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Impact of H_2S Content and Excess Air on Pollutant Emission in Sour Gas Flares

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Abstract

In sour gas flares, H_2S content like any other components in inlet gas influences adiabatic flame temperature, which, in turn, impacts on the pollutant emission. Wherever flame temperature increases, the endothermic reaction between O_2 and N_2 is accelerated, which means higher NO_x emission to the atmosphere. In this work, we developed an in-house MATLAB code to provide an environment for combustion calculations. Then, this written code was used to perform sensitivity analyses on H_2S content, air temperature, and excess air ratio in sour gas flares. We used Environmental Protection Agency (EPA) reports to assign weighting indexes to each air contaminant according to its harmfulness to environment; thereafter, sour gas flaring conditions were optimized for two real field case studies, namely Ahwaz (AMAK) and South Pars, to reach the minimum integrated pollutant concentrations. The results show that each 2% increase in the H_2S content of the entrance feed may produce 0.3% additional SO_x in the exhaust. The results also confirm that decreases of 20 °F and 50 °F in the oxidant temperature cause NO_x emission to reduce by 0.5% to 1% respectively. Finally, to verify and validate our results acquired from the written MATLAB code, FRNC 2012 industrial software was used to duplicate the oxidation results for the two sour flare case studies.

Keywords: Sour Gas Flares, EPA Environmental Reports, Pollutant Emission, FRNC Software

Introduction

In 2010, ten countries were accounted for 72% of the total oil and gas flare stacks worldwide. Top five leading countries were Russia (27%), Nigeria (11%), Iran (8%), Iraq (7%), and the United States (5%). Satellite data show that global gas flaring has decreased by about 20% in recent years. The most significant reductions in terms of volume were made in Russia (up to 40%) and Nigeria (up to 29%). One can expect Iran to rise to top of the list in the next few years. The existence of H_2S and CO_2 within the produced oil or natural gas is called sourness, which gives a high corrosive trait to flow, especially in the presence of H_2O. Whenever possible, it is preferred to install sweetening units to reduce H_2S contents in production lines. However, processes are not 100% effective, and some tail

