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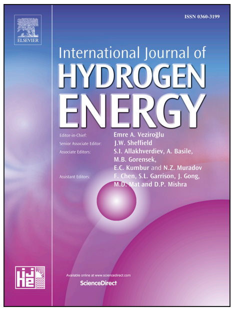

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H_2-Rich Syngas Strategy to Reduce NO_x and CO Emissions and Improve Stability Limits under Premixed Swirl Combustion Mode

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Abstract

The combustion performance of H_2-rich model syngas was investigated by using a premixed swirl flame combustor. Syngas consisting mainly of H_2 and CO was blended with components such as CH_4 and CO_2 in a mixing chamber prior to combustion at atmospheric condition. The global flame appearance and emissions performance were examined for high (H_2/CO = 3) and moderate (H_2/CO = 1.2) H_2-rich syngases. Results showed that higher H_2 fractions in the syngases produce lower NO_x emissions per kWh basis across all equivalence ratios tested. CO emissions are equivalence ratio dependent and are less affected by the H_2 fraction in the syngas. Increasing CO_2 diluent ratios result in the decrease of NO_x, particularly for moderate H_2-rich syngases. In contrast, syngas without CO shows an increase of NO_x with increasing CO_2 for fuel-lean mixtures. Addition of CO_2 increases the lean blowout limit of all syngases. Higher fraction of H_2 produces lower lean blowout limits due to the characteristics of high diffusivity of hydrogen molecules and high flame speed that assist in the stabilisation of the flame under flame-lean conditions. The range of blowout limits for moderate and high H_2-rich and pure hydrogen syngases under diluent ratios up to 25% were within the range of ϕ = 0.12-0.15.

Keywords: syngas, lean blowout, NO_x, swirl, emissions, CO_2 diluent

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1. Introduction

Synthesis gas, also known as syngas, produced from gasification processes is regarded as one of the promising alternative energies due to its clean fuel characteristics. The main components of syngas are H_2 and CO, which can be produced via gasification of various feedstock, including coal, biomass and solid waste [1]. The use of syngas as fuel source can potentially reduce CO_2, NO_x and other pollutants [2]. One example of syngas application is in Integrated Gasification Combined Cycle (IGCC) power plants where syngas fuel is combusted in gas turbines to generate power and electricity [3, 4]. Despite the proven feasibility of syngas, the challenges that syngases face are the variation in their composition due to different feedstock and production methods. Furthermore, the lack of understanding of the combustion characteristics of syngases poses difficulty in the design of syngas-specific systems and combustors [5].

Most gasification processes typically produce syngases that are CO-rich or H_2-rich depending on feedstock. CO-rich syngas has been produced by coal gasification with blends comprising 60% CO and 30% H_2 by volume [6]. The relative molar fraction of H_2 to CO for coal-derived syngas ranges from 0.4 to 1.0 [7]. The use of catalytic gasification technique to gasify biomass was shown to produce H_2-rich syngas with a composition of up to 50% H_2 and 17% CO by volume [8]. The volume ratio of H_2/CO in most syngas mixtures typically exceeds 0.25, where chemical kinetic and reaction mechanisms of hydrogen play a dominant role in syngas combustion. Hence, syngas generally exhibits large burning rates with small autoignition time, comparable to those of pure hydrogen combustion [9].

CO-rich syngas combustion shows combustion characteristics differ from those of H_2-rich syngas. Low concentration of H atoms in the former affects fast oxidation pathways of CO, resulting in unstable combustion and high CO emissions that are not well understood [10]. Previous studies of syngas combustion are restricted to limited composition of mixtures. Zhang *et al.* [11] studied the effect of dilution of N_2 and CO_2 on the propagation and extinction of lean premixed syngas flames using an opposed-jet twin flame configuration. CO-rich syngas with 50-90% CO and 8-50% H_2 were used in the study. Results showed that the dilution effect of CO_2 was more profound on flame propagation and extinction than that of N_2. Syngas emissions were not measured. Joo *et al.* [12] investigated the emissions of H_2-rich syngas in a partially premixed model gas turbine combustor. Syngas compositions of 25-50% H_2 and 25-100% CH_4 by mol were used as test fuels with the presence of CO ranging from 0-25% by mol. NO_x emissions index showed proportional trends to the flame temperature within the equivalence ratio range of $\phi = 0.7$-1.3 under fixed heat inputs.

The effects of hydrogen composition in syngas on emissions performance have been studied by several groups. Azimov *et al.* [13] used syngas derived from the gasification of biomass and oven coke in a diesel-type dual-fuel engine. The use of higher H_2 concentration syngas with H_2/CO ratio of 20/22.3 compared to syngas with ratio of 13.7/22.3 resulted in reduced CO and HC emissions but an increase in emitted NO_x. The baseline of pure H_2 fuel without dilution produced low emissions at equivalence ratios $\phi \leq 0.3$. Diluting pure H_2 with N_2 at $\phi = 0.35$ resulted in a decrease of NO_x emissions but an increase in CO emissions. Similar trend of higher NO_x emissions for H_2-rich syngas was found by Lee *et al.* [14] in a 60 kW industrial gas turbine using pure syngas without diluent. NO_x emissions were reported to increase exponentially as the heat input increased. When operating with H_2-rich syngas at higher equivalence ratios, an increase in flame temperature and higher NO_x emissions were reported. Higher CO was emitted with lower combustion efficiency when the gas turbine was operated at low load or idle mode. However, Ouimette *et al.* [15] reported a different NO_x emissions trend for syngas under partially premixed combustion mode. Syngas mixtures with H_2/CO ratio of 0.8-1.3 produced rather constant NO_x emissions, but NO_x showed a decreasing trend at higher H_2/CO ratios (> 1.3).

Numerical studies on the extinction limits and emissions for various syngas compositions have been conducted by Ding *et al.* [16]. Results showed that syngases with higher H_2 content exhibit higher flame temperature profiles and wider flame front. The high H_2 content in syngas resulted in high diffusion rate of hydrogen into the reaction zone of the flame compared to CO, thus accelerating the combustion rate. Huynh *et al.* [17] investigated the NO_x emissions of syngases produced from pine, maple-oak mixture and seed corn via gasification process. The syngases produced contained a maximum of 16% H_2, 20% CO, 20% CO_2, 40% N_2 and 55% H_2O. Syngas blends were combusted in an 879 kW industrial burner that operates under the principles of non-premixed combustion with staged air supply. Combustion of the syngas derived from seed corn resulted in NO_x emissions in the range of 450-990 ppm, which is significantly higher than those of wood-derived syngas due to the high nitrogen content in seed corn. Watson *et al.* [18] compared the emissions of methane with a syngas consisting of 37.5% H_2, 37.5% CO, 5% CH_4 and 20% CO_2 by volume and syngas-biogas blends with a composition of 18.8% H_2, 18.8% CO, 52.5% CH_4 and 10% CO_2 by volume using a jet-wall stagnation flame configuration. Result showed that the syngas and biogas mixture produced lower NO emissions than methane when the fuels were compared at equal power outputs.

