

Effect of reaction parameters on the quality of captured sulfur in Claus process



H. Selim^a, A.K. Gupta^{a,*}, A. Al Shoaibi^b

^a Department of Mechanical Engineering, University of Maryland, College Park, MD 20742, United States

^b The Petroleum Institute, Abu Dhabi, United Arab Emirates

HIGHLIGHTS

- Examined quality of collected sulfur deposits under different reaction conditions.
- All sulfur deposits were analyzed and proved to be of cyclo-S₈ (α-sulfur) allotrope.
- Equivalence ratio did not show significant effect on the quality of sulfur deposits.
- Carbon dioxide did not show considerable effect on sulfur deposits quality as well.
- Methane and propane caused sulfur deposits to contain considerable amounts of carbon.

ARTICLE INFO

Article history:

Received 19 June 2012

Received in revised form 3 December 2012

Accepted 4 December 2012

Available online 29 December 2012

Keywords:

Claus process

Sulfur quality

H₂S combustion

Sulfur chemistry

ABSTRACT

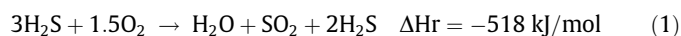
Quality of sulfur deposits collected from hydrogen sulfide combustion has been examined. Experimental conditions representing selected practical systems have been investigated. Combustion of hydrogen sulfide was allowed to occur under the different conditions with sulfur deposits collected for analysis. Experimental conditions represented effect of equivalence ratio of rich (Claus) conditions and stoichiometric conditions; effect of contaminants, other than H₂S, in the acid gas stream; combustion of H₂S with hydrocarbon fuels (methane and propane). Sulfur deposits from H₂S combustion under various conditions were captured and analyzed using X-ray powder diffraction and laser induced breakdown spectroscopy (LIBS) diagnostics. X-ray powder diffraction was used to determine the type of deposited sulfur allotrope, while LIBS was used to identify the elemental analysis of deposited sulfur.

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1. Introduction

Hydrogen sulfide is known to be present in different quantities in crude natural gas extracted from the gas/oil wells. The use of crude natural gas in any chemical to thermal energy transformation process, e.g. combustion, furnaces or power system results in the formation of acid gases, such as, SO₂ and SO₃ which are then transformed to sulfurous and sulfuric acid, if released to the atmosphere. In addition, the presence of these gases lowers the dew point of the combustion gases so that exhaust gases must be released at higher temperatures. Therefore, hydrogen sulfide must be removed from natural gas prior to its utilization. Amine extraction process [1–3] is used for the removal of acidic gases, mainly H₂S and CO₂, from crude natural gas wherein alkaline-based organic compounds are used to absorb H₂S and CO₂ from the fuel stream. Although the concentration of the absorbed H₂S is fairly low, it is crucial that hydrogen sulfide undergoes treatment process to hin-

der its harmful effects on both the human health and environment from the industrial process. Claus process [4–8] is commonly used for the treatment of hydrogen sulfide wherein reaction between H₂S and O₂ occurs under rich conditions ($\Phi = 3$) to form elemental sulfur. During this reaction one third of H₂S is burned to form SO₂ (reaction (1)). Afterwards, the reaction continues between the so formed SO₂ and unburned H₂S to form sulfur (reaction (2)) which is then captured in liquid (in hot region) or solid form (in cold region). Practically, the process is divided into two main stages consisting of thermal stage and some catalytic stages. Both stages have same chemical reaction, but catalysts are used for the later stage wherein H₂S concentrations are considerably low.



Sulfur collected from Claus process is considered a value-added product since it has many usage in several industrial and other applications. Selim et al. [9] analyzed the sulfur deposits collected from the reactor walls maintained at relatively low-temperatures.

* Corresponding author.

E-mail address: akgupta@umd.edu (A.K. Gupta).

These results revealed that sulfur deposits primarily consist of cyclo-S₈ (α -sulfur) with orthorhombic crystal structure. However, the quality of collected sulfur was not examined. In this paper we investigate the quality of sulfur collected with respect to reactor conditions (gas stream composition and equivalence ratio).