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gases with high H_2S contents are always flared. The formation of the sulfur oxides like SO_2 and SO_3 and the nitrogen oxides like NO and NO_2 during sour gas combustion causes air pollutant emissions to the atmosphere. Various air pollutants and greenhouse gases (GHG's) show different levels of harmfulness to the environment, while these effects can change yearly depending on several factors such as local climates and the ozone layer condition. Hence, several attempts and progresses were made in achieving air quality standards. Organizations such as Environmental Protection Agency (EPA) gather information and process data and publish yearly reports on the current conditions of the atmosphere and local environments. Several calculational and experimental studies were performed in the literature to simulate combustion reaction in flares and to investigate the effects of different operational parameters on the process efficiency. McMahon et al. (1994) studied several flare stacks and reported that the type of gas, average heating values, and ambient temperature are effective in performance and pollutant emission from flares. Al Rubai et al. (1999) used mathematical kinetic models to express oxidation reaction formulas for sour feed in flares located at Al Daura refinery and estimated SO_2, CO, and NO_2 concentrations within the flue gas. Gzar et al. (2009) employed mathematical formulas which used EPA impact factors to develop codes in Visual Basic programming language for simulating combustion in flares. The concentrations of CO and NO_x compounds in flue gas were calculated and compared with the experimental data from some real field stacks in Basra state, southern Iraq. Talebi et al. (2014) performed an experimental study over intake gas composition and excess air using pilot scale flare. It was shown that whenever the contents of light hydrocarbon decrease, more CO emits from flare, while the amount of NO_x emission remains constant. Kahforoushan et al. (2010) measured CO_2 concentrations in the outlet flow of several gas flares which contain various composition gases. The measured values were then compared with the EPA reports on flare emission factors. The results showed high difference wherever CO_2 existed in feed. Kahforoushan et al. (2014) performed an experimental study on the effects of parameters such as fuel and oxidant flow conditions. It was confirmed that NO_x formation is greatly dependent on oxidant temperature. The results also showed that CO and CO_2 concentrations in flue gas were only affected by the inlet composition. Umukoro et al. (2015) developed computer programs in MATLAB to simulate the combustion of oil and gas by considering several types of reactions, including complete and incomplete combustion of sweet and sour gases. Their code calculated the NO_x emission in types of NO, NO_2, and SO_x formation in type of SO_2 only. Then, they compared the calculated concentrations of emitted GHG and pollutants in twelve samples from all around the world and illustrated that the entrance composition impacted on the formation of various air contaminants. Ismail et al. (2014) wrote MATLAB codes and performed calculations to show the influence of various parameters such as excess air ratios, natural gas type, carbon mass contents, fuel impurities, and flare systems on the composition of the flame outputs. The calculations were founded on the consideration of various combustion reaction types, including CO_2, CO, NO, NO_2, and SO_2 formations. Finally, several comparisons were made among the predicted emissions in some real field flare stacks from all over the world to investigate the effects of the entrance composition on the dispersion rates of air contaminants. Abdulkareem et al. (2009) studied environmental influences of the various flue gas components by accounting on complete and incomplete fuel oxidation and NO_x and SO_x formations. The dispersion of the air pollutant components such as SO_2, NO_2, and CO_2 against the distance from flare stacks was experimentally measured in some oil fields in Nigeria. The illustrated results showed that CO_2 and NO_2 concentrations were reduced by an increase in distance, while conversely SO_2 concentrations were measured higher farther from the flare stacks. Kahforoushan et al. (2008) used MATLAB to model sour gas flares in petroleum industry which emits pollutants like CO, CO_2, NO_x, and SO_x. The impact of operating parameters such as atmospheric conditions on long term reactions of air contaminants was also estimated. Susu et al.

(2005) utilized mathematical models to calculate pollutant concentrations in the flue gas and the neighbor environment considering sulfur, nitrogen, and carbon oxides. The late reaction of air contaminants with excess air during dispersion was taken into account. Abdulkareem et al. (2005) used Visual Basic to simulate CO, NO, and SO_2 emission from flares. It was concluded that the air temperature and sour gas composition influence the pollutant emission. In this study, we firstly developed some codes in MATLAB R2015 to provide an environment for combustion calculations. In order to do so, we considered the independent oxidation of the fuel components such as C_xH_y, H_2S, O_2, and N_2. The written code was then used to perform sensitivity analyses on H_2S contents, excess air temperatures, and air to fuel ratio (AFR) to obtain an optimum condition in which the pollutant emission (SO_x+NO_x) is minimized. The novelty of this study compared to previous works in the literature is that, in our viewpoint, GHG's like CO and CO_2 are not directly dangerous to the ecological systems, so we presume one may need to minimize SO_x and NO_x in flares. The other difference is that we used Environmental Protection Agency (EPA) reports (2012-2013-1999) to weigh the detriments caused by each pollutant, and we proposed some optimum excess air ratios for sour gas flares as a function of their H_2S contents.

Computational methodology

In this paper, our focus is on the magnification of H_2S existence within the flare entrance and its impact on flame temperature and pollutant emission. In sour gas flares, H_2S is oxidized to SO_2 and SO_3 compounds which participate in acidic rains. Furthermore, the presence of H_2S affects NO_x emission via influencing adiabatic flame temperature. In addition to fuel composition, air temperatures and air-to-fuel proportions, which are also presented in excess air ratios, determine the adiabatic flame temperature of the flares. In order to calculate the amount of greenhouse gases and the air pollutants emitting from flare stacks, we need to simulate the combustion reaction of the fuel mixture and determine the composition of the flue gas. Each hydrocarbon component within the inlet sour gaseous phase participates in an exothermic reaction, which, in the presence of enough excess air, will go through complete combustion yielding only carbon dioxide and water vapor. The general form of the complete combustion we adjusted in our written MATLAB code is given by Equation 1:

$$C_xH_y + zO_2 + 3.77zN_2 + iCO_2 + jN_2 \rightarrow xCO_2 + 0.5yH_2O + 3.77zN_2 + iCO_2 + jN_2 \qquad (1)$$

