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Optimisation of Syngas Composition for Emissions Minimisation and Cost Reduction through Selective Catalytic Reduction (SCR) and Gas Mixtures in a Gas Turbine

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Optimisation of Syngas Composition for Emissions Minimisation and Cost Reduction through Selective Catalytic Reduction (SCR) and Gas Mixtures in a Gas Turbine

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Abstract. The fuel flexibility of a gas turbine allows it to operate on a large variety of fuels, with syngas gaining prominence in recent years due to its versatility to be produced from any hydrocarbon-based feedstock. The feedstock versatility also meant that the syngas produced could be hydrogen-rich or carbon monoxide-rich in its composition. In this experimental study, the emissions characteristics of syngas combustion in a gas turbine, and their associated cost to minimise emissions using the selective catalytic reduction (SCR) method and gas mixtures were investigated. The syngas composition comprised of four gases, namely H_2, CO, CO_2 and CH_4. The syngas mixture is combusted under an equivalence ratio (ER) range of 0.4-0.9. Using Design of Experiments (DOE) optimisation procedures for simultaneous NO_x-CO reduction, the emission indices of the NO_x and CO pollutants for the best H_2-rich syngas (ER=0.5) were found to be 0.0189 g/kWh and 0.0028 g/kWh lower than that of the optimum CO-rich syngas (ER=0.5), respectively. This implies that H_2-rich syngas has greater potential for emissions reduction. However, the general combined costs for an optimum H_2-rich syngas mixture with the SCR post-treatment method is about six folds greater than the CO-rich syngas counterparts. The actual cost is even greater on a cost per mass reduction basis, with the NO_x and CO emissions being 21.6 and 8.9 times more for the H_2-rich mixture. It was also determined that the combined use of SCR method and optimum gas mixture is a more cost-effective emissions control measure than purely using direct gas mixture.

1. Introduction

Synthesis gas, more commonly known as syngas is being regarded as one of the promising alternative fuels for future energy demand due to its cleaner fuel characteristics. Syngas is comprised of gases, primarily hydrogen (H_2) and carbon monoxide (CO), with other gases like methane (CH_4) and carbon dioxide (CO_2) occasionally included in the mixture as well. The cleaner nature of syngas combustion in gas turbines meant that greenhouse gas pollutions can be reduced [1]. The most common method of syngas production is through the gasification process. Syngas composition can be varied widely due to the variety in the feedstock sources and the gasification process [2-3]. Therefore, the main challenge of using syngas as the fuel for gas turbine combustion will be the variability of the syngas compositions, in addition to the complex characteristics of swirl flames involved, as it can significantly affect the combustion behaviours. Therefore, stringent control of the combustion protocol is required for emissions abatement, to mitigate the formation of carbon-rich by-products.

The effects of syngas mixture components on combustion characteristics and emission performances have been studied. The flame appearances and the emission characteristics in a gas turbine fuelled with $H_2/CO/CO_2/CH_4$ syngas blends in an equivalence ratio of 0.4 to 1.1 [4]. Syngas with higher fractions of H_2 exhibited lower NO_x emission per kWh in the range of equivalence ratio investigated. The study also showed that the CO emissions were more affected by equivalence ratio as compared to H_2 fraction in the syngas. The addition of CO_2 diluent reduced the combustion temperature, leading to a reduction in nitrous oxide emissions. Numerical simulations studies have been conducted on the effects of syngas mixture compositions and the gas turbine The current state-of-the-art syngas fuel composition was simulated using Large Eddy Simulation (LES) to investigate the effects of syngas composition on the combustion characteristics [5]. Flame characteristics of swirled non-premixed H_2/CO syngas fuel mixtures have been studied. The simulation allows comparison of syngases with varied compositions and their associated flame behaviours.

This present study is conducted to provide a detailed analysis on the emission characteristics and their associated cost, aimed at minimising pollutant emissions in conjunction with operation cost reduction, using selective catalytic reduction (SCR) and gas mixtures. The current study also recommended the operating parameters for the desired outcome, based on a varying range of equivalence ratio, to replicate H_2-rich or CO-rich operating conditions, and address the lack of knowledge about the optimal configuration of H_2/CO syngas fuel mixture for achieving cost-effectiveness and emission reductions.

2. Methodology

2.1. Experimental setup

A single annulus swirl flame burner was utilised for the establishment of a continuous syngas swirling flame. The schematic of the experimental setup and the flow deliver system is shown in figure 1. The axial swirler consists of six straight vanes with thickness of 1.5 mm which were positioned at an angle of 45° to the axial centreline. The swirler was connected concentrically to the burner wall, which consists of a quartz tube with a length of 400 mm and diameter of 120 mm. The estimated swirl number is S_N ~ 0.84, which is considered strong for generation of high intensity flow recirculation for flame stabilisation [6].