The inherent variability in composition and heating value in synthesis gases cause significant challenges towards their usage in practical combustion system, especially for low emission combustors that operate near the blowout limit. Blowout occurs when the flame becomes detached from the anchored location and is “blown out” of the combustor. For syngas, the lean blowout limit (LBO) varies significantly depending on the fuel composition and chemical kinetic rates. Several groups have investigated the blowout phenomena of syngas at lean burning region [19, 20]. Li *et. al.* [21] studied the effects of diluents on the LBO limits of premixed syngas flames by using an atmospheric swirl flow combustor. Results showed that the LBO limits increase with the dilution ratio of N_2, and that the inert dilution dominates the LBO behaviour of syngas with low H_2 content. The effect of swirl intensity on the flame stability limit of a premixed variable swirl burner operated with syngas was investigated by Sayad *et al.* [22]. It was shown that LBO limit was not significantly affected by the change of swirl number, concurring with the result shown by Garcia-Armingol and Ballester [23]. The latter reported that the LBO limit of a turbulent premixed swirl burner ranges between ϕ=0.4-0.7 for syngases with 0-50% H_2 by volume. The stability limits for high H_2-enriched syngas was not investigated.

Syngas produced from most gasification processes contains different H_2, CO and diluent compositions. CO and H_2 constitute the main reactive components in syngas ranging from 10% to 70% by volume [8, 24-26]. Diluents such as CO_2, N_2 and H_2O are typical non-reactive components present in syngases derived from biomass or coal gasification. Due to a wide range of possible syngas composition produced from different feedstock and processes, thorough investigation of the combustion performance for any syngas is needed to allow further practical application and design of syngas-specific combustors. On the previous note, there is a lack of studies on the combustion characteristics of H_2-rich syngases. The present study focuses on the combustion performance of H_2-rich syngases under a continuous swirl flame mode. The flame appearance, post-flame emissions and lean blowout limits are investigated and compared against baseline pure hydrogen/air flames.

2. Experimental

2.1 Swirl burner and flow delivery system

An axial swirl flame burner was utilised to establish continuous swirling syngas flames. The schematic of the swirl flame burner and flow delivery system is shown in Fig. 1a. The axial swirler (Fig. 1b) consists of 6 straight vanes with thickness of 1.5 mm. The swirler vanes are positioned at an angle of 45^o from the axial centerline. The outer diameter of the swirler is 40 mm, while the swirler hub diameter is 20 mm. The swirler was placed at the burner outlet, concentric to the burner wall which consists of a quartz tube with a diameter of 120 mm. The use of a quartz tube allows optical access and enables visualisation of the flames. The geometric swirl number is approximate to be $S_N \sim 0.84$ based on the equation,

$$S_N = \frac{2}{3}\left[\frac{1-\left(D_h/D_s\right)^3}{1-\left(D_h/D_s\right)^2}\right]\tan\theta \qquad (1)$$

where D_h and D_s represent the swirler hub diameter and the swirler diameter, respectively, and θ is the angle of the swirl blade from the centreline. The present swirler was able to generate sufficiently high swirl ($S_N > 0.6$) to assist in flame stability [27]. For the flow delivery, hydrogen (99.99% purity) and carbon monoxide (99.97% purity) gases were supplied and regulated via mass flow controllers (Sierra) with an accuracy of ±1.5% full scale. The supplies of CH_4 (99.95% purity) and CO_2 (99.9% purity) were regulated using flow meters (Key Instrument) with an accuracy of ±5% respectively. The gases were premixed at room temperature of 298 K in a mixing chamber to ensure thorough mixing. The mixed gases then premixed with dried air at the burner plenum. The air was supplied and regulated using a mass flow controller (Sierra) with an accuracy of ± 1.5% full scale. The uncertainties of the flow meters for inlet gases (air + fuel) are 1.5-5%. A flame torch was used to ignite the combustible mixture at the burner outlet exposed to the atmosphere.

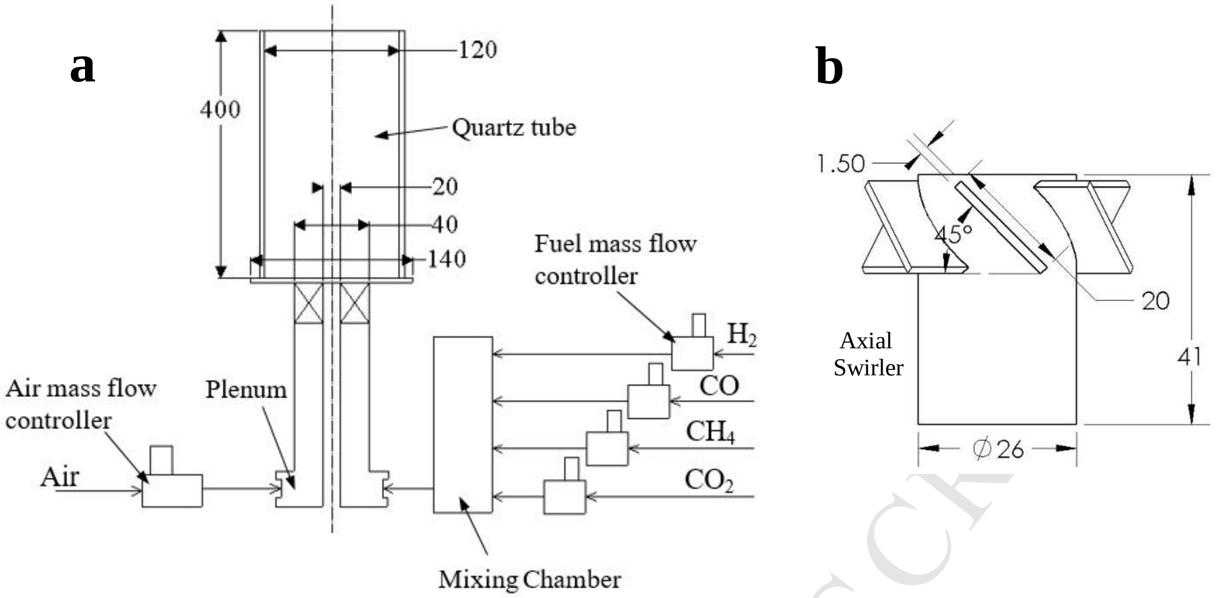

Figure 1: Schematic diagram of (a) swirl burner with flow delivery system and (b) axial swirler. Dimensions are in mm.

2.2 Operating conditions

In the present study, H_2-rich syngases blended with CO_2 and CH_4 as diluents are investigated. Table 1 shows all model syngases tested. For case S1-S5, the syngas H_2/CO ratio is fixed at 3, while case S6-S10 set the H_2/CO ratio to 1.2, representing high H_2-rich syngas for the former and moderate H_2-rich for the latter cases respectively. By fixing the main air flow rate at 2 g/s, the syngas fuel flow rates were regulated to form mixtures with equivalence ratio ranging from ϕ = 0.5 to 1.1, covering fuel-lean, stoichiometric and rich conditions. For the syngas baseline case B1-B5, the main air flow rate was fixed at 2.5 g/s. A constant 5% of CH_4 was added to all test cases to increase the calorific value of the mixture. For emissions measurements, cases S1 and S6 were studied and compared against baseline B1. S1 is a high H_2-rich syngas (67.5% H_2) while S6 is a moderate H_2-rich (49.5% H_2) case. The baseline established in the present study consists of 90% H_2, 5% CO_2 and 5% CH_4 by volume without CO. The effect of

CO_2 as diluent on the emissions is investigated by incrementally adding CO_2 at 5% for both sets of syngas, S1-S5 and S6-S10 and compared against baselines B1-B5.