Nitrogen which participates in NO_x formation comes from two main sources, firstly from the oxidant mixture and secondly from N_2 contents within the gaseous fuel. Similarly, the GHG products of the complete oxidation, namely CO_2 and H_2O, which appear in the flue gas may come from fuel oxidation or from the entrance. Conversely, in situations with limited oxidant, hydrocarbon components will go through an incomplete combustion, in which hydrogen and carbon monoxide are produced in addition to carbon dioxide and water vapor. The general form of the incomplete hydrocarbon oxidation we used in our written MATLAB code reads as:

$$\begin{aligned} &C_xH_y + zO_2 + 3.77zN_2 + iCO_2 + jN_2 \\ &\qquad \rightarrow wCO_2 + vCO + pH_2O + qH_2 + 3.77zN_2 + iCO_2 + jN_2 \end{aligned} \qquad (2)$$

Reaction yields and enthalpy changes relevant to fuel component combustions were calculated from thermodynamics, in which the difference among the total Gibbs free energy of the products and those of the reactants determines the exothermic or endothermic condition of each oxidation. Conversely, the rate of the conversion of each reactant was calculated using kinetics via considering especial orders and constants for each fuel compound within its combustion reaction. The enthalpies calculated for all the hydrocarbon combustions showed negative values, which exhibit exothermic oxidations, while the calculated rates represented the instant conversion of reactants to products. On the contrary, N_2, O_2, and H_2S oxidations to NO_x and SO_x disobey the above rules. Nitrogen oxides, which are also called NO_x, are emitted from flares and are mainly consisted of NO (about 90% to 95%) and much less of NO_2 (about 5% to 10%). Other nitrogen oxides like nitrous oxide appear only in negligible amounts. Nitrogen oxides which are formed in combustion reactions also came from two sources, namely air and fuel. Two main formation mechanisms which are called prompt and thermal contribute in the formation of nitric oxide from the oxidant resource. The prompt mechanism presents that firstly some available radicals react with nitrogen molecule to form HCN, HN, and CN molecules, and then these compounds are easily oxidized to nitric oxide. Oppositely, the thermal mechanism presents that some high energy molecules dissociate oxygen molecule to form oxygen radical, and this radical then attacks nitrogen molecule to form nitric oxide. Regardless of what extent each of the mentioned mechanisms participates in O_2 and N_2 oxidation, we adjusted NO endothermic formation equation in our code in the form of Equation 3:

$$xO_2 + xN_2 \rightarrow 2xNO \tag{3}$$

However, the slow spontaneous conversion of nitric oxide to nitrogen dioxide was considered to take place in small amounts according to the exothermic equation described by:

$$xNO + xO_2 \rightarrow 2xNO_2 \tag{4}$$

It is noticeable that the overall NO_x formation is definitely endothermic, so wherever adiabatic flame temperature is high, the oxidation is accelerated, leading to more NO_x emission.

Sour flares contain H_2S component that is oxidized into sulfur dioxide and trioxide. If oxygen is present in high amounts, the dominant product is SO_3, while limited oxygen tends to form more SO_2. Both reactions are exothermic, and while SO_2 participates in H_2SO_3 formation in acidic rains, SO_3 conspires against H_2SO_4 formation which shows more corrosive characteristics. The oxidation reactions are similar with the only difference that SO_3 formation consumes more oxygen. We considered adjusting SO_x formation equations in our MATLAB code in the forms of below equations:

$$2xH_2S + 3xO_2 \rightarrow 2xSO_2 + 2xH_2O \tag{5}$$

$$xH_2S + 2xO_2 \rightarrow xSO_3 + xH_2O \tag{6}$$

To provide an environment for calculations, we developed an in-house code in MATLAB R2015 which considers identical oxidant distribution among all the components. Under the existence of excess air, the written code automatically enables complete combustion option and calculates the enthalpy change within each independent reaction with the aid of the reference Gibbs free energies associated to each product or reactant component. The rate of reactions were defined as power law equations in which each component concentration influences conversion rate with a power factor equal to its stoichiometric coefficient within the reaction equation, i.e. its reaction order. The adiabatic flame temperature was then calculated according to total enthalpy change and intake air temperature. Finally, the conversion of reactants to NO_x and to SO_x was calculated. Almost the same progress happens when the air resources are limited. In such conditions, the written code automatically activates the incomplete combustion option, which replaces the equations relevant to the complete reaction with those of incomplete combustion. Total enthalpy change and adiabatic flame temperature were calculated in the same manner. Only difference is that the code coincidentally calculates the amounts of the additional products like CO and H_2 which appear only in the incomplete combustion.

Afterwards, to investigate the effects of parameters affecting contaminants emission from sour flares, we performed several sensitivity analyses on H_2S content within the entrance stream, the higher concentration of which is expected to accelerate pollutant emission, on excess air ratio, which determines adiabatic flame temperature and consequently impacts on NO_x and SO_x emission, and on the inlet air temperature. One must notice that since in combustion reactions, oxidant is consumed in much larger volumes than fuel, inlet stream temperature is less affected with fuel rather than air. Based on the results of the analyses, we proposed some optimal excess air ratios as functions of H_2S content and intake air temperature to minimize pollutant emission from sour flares. Figures 1 and 2 show the schematic flowchart of the calculations and the developed MATLAB code. The calculated results were then verified and validated with FRNC 2012 industrial software using its Combustion Simulator Module.

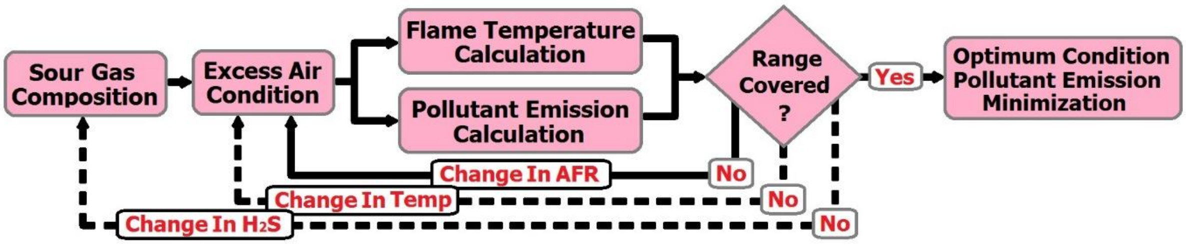
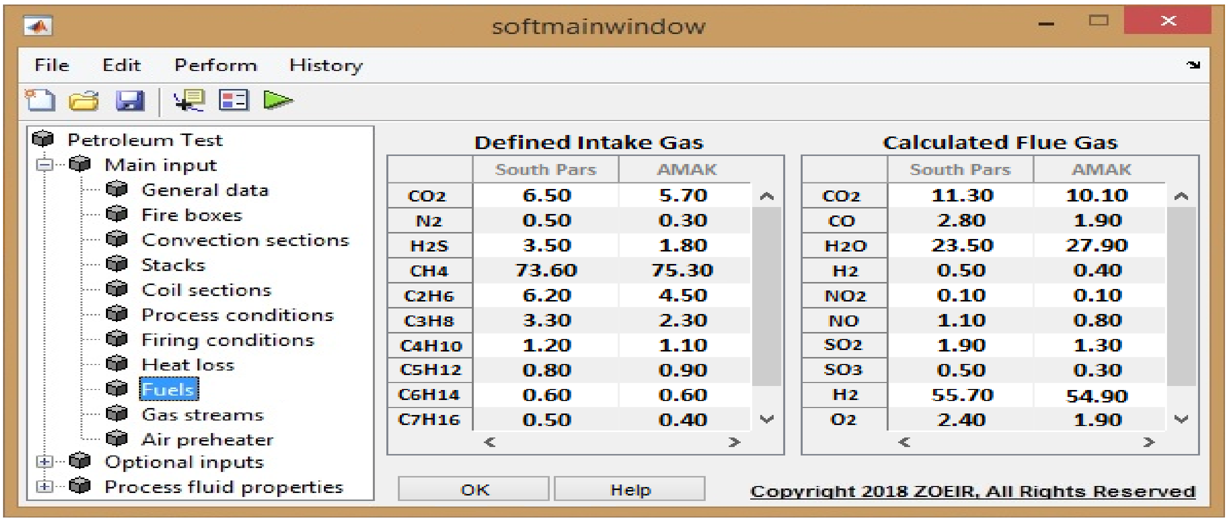