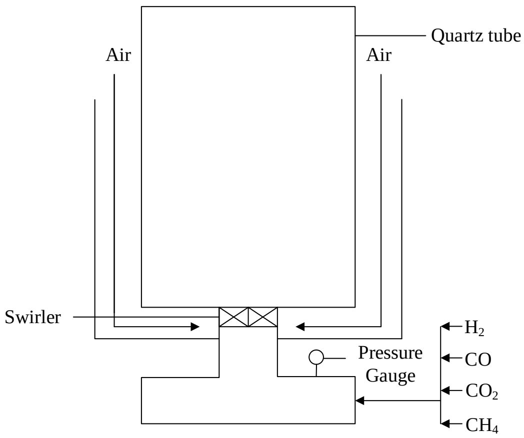

Figure 1. Schematic diagram of swirl burner with flow delivery system.

The combustion characteristics of H_2-rich and CO-rich syngases at different ratios of H_2/CO, namely H_2/CO=3, H_2/CO=1.2, CO/H_2=1.2, CO/H_2=3 are investigated. The effect of CO_2 in the syngas mixture is also investigated by incrementally adding 5% of CO_2 for every blend. The gases were first premixed in a mixing chamber, then delivered to a plenum and mixed with dry air. The sampling probe was placed at the burner outlet across eight equally-spaced spatial locations to perform measurements of post combustion emissions. Detailed specifications of the equipment used are shown in table 1.

Table 1. Specification of the temperature sensors and gas analyser.

Sensors	Range	Resolution	Accuracy	Propagated error
Temperature	0-800 ˚C	±1 ˚C	±0.3%, ±1 ˚C	±1.3%
NO	0-1000 ppm	±1 ppm	<100ppm; ±5 ppm >100ppm; ±5%	±22.3%
CO	0-10,000 ppm	±1 ppm	<100ppm; ±5 ppm >100ppm; ±5%	±1.3%

2.2. Statistical analysis

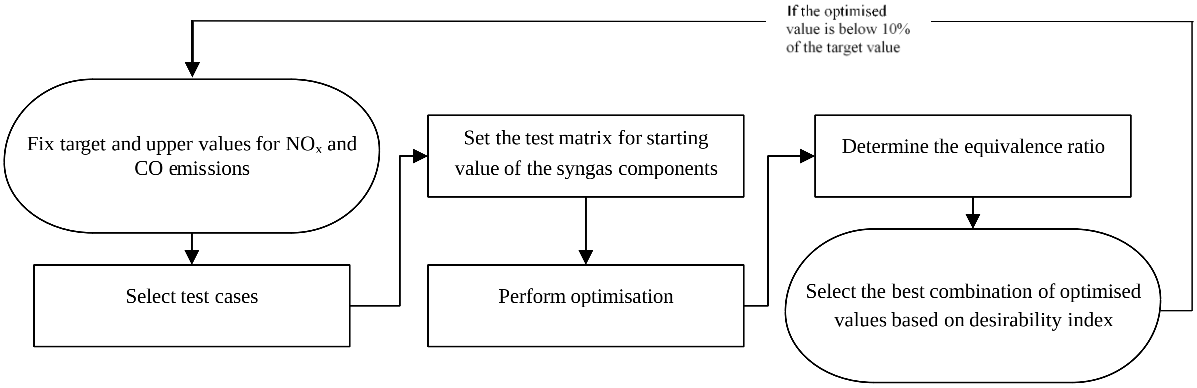

Figure 2. Methodology for the optimisations.

In this study, the Minitab 18 statistical software was used to perform statistical analyses. The custom mixture design was chosen and the expression for the sum of the syngas mixture is as shown below:

$$H_2 + CO + CO_2 + CH_4 = 1.0 \quad (1)$$

The mixture design utilises four mixture variables namely, H_2, CO, CO_2 and CH_4 and one process variable which is equivalence ratio (∅). Figure 2 shows the methodology used in optimisation process. The 'target' values for the optimisation are fixed as the lowest value obtained from the set of experimental data. However, the values from pure H_2 and pure CO syngases were not considered. The 'upper' value was defined as +20% of the 'target' value. The ranges of the starting values of each syngas component is shown in table 2. The starting value for equivalence ratio in the range of ER0.4-0.9 was increased by a fixed increment of 0.1. No response variable is being prioritised as the importance was set at 1.0.

Table 2. The ranges of optimisation for each component.