Table 1: Syngas mixture composition tested (vol. %)

Mixture	**H_2 (vol. %)**	**CO (vol. %)**	**CO_2 (vol. %)**	**Lower heating value (MJ/kg)**
S1	67.5	22.5	5	25.0
S2	63.8	21.3	10	20.4
S3	60.0	20.0	15	17.0
S4	56.3	18.8	20	14.3
S5	52.5	17.5	25	12.2
S6	49.5	40.5	5	17.9
S7	46.8	38.3	10	15.5
S8	44.0	36.0	15	13.5
S9	41.3	33.8	20	11.8
S10	38.5	31.5	25	10.4
B1	90	0	5	53.3
B2	90	0	10	35.4
B3	90	0	15	25.8
B4	90	0	20	19.8
B5	90	0	25	15.8

* CH_4 is supplied as diluent at 5% by volume for all test cases
* S1-S5 are high H_2-rich syngases with H_2/CO ratio of 3
* S6-S10 are moderate H_2-rich syngases with H_2/CO ratio of 1.2
* B1-B5 are pure hydrogen syngases without CO

2.3 Emissions measurements

Measurements of post combustion emissions were performed by using a gas analyzer (Tempest 100) at the combustor outlet. The sampling probe was placed at the burner outlet to measure the emissions radially at 8 spatial locations that were equally spaced. The spatial emissions values were averaged and weighted by area-velocity to obtain the final global emissions value for each test case [28]. The emissions measured include NO, CO, O_2 and CO_2, with the measurement range of 0-1000 ppm, 0-10000 ppm, 0-25% and 0-99.9% respectively. The emissions readings from the gas analyser are reported in dry basis with the uncertainty of ± 5 ppm for NO, ± 5 ppm for CO, ± 0.2% for O_2. The NO, CO emissions were measured by chemical sensors in the gas analyser, while the CO_2 emissions was calculated based on the chemically measured O_2. Sampling of the post-combustion gases was performed on the steady state, continuous swirl flame for 1 minute until the reading became stabilised. The propagated errors of the emission measurements for NO_x, CO, O_2 and CO_2 are ± 22.3%, ± 1.3%, ± 1.3% and ± 4.2% respectively. Detailed specifications of the gas analyser and temperature sensors are summarised in Table 2.

Table 2: Specification of the gas analyser and temperature sensors.

Sensors	Range	Resolution	Accuracy	Propagated error
Temperature	0 – 800 °C	±1 °C	±0.3 %, ±1 °C	± 1.3 %
NO	0-1000 ppm	±1 ppm	< 100 ppm; ±5 ppm >100 ppm; ±5 %	± 22.3 %
CO	0-10000 ppm	±1 ppm	< 100 ppm; ± 5 ppm > 100 ppm; ± 5 %	± 1.3 %
O_2	0-25 %	0.1 %	±0.2%	± 1.3 %
CO_2*	0-99.9 %	0.1 %	N/A	± 4.2 %

*CO_2 is calculated based on the measured O_2

2.4 CO_2 diluent addition in syngases

The effect of CO_2 diluent on H_2-rich syngases was investigated by varying the CO_2 diluents while keeping the ratio of H_2/CO fixed and CH_4 constant at 5%. The diluent ratio $X_{diluent}$ is defined by, $X_{diluent}=V_{diluent}/(V_{diluent}+V_{fuel})$, where $V_{diluent}$ and V_{fuel} are the volume fractions of diluents and fuel (syngas) respectively in the reactant mixtures [29]. Dilution of CO_2 was performed at 10-25% for high (S2-S5) and moderate H_2-rich syngases (S7-S10). The emissions and lean blowout limits are compared to baselines (B2-B5) with corresponding diluent percentage.

2.5 Blowout limit of syngases

The blowout phenomena, often regarded as the upper static stability limit of combustors, occurs when the flame detaches from its anchored location and physically blows off [30]. Measurements of the high and moderate H_2-rich syngases were performed in the present study to determine the blowout limit under non-diluted and CO_2-diluted conditions. By gradually increasing the air flow rate of an established flame until the flame blows out, the equivalence ratio at which blowout occurs was recorded as the lean blowout limit. Comparison of the blowout limit of H_2-enriched syngases against the baseline syngas was performed.

3. Results and discussion

3.1 Flame imaging

A digital camera (CASIO; model EX-ZR1000) was utilised to image the global flame appearance of syngas swirl flame established at different equivalence ratios through the optically accessible quartz wall. The focal length and exposure time of the camera were set at 4 mm and

1/15 second, respectively. Figure 2 shows two sets of syngas flame images, case S1 and S6, established at ϕ = 0.5-1.1 with a constant composition of 5% CO_2 and 5% CH_4. Despite the differences in H_2/CO ratios, syngas flames exhibit rather similar attributes. For fuel-lean mixtures of ϕ = 0.5-0.7, bluish flames were observed as a result of the intense heat reaction zones. The fuels chemically react in this region under sufficient oxygen, leaving no unburned hydrocarbon for post-flame reaction or soot formation. Increasing the equivalence ratio to near stoichiometric region results in the luminous orange-yellow region downstream of the main reaction zone. Visible orange-yellowish post flames are indicative of soot formation. The luminous post-flame region is evident for fuel-rich mixture, as shown in Fig. 2a and 2b for S1 and S6 respectively. The typical high temperature environment under stoichiometric or fuel-rich conditions increase the concentration of vapour consisting of combustion product of H_2 component, e.g. H_2O, and formation of C/H radical, paving the way for the production of acetylene and propargyl formation which are essential precursors for soot formation [31, 32].

It is observed that high H_2-rich syngas (S1) shows less luminous post-flame region compared to moderate H_2-rich syngas (S6) at the equivalence ratio ϕ = 1.1, indicating the lower tendency for soot formation for the former due to reduced availability of C radicals. The higher H_2 concentration in S1 results in the overall decrease in flame reaction zone length due to reduced flame thickness [33]. Hydrogen has the intrinsic characteristic of greater molecular diffusivity. Increase of H_2 concentration in fuel-air mixtures results in reducing the convective time scale for H_2 to the flame front [34]. As a result, the burning intensity is increased while the main reaction zone length is decreased, as evident across the lean and rich region for S1 flames. The increase of equivalence ratios results in the decrease of diffusive-thermal instabilities for S1 and S6 due to the increase of Lewis number, *Le* (ratio of heat diffusivity to mass diffusivity). Correspondingly, hydrodynamic instability increases with increasing equivalence ratio as a result of reduced flame thickness and increased thermal expansion ratio (ratio of unburned gas density to burned gas density) [33]. Li *et al.* [33] quantitatively showed that the effect of diffusive-thermal instability is more dominant than hydrodynamic instability for the syngases with hydrogen fractions of 50% and 70% in an outwardly propagating flame test. No evident signs of intrinsic instabilities were observed for the present S1 (67.5% H_2) and S6 (49.5% H_2) syngas flames, partly in due to the weak flame intrinsic instability effect on the smallest wrinkled scale, and the sufficiently strong turbulent flame where the effect of intrinsic instability is nullified by the turbulence vortex scale. The effect of preferential diffusion of H_2 in syngas is expected to affect the diffusive-thermal instability near the ultra-lean region ($\phi < 0.4$) of syngas where the intensity of the flame front wrinkling in turbulent flame is enhanced.