Figure 1

A schematic flowchart explaining calculational procedure within the MATLAB code.



Defined Intake Gas

	South Pars	AMAK
CO_2	6.50	5.70
N_2	0.50	0.30
H_2S	3.50	1.80
CH_4	73.60	75.30
C_2H_6	6.20	4.50
C_3H_8	3.30	2.30
C_4H_{10}	1.20	1.10
C_5H_{12}	0.80	0.90
C_6H_{14}	0.60	0.60
C_7H_{16}	0.50	0.40

Calculated Flue Gas

	South Pars	AMAK
CO_2	11.30	10.10
CO	2.80	1.90
H_2O	23.50	27.90
H_2	0.50	0.40
NO_2	0.10	0.10
NO	1.10	0.80
SO_2	1.90	1.30
SO_3	0.50	0.30
H_2	55.70	54.90
O_2	2.40	1.90

Figure 2

A screenshot of the developed MATLAB GUI code to calculate flue gas compositions.

Results and discussion

In order to perform the sensitivity analyses, we firstly selected two real field sour gas case studies from southern Iran, namely AMAK resource in Abadan refinery, which originally comes from Asmari field in Ahwaz, and another resource coming from South Pars field in the Persian Gulf. Next, we performed the sensitivity analyses on H_2S content within the flare entrance via imaginarily changing H_2S to CO_2 ratio while keeping the overall composition of the sour components constant. In this way, one can assure that the adiabatic flame temperature is least affected with changes in the inlet fuel composition. Then, we fed all the defined cases into the written MATLAB code and calculated the optimum excess air ratios and the relevant flue gas compositions. The exhaust gas compositions determined at the optimum excess air ratios, including CO and CO_2, SO_x, and NO_x are presented in Table 1.

Table 1

Exhaust gas composition for South Pars case study at the optimum excess air condition.

Intake sour compositions		$\mathbf{CO + CO_2}$ (%)	$\mathbf{SO_2}$ (%)	$\mathbf{SO_3}$ (%)	**NO** (%)	$\mathbf{NO_2}$ (%)
H_2S = 2.5%	CO_2 = 7.5%	13.9%	1.5%	0.3%	1.1%	0.1%
H_2S = 4.5%	CO_2 = 5.5%	13.7%	1.7%	0.4%	1.1%	0.1%
H_2S = 6.5%	CO_2 = 3.5%	13.5%	1.9%	0.5%	1.1%	0.1%
H_2S = 8.5%	CO_2 = 1.5%	13.3%	2.2%	0.6%	1.1%	0.1%