Type of Syngas Investigated	H_2 (vol/100)	CO (vol/100)	CO_2 (vol/100)	CH_4 (vol/100)
H_2-rich	0.5-0.8	0.1-0.4	0.05	0.05
	0.4-0.6	0.1-0.3	0.25	0.05
CO-rich	0.1-0.4	0.5-0.8	0.05	0.05
	0.1-0.3	0.4-0.6	0.25	0.05

2.3. Analysis of costs

The costs of industrial fuel gases of cylinder tank are adopted from NorLab, the Specialty Gas Division of Norco, Inc [7]. The cost of reducing NO_x using SCR technology is obtained and investigated [8]. The cost considered for the SCR technology are capital, performance loss, catalyst, O&M, ammonia and

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catalyst disposal. The cost value of each term is recalculated to fit the output of the model. All cost values are calculated with the correlation value of R_2 higher than 0.9.

3. Results and Discussion

3.1. Optimisation for NO_x and CO emissions

Figure 3 shows the optimisation desirability plot of the two common gas turbine pollutant gases, which are NO_x and CO emissions for the minimisation process for pollutant emissions. The belt regions of low diluent denote the mixtures covered by the experimental data, while the regions outside are extrapolated desirability. Two regions in the contour plot show relatively higher desirabilities, where the H_2 or CO content exceeds 80% by volume within the mixtures which give maximum desirabilities of 0.75-0.85 and 0.25-0.35, respectively. A very high H_2-content syngas and a very high CO-content syngas are obtained as the optimised syngas mixture composition. Low formation of NO is due to the lack of C radicals for H_2-rich syngas, likewise the lack of H radicals for high CO-content syngas. The lack of C radicals prohibit the production of CH and CH_2 that are critical for the formation of the intermediate species, HCN in the formation of prompt NO. The lack of H radical, however leads to slower oxidation rate of HCN. The reactions involved in the oxidation of HCN are shown as below [9]:

$$HNCO + H \longleftrightarrow NH_2 + CO \quad (2)$$

$$NH_2 + H_2 \longleftrightarrow NH_3 + H \quad (3)$$

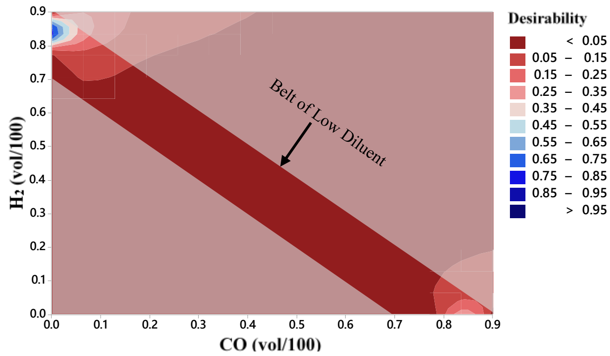

Figure 3. Optimisation desirability for NO_x and CO emissions against H_2 and CO in the range of ER 0.4 and ER 0.9.

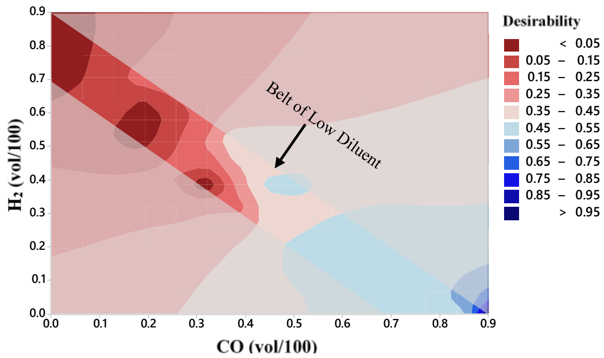

Figure 4. Optimisation desirability for the costs of SCR and fuel gas against H_2 and CO in the range of ER 0.4 and ER 0.9.

Table 3 summarises the optimum combinations of syngas mixtures and equivalence ratio that give the possible minimised values for NO_x and CO emissions using the optimisation method. The cases with H_2-rich and CO-rich syngases give the desirability index of 0.8728 and 0.8823, respectively. The values of fuel gas costs for H_2-rich syngas and CO-rich syngas are also included. Using Design of Experiments (DOE) optimisation for simultaneous NO_x-CO minimisation, the emission indices of NO_x and CO pollutants for the optimum H_2-rich syngas at equivalence ratio of 0.5 were found to be 0.0189 g/kWh and 0.0028 g/kWh lower than that of the best CO-rich syngas, respectively. The low optimised emission index value of CO for H_2-rich syngas case is attributed to the increase of OH radicals as H_2 content is high. The activation energy of reaction (4) involving OH radicals that converts CO to CO_2 are lower than other reactions (5)(6). The reactions of conversion of CO to CO_2 are [10]:

$$CO + OH \longleftrightarrow CO_2 + H \quad (4)$$

$$CO + O_2 \longleftrightarrow CO_2 + O \quad (5)$$

$$CO + O + M \longleftrightarrow CO_2 + M \quad (6)$$

The optimum values of equivalence ratio in the pollutant minimisation process for both cases are the same at ϕ=0.5. It is suggested that the low emissions of NO_x are due to the low formation of thermal NO. This is because the flame temperature for fuel-lean mixture is relatively low, resulting in slower oxidation rate of nitrogen. Meanwhile, the emission of CO is found to have optimised value at ϕ=0.5 as a large decrement always observed at very fuel-lean region.