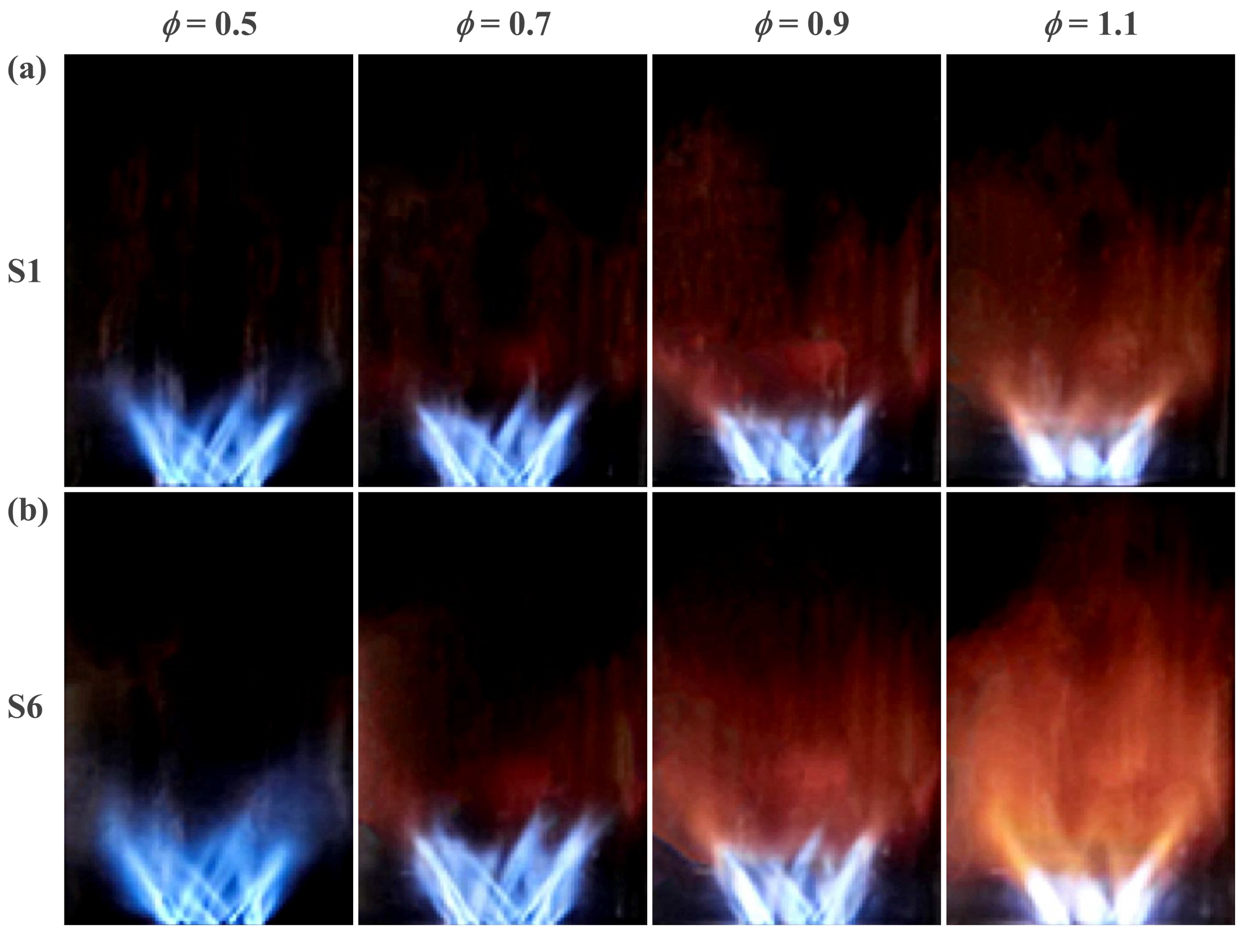

Figure 2: Global flame images of (a) high (S1) and (b) moderate (S6) H_2-rich syngas at different equivalence ratios.

3.2 Effect of equivalence ratio on syngas emissions

Comparison of the NO_x emissions indices for high H_2-rich syngas (case S1) and moderate H_2-rich syngas (case S6) against baseline (case B1) as a function of equivalence ratios is shown in Fig. 3a. The diluents of CH_4 and CO_2 for all the cases including baseline were both supplied at constant 5% by volume. For syngas S1, the trend is similar to baseline hydrogen fuel where fuel-lean region shows the lowest NO_x but increases exponentially as the fuel/air mixture approaches stoichiometric and rich regions. This is due to the increase of flame temperature as the mixture becomes richer towards stoichiometric and fuel-rich region that favours the formation of thermal NO_x [23, 35].

The NO_x emissions indices for moderately H_2-rich syngas S6 is comparatively higher than syngas S1 across the range of equivalence ratio (ER) tested. It also differs at the very lean range of $\phi = 0.4 – 0.6$ where NO_x emissions can be 2.5 to 3.3 times greater than that of the H_2-rich syngas S1. This can be attributed to the tri-effects of (i) the stack temperature which was found to be higher for S6 (467-517 °C) as compared to S1 (446-475 °C), (ii) the much lower power generated at the lower ER and (iii) the effect of CO on the NO_x emissions is significant for H_2-rich syngas. An increase of CO from 22.5% (S1) to 40.5% (S6) resulted in the average increase of NO_x by a factor of 2. This is partly in due to the presence of oxygen atom in CO, which further promotes the formation of NO_x via dissociations. The high H_2/CO ratio in S1 is another reason that retarded the formation of NO_x. The abundant H radical and lack of C radical prohibits the production of CH and CH_2 radicals that are critical for the formation of HCN, which is a precursor responsible for the formation of prompt NO_x emissions [15]. It is also observed that the ER range where NO_x tend to plateau before increasing tends to shift towards higher ER with decreasing H_2 proportion in the syngas.

The emissions index presented in log form for CO as a function of equivalence ratios is shown in Fig. 3b. Surprisingly the high H_2-rich syngas (S1) shows CO emissions close to that of moderate H_2-rich syngas (S6), except at $\phi = 0.4$ and stoichiometric region where S6 exhibits slightly higher CO emissions. The high production of CO at fuel-lean $\phi = 0.4$ is due the incomplete combustion of pockets of fuels and further aggravated by the presence of CO in the syngas fuels. The low flame temperature in fuel-lean region prohibits the conversion of CO into stable species of CO_2 [23]. An increase of equivalence ratio to $\phi = 0.5$ resulted in the reduction of CO formation by an order of magnitude. The combustion efficiency is increased with higher equivalence ratio as a result of higher flame temperature. Syngas S6 shows that $\phi = 0.9$ produces the lowest CO before increasing again at stoichiometric and rich conditions, whereas syngas S1 produces the least CO at $\phi = 1.0$. An increase of CO production in the fuel-rich region as shown by syngas S6 is attributed to incomplete combustion due to lack of oxygen. The baseline case B1 shows low CO emissions compared to H_2/CO syngases. At $\phi = 0.4$, the baseline shows CO emissions that is an order of magnitude lower than both syngas S1 and S6. High H_2-rich syngas shows NO_x emissions that are comparable to baseline but CO emissions is significantly higher in the lean region.

The emissions indices for CO_2 and O_2 for high (S1) and moderate (S6) H_2-rich syngases are shown in Fig. 4a. Both syngases show higher CO_2 emissions compared to baseline B1, in particular in the fuel-lean region of $\phi < 0.9$. Despite the significant differences in CO values in the fuel-lean region, CO_2 seems to converge in the stoichiometric region. The baseline hydrogen fuel shows significantly low CO_2 emissions, with approximately a factor of 3 lesser compared to case S1 and S6. The high CO_2 emissions for the latter in the fuel lean region were attributed to the presence of carbon monoxide in the fuels where part of the CO in the fuel was converted into CO_2 [36]. The O_2 emissions profiles were similar for cases S1, S6 and B1 as shown in Fig. 4b.