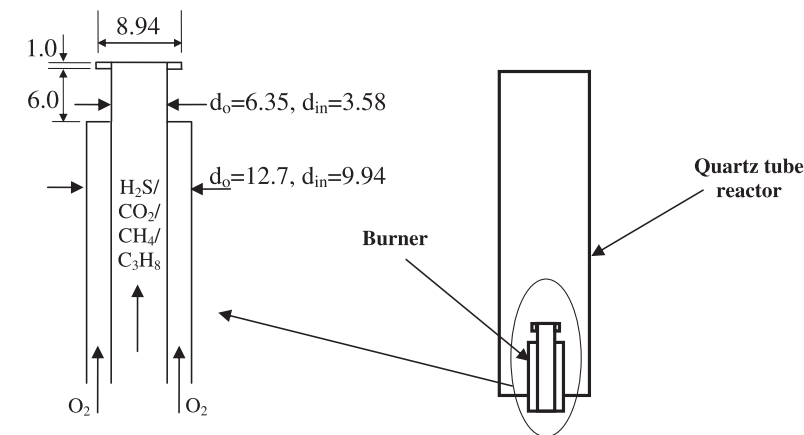
2. Experimental setup and diagnostics

A schematic diagram of the combustor used in this study is shown in Fig. 1. The facility consists of a quartz tube reactor of 19 cm length and 4 cm inner diameter. A double concentric tubular burner was designed and used for all experiments reported here. In addition, a bluff body was used to anchor the flame. In all the experimental results reported here, oxygen was used as an oxidizer and it was injected into the outer annulus of the burner. The hydrogen sulfide was introduced into the central tube of the burner wherein it was premixed with carbon dioxide, methane, or propane prior to their injection into the burner. Dimensions of the burner are given on Fig. 1. All flow rates were adjusted using thermal flow controllers. Quartz reactor was installed in a steel housing base which was used to collect the condensed sulfur. Fig. 2 shows a photograph of the sulfur formed with deposited onto the reactor housing. Average time of each experiment was about an hour in order to allow accumulation of sizeable amounts of sulfur deposit

form the tests. Laser induced breakdown spectroscopy (LIBS) was used for most of this study to analyze the chemical structure of deposited sulfur. The LIBS setup consisted of seven-channel spectrometer equipped with seven CCD cameras. The spectrometer covered a band of wavelengths extending from 200 nm to 970 nm, equally distributed on the seven CCD cameras. A 532 nm Nd:YAG was used to excite the samples inside the LIBS chamber. The sampling chamber is connected to the seven-channel spectrometer via a fiber optic cable. Fig. 3 shows a schematic diagram of the LIBS setup. Since LIBS gives only the elemental analysis of the sample, X-ray powder diffraction technique has been used primarily to determine the allotrope of collected sulfur.

3. Experimental conditions

Selected experimental conditions have been reported in this study. Firstly, we investigated the effect of reactants equivalence ratio on the quality of collected sulfur deposits. Two equivalence ratios examined here (stoichiometric and Claus conditions). Secondly, effect of other contaminant (carbon dioxide) likely to accompany hydrogen sulfide in the acid gas stream is examined. Finally, effect of hydrogen sulfide combustion in presence of hydrocarbon fuels (methane and propane) on the quality of



All dimensions are in millimeters

Fig. 1. A schematic diagram of the experimental reactor.

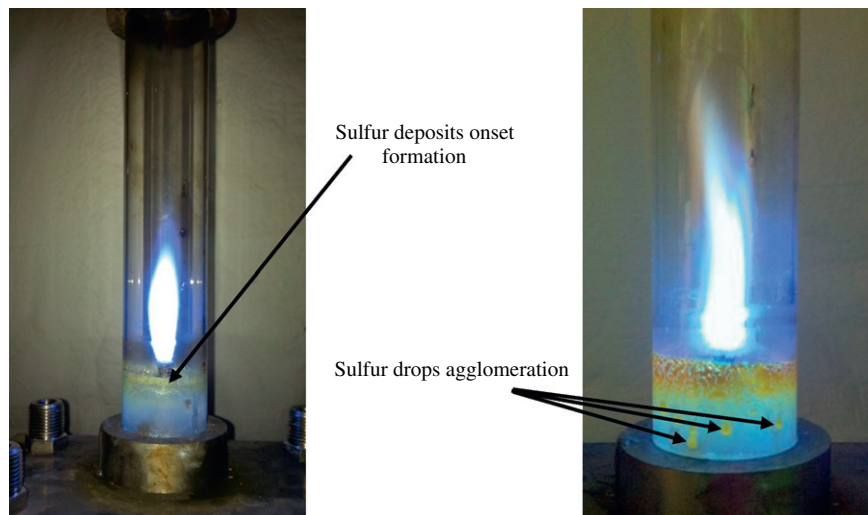


Fig. 2. The onset of sulfur formation, agglomeration and precipitation in reactor housing.

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