemical sorbents that are dissolved in water. That solution must then be heated up and boiled to remove the sulphide compounds, in order to prepare the sorbent for future use. Once the sulphide compounds are boiled off, the sorbent is then cooled and ready for use again. The repeated heating and cooling requires a lot of energy and markedly reduces the efficiency of the process, scientists say. In the adsorption process by nano Ferrite oxide, sweetening of liquefied petroleum gas is occurred with minimum heat flux comparing with the other sweetening methods. Also, approximately, 70 to 80 percentage of the initial amount of sulphur is removed from the liquefied petroleum gas by the proposed adsorption process. Also, Ferrite oxide catalyst is produced due to feasible method and is not expensive comparing with the other catalysts. So, this method is beneficial. Undoubtedly, the ferrite oxide nano particles as sorbents have large active surface. So, they can be reused again and again. This method will be developed as soon as possible and will be applied in industrial scale.

In this work, a fixed bed reactor is set up which is equipped by nano ferrite oxide catalysts. Some experiments have been held to investigate the effect of different operating pressure, temperature, catalyst diameter, bed height on the performance of sulphur removal.

**MATERIALS AND METHODS**

The materials and methods which are used in this work is investigated in this section.

**Synthesis method of nano-sized Fe2O3**

Using 0.01 M ferric nitrate which is prepared by dissolving Fe(NO\(_3\))\(_3\)·9H\(_2\)O in distilled water. After 15 min duration of ultrasonic the solution is rest for about 15 min. Then, Hydrazine monohydrate with concentration of 0.5 M is added dropwise to the solution under sonication (600 W, 20 kHz) for about 2 hours and the pH value of reaches about pH= 5. Black particles are precipitate after cooling the solution. Then the solution is centrifuged and washed using distilled water and acetone, respectively. Nano powder is reached after 5 h drying time at 70 C in air.

**Set up description**

One laboratory cylindrical vessel equipped with the nano-sized Fe2O3 catalytic fixed bed is applied for sulphur adsorption process, in this work. The process temperature is adjusted by one steam jacket around the vessel. Liquefied petroleum gas stream from a tank reservoir is mixed by sulphur and is fed into the bed containing ferrite oxide nano particles. The inside diameter of bed is designed 15 to 40 cm and the height of the vessel is 70 to 100 cm, respectively. All the instruments and equipments are made of stainless steel.

**RESULTS AND DISCUSSION**

Anyone knows, the sulphide compound is corrosive and toxic, severely. Meanwhile, this component is in several industrial. We know the current technologies use huge resources of energy for removing the sulphur component. Therefore, the researchers try to enhance the performance of sweetening process. So, in this paper the ferrite oxide are applied as nano catalysts for removal of sulphur. This metal oxide is not expensive comparing with the other metal oxides. Several
experiments were held to determine operational conditions that would optimize the amount of sulphur removed from gas in order to gas sweetening. Some major parameters are considered experimentally in the gas desulphurization process by nano particles. The effects of operating conditions, properties of catalytic bed and ferrite oxide catalyst are investigated on the process performance. The ratio of sulphur concentration in the product stream on the initial concentration in the input stream ($C/C_0$) represents the process performance. The purpose of the experiments is to decrease the amount of sulphide compounds below the 4 ppm in the outlet stream. Experimental results are presented in the following Figures.

**The effect of operating temperature and pressure**

The increase in the amount of operation pressure from 6 atm to 9 atm at temperatures of 82°C to 88°C decreases the amount of $C/C_0$. For higher amounts of pressure from 12 atm to 15 atm the increase in the amount of temperature from 86°C to 90°C decreases the amount of $C/C_0$. The strange behavior in the decreasing trend of $C/C_0$ is obtained at temperature of 90°C which the higher amount of $C/C_0$ is obtained at 9 atm comparing with 6 atm. Also, the equal amounts of

![Figure 1: The relation between operating conditions and process quality](image1)

![Figure 2: The effect of bed diameter on the amount of $C/C_0$.](image2)
C/C0 are obtained at 12 atm and 15 atm and at 90 C. The amounts of concentrations which are obtained at 15 atm are higher than those are obtained at pressure of 12 atm in ranges of temperatures from 86 C to 90 C. Figure 1 shows the changes in the amount of C/C0 due to the changes in temperatures and pressures. Results indicate the optimum amount of pressure and temperature is 12 atm and 66 C which leads to the amount of 0.061 ppm.

The effect of bed characteristics

The height and diameter of catalytic bed and also catalyst diameter are considered as the shape effects in this section. Bed length is changed in the range of 6, 7 and 9 cm. Also the bed diameter changes are in range of 1.25, 1.8 and 2.5 cm. The effect of bed characteristics are investigated on the amount of sulphur removal. The optimum amounts of height and diameter of the nano catalytic bed are studied in this section. Operating temperature and pressure is adjusted at 86 °C and 12 atm, respectively.

- Effect of bed diameter

The effect of changes in diameter of bed on sulphur removal is shown in Figure 2. The increase in value of bed diameter provides higher mass transfer area and also leads higher rate of mass transfer. So, the decrease trend is obtained with higher values of bed diameter. The diameter changes from 1.25 cm to 2.5 cm and the ratio of C/C0 changes from 0.048 to 0.04.

CONCLUSIONS

Nano catalyst development in various areas proposes to perform many processes economically and efficiently. The optimum operating conditions and reactor characteristics for sulphur removal with Fe2O3 nano catalyst are investigated experimentally in this work. The process performance is introduced as the ratio of the outlet concentration of sulphur per the inlet concentration and is presented as value of $C / C_0$. Experiments are done in the cylindrical reactor in different temperature, pressure, bed height, bed diameter and ferrite oxide catalyst diameter to find the best condition to reach the value of 0.04 for sulphur at the product. The experimental results indicate that the optimum adsorption performance is obtained at 86 Centigrade degree and 12 atm operating conditions using 80 ferrite oxide catalysts when the bed height is fixed 7 cm and the bed diameter is 30 cm.

REFERENCES