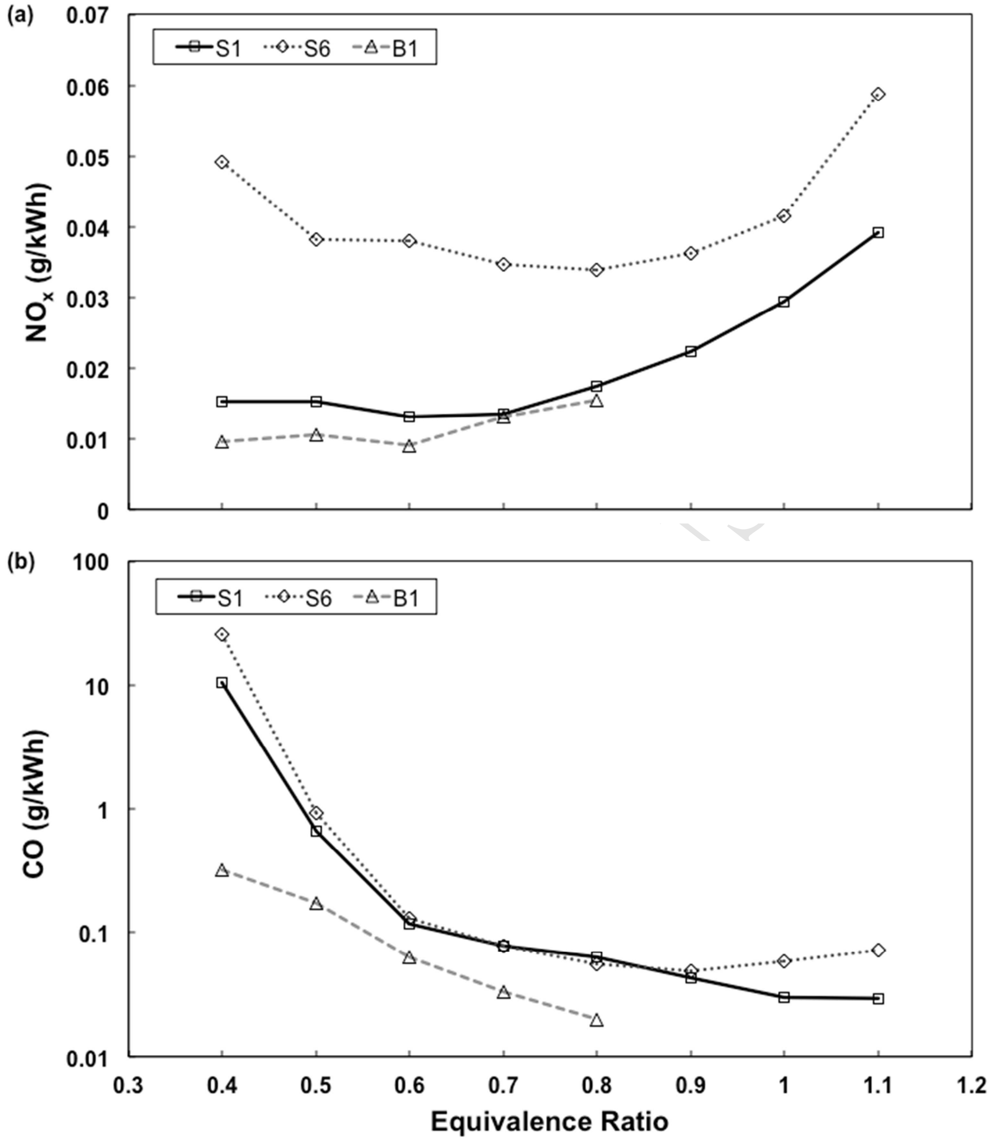

Figure 3: Emissions indices of (a) NO_x and (b) CO for high H_2-rich syngas (S1), moderate H_2-rich syngas (S6) and baseline pure hydrogen syngas (B1) as a function of equivalence ratio.

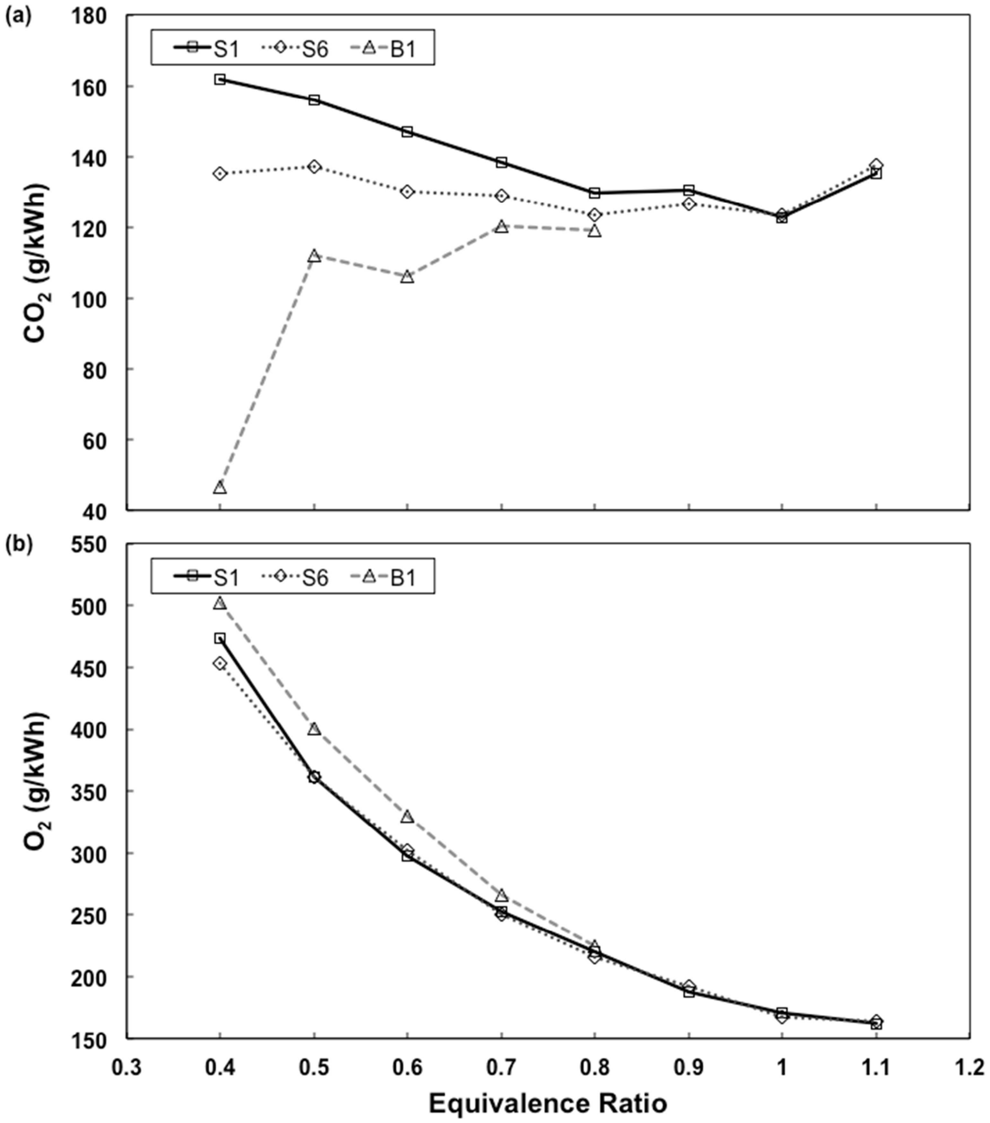

Figure 4: Emission indices of (a) CO$_2$ and (b) O$_2$ for syngas S1, S6 and baseline B1 as a function of equivalence ratio.