The calculated values of the GHG ($CO + CO_2$) compositions within the flue gas are almost constant. Only some small increases are seen in GHG emissions mainly due to the increment in CO_2 contents in the inlet feed. Conversely, SO_2 and SO_3 contents within the exhaust gas show unavoidable continuous rise with an H_2S increase in the inlet feed. It is noticeable that the calculated optimum excess air within each case study was different from the others, which shows that one optimum air fuel ratio (AFR) value cannot be adapted to all sour flares. As expected, the amounts of the NO and NO_2 in the flue gas show no change with any variation in H_2S to CO_2 proportions. We then conducted several sensitivity analyses on the intake air temperature. Air temperature can extremely be lowered via a humidification process using cheap air coolers, in which air goes through some adiabatic water saturation hypothetical lines. Low temperature oxidant mixtures, particularly with up to 80% inert nitrogen, play an important role in lowering the adiabatic flame temperatures, which consequently decrease NO_x productions. Table 2 tabulates the effects of the intake air temperature on each contaminant composition within the exhaust gas in the South Pars sour flare case study.

Table 2

Effects of intake air temperature on GHG and pollutant emission in the South PARS case study.

Intake temperatures		**Intake temperature 90 °F**	**Intake temperature 80 °F**	**Intake temperature 70 °F**	**Intake temperature 60 °F**	**Intake temperature 50 °F**	**Intake temperature 40 °F**
Air contaminant	NO %	1.4%	1.1%	0.9%	0.7%	0.6%	0.5%
	NO_2 %	0.2%	0.1%	0.07%	0.05%	0.04%	0.03%
	(SO_2+SO_3)%	2.4%	2.4%	2.4%	2.4%	2.4%	2.4%
	$(CO+CO_2)$%	13.5%	13.5%	13.5%	13.5%	13.5%	13.5%

The impact of the intake air temperature on the endothermic formation of NO and NO_2 shows that with a 50 °F decrease in the intake oxidant temperature, one can expect NO emission to increase threefold. The same trend appears in NO_2 emission with the difference that the effect is even larger. The production of the SO_x compound was not influenced by the intake air temperature perhaps due to the fact that SO_x formation reaction is only slightly exothermic. Also, GHG emissions were not affected with the intake air temperature and were only sensitive to the amount of excess air.

Moreover, it is noticeable that the inlet fuel temperature influences the flame temperature, but its effect is negligible. Therefore, one can offer to cool the air in sour gas flares as well as sweet flares using some throttling valves or air humidifiers prior to the premix entrance. Thereafter, we investigated the role of excess air in determining the adiabatic flame temperature and its effect on pollutant emission from sour flares. Table 3 illustrates the calculated results of flame temperatures and SO_x and NO_x emissions from two flare case studies.

Table 3

Effects of excess air on flame temperature and pollutant emission in AMAK case study.

	Excess air	**10.0%**	**15.0%**	**20.0%**	**25.0%**	**30.0%**
South Pars	Adiabatic flame temperature	2750	2830	2800	2780	2760
	Air pollutant (SOx +NOx)	3.8%	3.7%	3.6%	3.8%	4.0%
Ahwaz (AMAK)	Adiabatic flame temperature	3050	3100	3080	3050	3030
	Air pollutant (SOx +NOx)	3.1%	3.0%	2.9%	2.8%	3.1%