Table 3. Optimised syngas composition of H_2, CO, CO_2, CH_4 for minimised NO_x and CO emissions.

Operational Condition		Optimised Process Variable [Mass-based]	Optimised Emission Indices [g/kWh]		Optimised Syngas Mixture Composition [vol/100]				Cost Associated [$/kWh]
		ER	NO_x	CO	H_2	CO	CO_2	CH_4	Fuel Gas
ER 0.4-0.9	H_2-rich	0.5	0.017	0.040	0.819	0.000	0.131	0.050	32.66
	CO-rich	0.5	0.035	0.043	0.105	0.795	0.050	0.050	6.38

3.2. Optimisation for costs

The desirability of SCR costs and fuel gas costs for the cost optimisation is shown in figure 4. A high CO-rich syngas is shown to give a higher desirability as compared to a high H_2-rich syngas. Table 4 also summarises the optimum combinations of syngas mixture composition and equivalence ratio in the minimisation for SCR costs and fuel gas costs. Both cases give the desirability index value of 1.0. The combined costs for an optimum H_2-rich syngas mixture with the SCR treatment is $22.97/kWh greater than that its CO-rich syngas counterpart. This is because the industrial H_2 fuel gas has relatively higher cost than that of CO fuel gas. The cost of SCR for H_2-rich syngas is $1.11/kWh, which is $0.08/kWh higher than that for CO-rich syngas. The slightly higher value of SCR cost for H_2-rich syngas may be attributed to the lower output power. The optimised values for equivalence ratio for both H_2-rich and CO-rich syngas are ER 0.9, which is the highest value in the range of equivalence ratio investigated. The combustion efficiency is high with higher equivalence ratio as a result of higher flame temperature, therefore leading to a lower power generation cost.

Table 4. Optimised syngas composition of H_2, CO, CO_2, CH_4 for minimised costs of SCR and fuel gas.

Operational Condition		Optimised Process Variable [Mass-based]	Optimised Costs [$/kWh]		Optimised Syngas Mixture Composition [vol/100]				Emission Indices [g/kWh]	
		ER	Fuel Gas	SCR	H_2	CO	CO_2	CH_4	NO_x	CO
ER 0.4-0.9	H_2-rich	0.9	25.92	1.11	0.496	0.404	0.050	0.050	0.028	0.053
	CO-rich	0.9	3.03	1.03	0.036	0.864	0.050	0.050	0.092	0.071

3.3. Recommendation

Using optimisation method for simultaneous NO_x-CO reduction, the emissions indices of NO_x and CO for the H_2-rich syngas were lower than that of CO-rich syngas. This indicates that H_2-rich syngas has greater potential in reducing the pollutants. However, the combined costs for H_2-rich syngas with the SCR post-treatment is six times as high when compared with that of CO-rich syngas equivalent. On a cost per mass of emissions reduction basis, the cost escalates to 21.6 and 8.9 times more for NO_x and CO output, respectively. For H_2-rich syngas, the SCR post-treatment method shows a total combined cost of $27.03/kWh, while the cost is $32.66/kWh for the method of using gas mixture. This implies that the combined use of SCR treatment and optimum gas mixture for the purpose of minimising pollutant emissions is cheaper than just that purely using direct gas mixture.

4. Conclusion
Using Design of Experiments (DOE) methodology, optimisations were performed on a four-component $H_2/CO/CO_2/CH_4$ syngas mixture, in order to achieve simultaneous NO_x-CO reduction and costs reduction. Optimum syngas mixture compositions paired with the equivalence ratios were proposed in the range of equivalence ratio from 0.4 to 0.9, which is considered as the fuel-lean combustion regime. In the optimisation for both pollutants, the emission indices of the NO_x and CO pollutants for the optimum H_2-rich syngas at equivalence ratio of 0.5 are found to be 0.0189 g/kWh and 0.0028 g/kWh lower than that of the optimum CO-rich syngas case, respectively. The result shows that H_2-rich syngas has higher potential for the emissions reductions. However, the general combined costs for an optimum H_2-rich syngas with SCR treatment method is about six folds higher than that of CO-rich syngas. The result also shows that the combined use of SCR and optimum gas mixture is more cost-effectiveness than purely varying the syngas mixture composition. The statistical approach presented in this study provides an effective way to perform analysis on syngas mixture effects. The optimised syngas mixture composition can be obtained in the process to achieve desired outcomes.

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