3.3 Effect of CO_2 diluent in syngases on emissions

The NO_x emissions index for moderate H_2-rich syngases (S6-S10) are shown in Figure 5a. Overall, at the range of equivalence ratio tested, from lean to rich mixtures, a non-linear decreasing trend of NO_x was shown with the increase of CO_2 diluent ratio. The lower NO_x at high CO_2 diluent ratios can be attributed to both the thermal and chemical effects. CO_2 diluent reduces the adiabatic temperature due to higher specific heat, which would result in a significant decrease in overall burning rate due to the reduction of laminar flame speed [11], subsequently reduces the production of thermal NO [37]. Addition of diluent to the air stream also causes a corresponding decrease in oxygen mole fraction. Consequently, the flame temperature and the mole fractions of H, O, and OH radicals reduce. On chemical effect, addition of diluents decreases the N and HCN mole fractions and subsequently reduces prompt NO [38]. At $\phi = 0.4$ for moderate H_2-rich syngases, thermal NO_x effects were not significant due to low temperature. Hence, prompt NO from the CH and CH_2 radicals and HCN from CO react to promote NO_x production, resulting in the overall higher NOx emissions compared to $\phi = 0.6$ and $\phi = 0.8$.

Figure 5b shows the NO_x emissions index for high H_2-rich syngases (S1-S5) as a function of equivalence ratio. Interestingly, all equivalence ratio tested showed similar trend of non-monotonic emissions of NO_x with diluent ratios. A slight increase of NO_x was observed when CO_2 diluent ratio increased from 5% to 10%, but the values drop to minima at diluent of 15% before increasing again at both the 20% and 25% CO_2 dilutions. However, the absolute NO_x emission (by volume) is reducing with increasing CO_2 content in the fuel. When a linear trendline is used for S1-S5, then a flatter line can be seen with increasing CO_2 content, signifying that NOx reduction is balanced out by the reduction in energy content, which in turn leads to the thermal NO producing higher temperature.

It is noted that for high H_2-rich syngases, the high content of H_2 and lack of C radicals are factors that prohibits NO_x formation. At low dilutions of 5% and 10% for $\phi = 1.0$ and $\phi = 1.1$,

NO_x emissions are evidently high. Variation of the NOx emissions is not significant between $\phi =$ 0.4, 0.6 and 0.8 at all diluent ratios. The emissions index of NO_x for baseline hydrogen (B1-B5) is shown in Fig. 5c. The NO_x emissions for fuel-lean pure hydrogen syngas shows an increase in NO_x production which is in reverse trend compared to moderate H_2-rich syngas (S6-S10). The addition of CO_2 increases the C radicals that promotes the formation of NO_x. Overall, the lean pure hydrogen baseline shows comparatively less NO_x emissions compared to high and moderate H_2-rich syngases at all equivalence ratios.

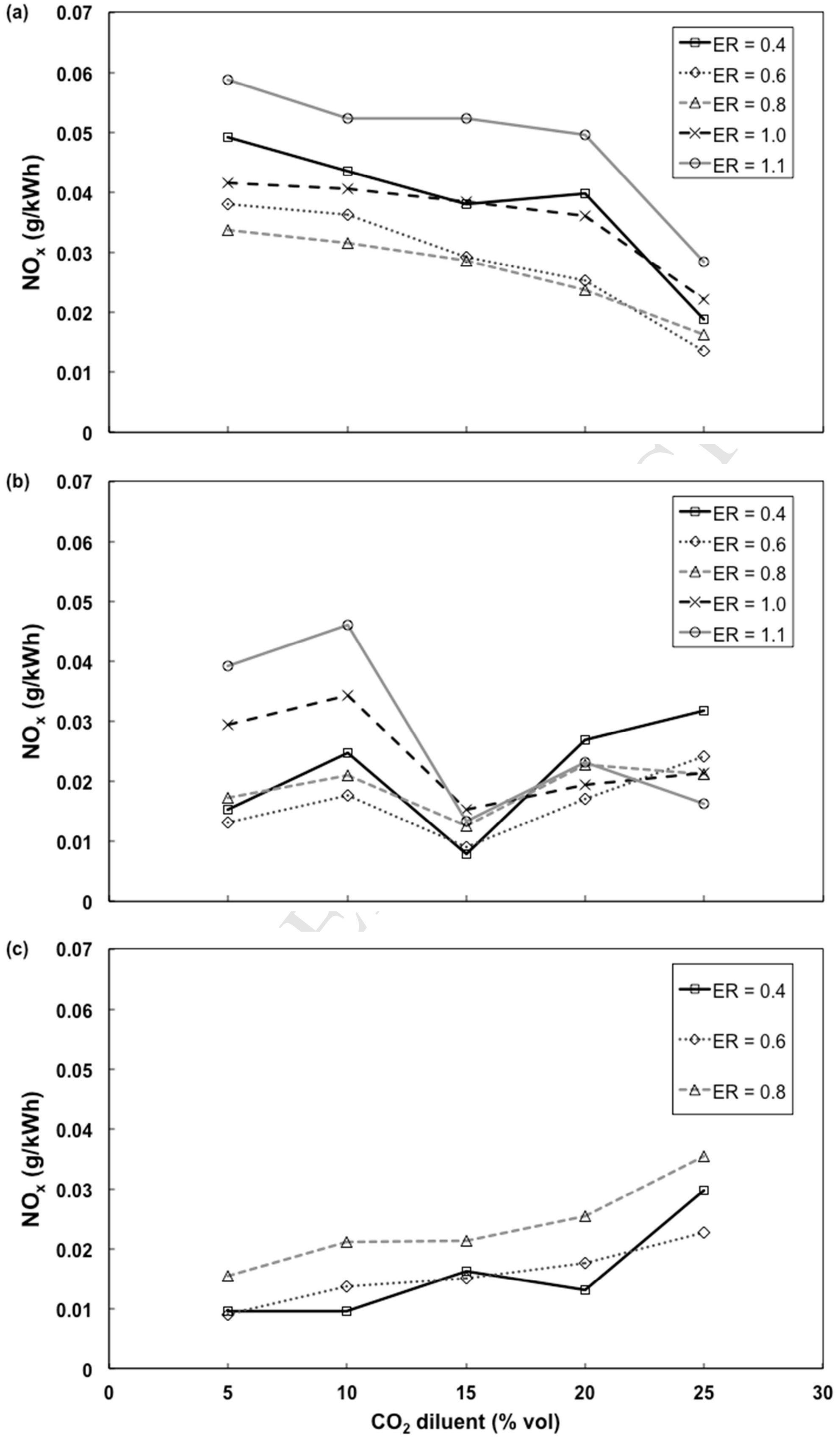

Figure 5: Emission indices of NO_x for (a) moderate (S6-S10) and (b) high H_2-rich syngas (S1-S5) and (c) baseline hydrogen (B1-B5) at different CO_2 dilutions and equivalence ratios.