While theory states that exact stoichiometric oxidant ratios maximize the flame temperatures, the experiences show that some excess air is always needed to achieve this goal. Our calculated flame temperatures versus the excess air show increases in exhaust gas temperatures when excess air quantities change from 0.0% to 15.0%. This value appears to be the critical excess air above which the adiabatic flame temperatures decrease with a further increase in excess air. Similarly, in the two case studies of South Pars and AMAK (Ahwaz), 15% excess air maximizes the flame temperatures to values of 2830 °F and 3100 °F respectively. In lower oxidant quantities, the incomplete combustion of the fuel components occurs to some extent, which causes the flame temperature to drop. On the other hand, in higher excess air quantities, more energy consuming components appear in the exhaust, which lowers the adiabatic flame temperature. The harmfulness of each air contaminant, including NO, NO_2, SO_2, and SO_3 was weighed according to EPA environmental reports and was used in calculating the optimum excess air conditions. The oxidation of N_2 to NO_x is extremely endothermic, so it shows high yields at high flame temperatures and falls by increasing excess air quantities. On the contrary, the presence of the high amounts of O_2 within the reaction causes more SO_3 to be produced instead of SO_2. The EPA reports that the impact of SO_3 is several times more destructive than that of SO_2 in acidic rains. It goes through a slow equilibrium with water vapor present in the atmosphere and forms H_2SO_4, which is known as a highly corrosive compound within acidic rains. However, in an opposite way, SO_2 participates in the formation of H_2SO_3, which is several times weaker than H_2SO_4. We calculated optimum excess air quantities with respect to these effects reported by EPA (United States Environmental Protection Agency). In the South Pars case study, a 20% excess air with a tolerance of 5% was calculated as the optimum, while in the case of AMAK (Ahwaz) a value of 25% with a tolerance of 5% was determined as the optimum excess air to minimize the pollutants emission.

In order to verify data resulted from the written MATLAB code, we used FRNC 2012 software to simulate flaring processes in the two previously selected field case studies, namely AMAK (Ahwaz) and South Pars. Inlet gas compositions and oxidant mixture conditions were defined in FRNC 2012, and the options for incomplete reaction, i.e. NO_x and SO_x formation, were activated; then, the simulation was performed to give the resultant flue gas compositions. In each case study, several simulation runs at various air temperatures and excess air ratios were performed to find the conditions which minimize pollutant emission. In addition, we compared the results with the experimental data acquired from the gas chromatography (GC) tests of the exhaust gases in the two selected case studies. A comparison among the calculated composition of air pollutants and GHG's, the simulated results, and the experimental data is tabulated in Table 4.

Table 4

Validation of the written code results with FRNC Industrial Software and experimental data.

Flue gas compositions		$CO+CO_2$ (%)	SO_2 (%)	SO_3 (%)	NO (%)	NO_2 (%)
South Pars	via Code	13.50%	1.90%	0.50%	1.10%	0.10%
	via FRNC	13.80%	1.80%	0.60%	1.00%	0.20%
	Experimental data	13.95%	1.77%	0.53%	1.15%	0.15%
Asmari (AMAK)	via Code	15.40%	1.30%	0.30%	0.80%	0.10%
	via FRNC	15.60%	1.20%	0.30%	0.70%	0.10%
	Experimental data	15.73%	1.15%	0.28%	0.65%	0.15%

Conclusions

Several key findings of the present work can be summarized as follows:

1- An increase in H_2S content of the flare entrance by 2.0%, 4.0%, and 6.0% causes SO_x emission to raise 0.3%, 0.6%, and 0.9% respectively.

2- Low H_2S contents in sour gases do not significantly affect CO and CO_2 emissions. Hydrocarbons consume much larger volumes of oxygen in a way that greenhouse gas emissions are not affected with the presence of hydrogen sulfide.

3- A decrease in the temperature of the entrance air significantly reduces the NO_x formation. A drop of 20 °F and 50 °F in the oxidant temperature causes NO_x emission to respectively fall by 0.5% to 1%.

4- Pollutant emissions in sour flares are not necessarily minimized at optimum AFR's by flame temperature maximization. The independent optimum AFR's were calculated regarding the harmfulness of air contaminants.

5- The optimum AFR values to minimize pollutant emission in Ahwaz AMAK and South Pars case studies are calculated 25% and 20% respectively.

6- As EPA annually reports different indexes for the local harmfulness of each GHG and pollutant, the optimum flaring conditions vary locally and with respect to time.

7- Each sour flare seems to have a particular optimum excess air ratio which depends on H_2S content and other flow characteristics. One can expect the optimum condition to differ per case.

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Nomenclature

AFR	Air fuel ratio
EPA	Environmental Protection Agency
FRNC	Heater simulation program
GC	Gas chromatography
GHG	Greenhouse gas

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