Figure 6a shows the emissions index of CO in log scale for moderate H_2-rich syngases (S6-S10) at different CO_2 diluent ratios. The increase in CO_2 diluent ratios result in insignificant variation of CO for all equivalence ratios tested. For $\phi = 0.4$, CO emissions index is higher than $\phi = 0.6$ by two order of magnitude which is attributable to incomplete combustion of pockets of fuels. Emissions of CO is lower at higher equivalence ratio as a result of higher combustion efficiency. CO emissions level at $\phi = 1.0$ and $\phi = 1.1$ are rather similar across all diluent ratios tested. Compared to moderate H_2-rich syngases (Fig. 6a), high H_2-rich syngases show lower emissions for CO_2 diluent ratios of < 20% at all equivalence ratios. However, slightly higher CO emissions were observed for high CO_2 diluents ratio (20% and 25%), as shown in Fig. 6b. The increase of CO_2 diluent prevents complete oxidation of fuels at high temperatures, causing an increase in CO concentrations as the reaction rate of combustion decreased. The existence of CO_2 reduces the reactivity of O_2 for atomic hydrogen and leads to the formation of CO [39]. The baseline hydrogen syngas flames (B1-B5) show significantly low CO emissions at fuel-lean conditions in Fig. 6c. In particular, for $\phi = 0.4$, the average CO emissions is of two order of magnitude lower compared to moderate H_2-rich syngas. Further evidence of lower CO emissions was shown when equivalence ratio was increased to $\phi = 0.6$ and $\phi = 0.8$, highlighting the significant effect of CO in syngas on CO emissions.

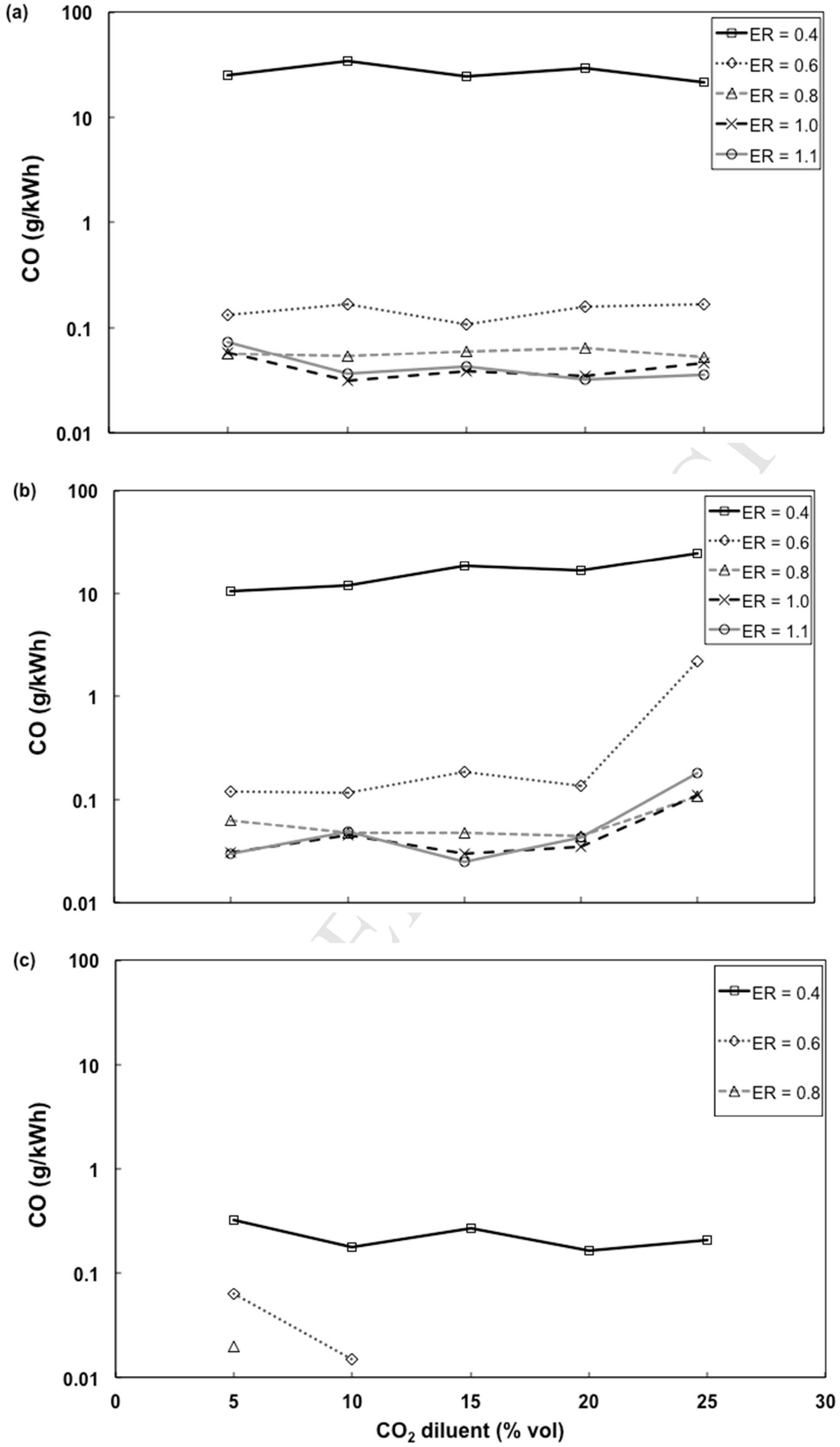

Figure 6: Emission indices of CO for (a) moderate (S6-S10) and (b) high H_2-rich syngas (S1-S5) and (c) baseline hydrogen (B1-B5) at different CO_2 diluent ratios and equivalence ratios.

3.4 Effect of CO2 diluent addition in syngases on lean blowout limit

Blowout limit can be related to Damkohler number, *Da*, where the order of magnitude of *Da* at blowout can be estimated from the values of the reference variables at the point of blowout given byt: $Da_{bo} = (\tau_{res}/\tau_{chem})_{BO} = (d_r/u_r)/(\alpha/S_L^2)_{bo}$, where τ_{res} and τ_{chem} represents the characteristic residence time and chemical time scales respectively in the combustor while, d_r is the characteristic flow length, u_r is the velocity, α is thermal diffusivity and S_L represents the laminar flame speed [23]. Blowout occurs when the characteristic chemical time exceeds the characteristic residence time.

The lean blowout limit for high (S1-S5) and moderate (S6-S10) H_2-rich syngases at different CO_2 diluent ratios is shown in Fig. 7a. In general, moderate H_2-rich syngases show consistently higher LBO limit than high H_2-rich syngases across all CO_2 dilution rates. As expected, the baseline of pure hydrogen cases (B1-B5) show lower LBO limit compared to high H2-rich syngases. By increasing the diluent ratio of CO_2, the LBO limit increases for all syngas composition as a result of reduced H_2/CO ratio. Lower H_2 fraction in the syngas leads to reduced laminar flame speed [22], resulting in short residence time that induces flame blowout. This effect is reflected in all syngas compositions including baselines. The addition of CO_2 was shown to reduce Markstein length and Lewis number apart from reducing flame speed, causing the flame to be more sensitive to stretch and increases the susceptibility to diffusive-thermal instability [29].

Figure 7b shows the lean blowout limit as a function of H_2 fraction in syngas at different CO_2 diluent ratios, combining the cases of high, moderate H_2-enriched syngases and baselines. Results show that higher H_2 content extends the flammability limit of syngas and subsequently lowering the lean blowout limit, concurring with previous reports [30, 40]. The LBO trend for all tested cases converges to show the same trend when plotted as a function of H_2 fraction by volume, highlighting the consistency of LBO limit depending on the H_2 fraction in syngases.

Higher H_2 content in the mixture leads to higher mixture reactivity rate which increases flame speed and reduces chemical time for complete reactions [22]. The relative high flame speed of H_2-rich syngases delays blowout phenomena as the velocity of the incoming unburnt reactants matches those of the flames to enable stabilisation of flame at the burner outlet [40-43]. With higher H_2 fraction in syngas, the additional H_2 in the free stream increases the concentration of radicals, thus increases the mixture's reactivity [44].

The preferential diffusion of H_2 in syngas strongly affects the diffusive-thermal instability and laminar flame speed. For syngas/air mixture, the intensity of flame front wrinkle is promoted with the increase of hydrogen fraction in the syngas due to the increased preferential diffusive-thermal instability [33]. Lewis number for lean syngas/air mixture is smaller than unity, resulting in flame cellular instability and enhances laminar flame speed. For any protuberance that appears on the flame front (stretch increasing), the local flame speed will increase, leading to flame front instability and flame wrinkling due to the development of fractal structure on the flame front [45]. It has been shown that the wrinkled flame fronts for syngas possess much finer cusps structure and higher turbulent speed compared to methane flame, as the flame intrinsic instability promotes turbulence wrinkling leading to the increase of flame front area [46, 47]. The intensity of flame front wrinkle is promoted with the increase of hydrogen fraction in syngas due to increased preferential diffusive-thermal instability, resulting in the extension of lean blowout limit.

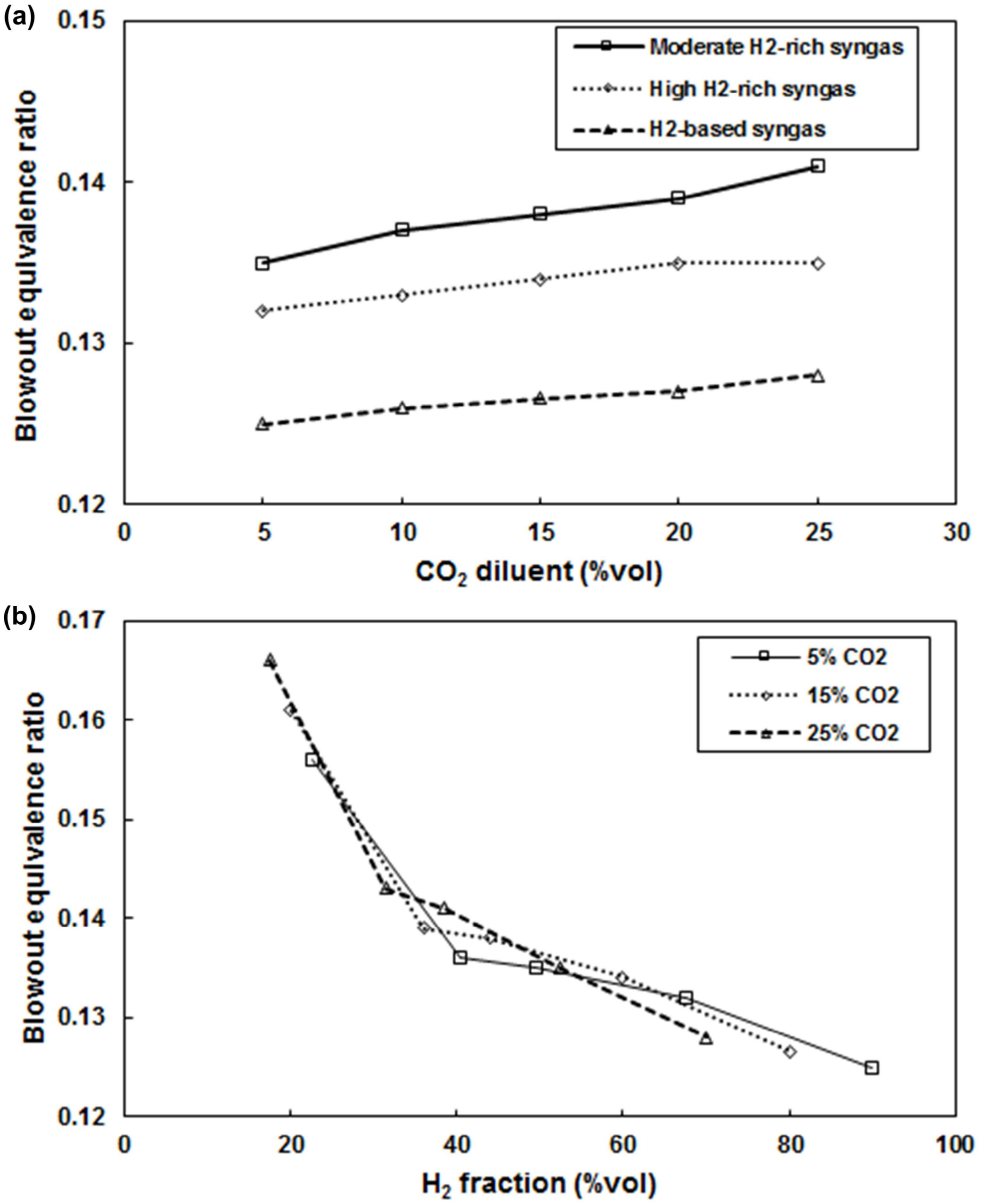

Figure 7: Lean blowout limit (a) for high (S1-S5) and moderate (S6-S10) H_2-rich syngases compared against baseline (B1-B5) H_2 flames and (b) as a function of percentage of H_2 fraction in syngas at different CO_2 diluent ratios.

4. Conclusion

The combustion characteristic of rich-H_2 syngases were investigated in a medium swirl stabilised premixed combustor at various equivalence ratios. Direct flame imaging shows that high H_2-rich syngas exhibits cleaner flames with less luminous orange-yellowish flame in the post-flame zone, indicating the low tendency for soot formation. The high H_2-content syngas shows a flame reaction zone length that is more intense and shorter due to reduced flame thickness compared to moderate H_2-enriched syngas. From the emission tests, moderate H_2-rich syngas shows higher NO_x emissions than high H_2-rich syngas at all equivalence ratios tested attributable to the presence of C from CO that promotes the formation of NO_x. High H_2-rich syngas shows slightly higher NO_x emissions compared to baseline pure hydrogen gas. For CO emissions, both the high and moderate H_2-rich syngases show similar CO profiles. Both syngas type show higher CO than pure hydrogen baseline at fuel-lean regions. The effect of CO_2 diluents on NO_x emissions is pronounced for moderate H_2-rich syngas, where a decreasing trend of NO_x emissions was shown with increasing CO_2 diluent ratios. The effect of diluents on CO is not evident for moderate H_2-rich syngases, but slightly more evident for high H_2-rich syngas at high diluent rates (>20%). The results show that higher fraction of H_2 in the syngas relative to CO can potentially achieve both low NOx and CO simultaneously Diluting the syngas with CO_2 results in blowout at higher equivalence ratio as the laminar flame speed decreases. High fraction of H_2 in syngas exhibits extended flammability limit and lower lean blowout limit. The effect of preferential diffusion of H_2 leads to higher mixture reactivity rate, increases flame speed and stability due to extended lean blowout limit.

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Title: H_2-Rich Syngas Strategy to Reduce NO_x and CO Emissions and Improve Stability Limits Under Premixed Swirl Combustion Mode

Highlights

- Higher fraction of H_2 in syngas produces lower NO_x per kWh basis.
- CO emissions is minimally affected by the H_2 fraction in syngas.
- Increasing CO_2 diluent ratio in H_2-rich syngas reduces NO_x emissions but minimally affects CO.
- Higher fraction of H_2 in syngas results in lower lean blowout limit.
- Higher CO_2 diluent ratio increases lean blowout limit for syngas.