AMMONIA AND BIOMASS CO-FIRING IN A PILOT-SCALE FLUIDIZED BED REACTOR: A NUMERICAL STUDY FOR COAL PHASE-OUT

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Abstract -Recent advancements point towards the co-firing of coal with biomass and NH3 as a state-of-art strategy to phase-out coal from coal-based power stations. The implementation of NH3 to the coal-biomass fuel mix allows not only to decarbonize coal-fired processes but also reduces biomass exploration costs to avoid feedstock availability disruption while increasing the power station production efficiency. Moreover, such a strategy brings an alternative player into the environmentally friendly power mix portfolio, accelerating the coal phase-out from electricity production. However, the use of NH3 for power generation still presents some research gaps. This study delivers a 2D Eulerian-Lagrangian numerical model to describe the co-firing of coal with biomass and NH3 in a pilot-scale fluidized bed reactor. The numerical model is validated against experimental data on coal and biomass combustion to assess the model's accuracy. The coal, biomass, and NH3 co-firing ratios are varied to the point where coal is gradually reduced from the fuel mix, and its effect on the combustion processes is broadly investigated to determine the biomass and NH3 impact on the reactor's emissions. The use of both biomass and NH3 showed to broadly reduce carbon and NO emissions from coal-fired energy systems, emphasizing the potential addressed to biomass as an alternative to coal and NH3 as an effective carbon-neutral fuel of the future.

Keywords -Ammonia, Coal, Biomass, Co-firing, Simulation, Emissions reduction

I. INTRODUCTION

Currently, coal still supplies over one-third of the global electricity generation, with a total share of 36.7% in the global power mix in 2019. In 2018, coal-fired power generation emitted more than 10 Gt of CO2 [1]. Given this purview, the ongoing issues related to climate change have increased the interest in decreasing coal energy reliance.

Ammonia (NH3) is a necessary commodity chemical with a mature storage and transportation network. As a carbon-free fuel, NH3 carries great potential with promising applications in energy systems. Plus, NH3 can be obtained through renewable energy sources, known as green NH3, highlighting NH3 as the next sustainable fuel solution for power generation. NH3 is also a suitable hydrogen (H2) energy vector, enabling future CO2-free energy systems [1].

At the UN's Climate Change Conference (COP26), more than 40 countries pledged to shift away from coal. However, despite the clean-power transition hype, the current global energy crisis triggered a dash for coal again, showing that the rapid transition to renewables may indeed be far more challenging than expected [1]. To work around this issue, one should consider a gradual fossil fuel phase-out allowing time for zero-carbon technologies to establish fully. The co-firing of coal and NH3 is an example of such promising alternative technologies, allowing the retrofitting of coal power facilities without major modifications while contributing to global decarbonization goals.

To the best of our knowledge, no biomass-NH3 co-firing studies can be found. However, the co-firing of coal and NH3 alone in coal-fired thermal power plants has been attempted through various approaches. The Chugoku Electric company conducted the first NH3-coal co-firing in 2017 at its 156 MW Mizushima Thermal Power Station, using a fuel mix of 0.6% to 0.8% NH3. Japan's Green Growth Strategy already laid out a strategy to begin testing co-firing NH3 in coal power plants in 2021, boosting its usage by 2030. However, NH3 co-firing is still in its early stages, with a recently achieved 20% NH3 fuel mix (calorific value) by the IHI Corporation [2]. Some works have already simulated the co-firing of NH3 with coal in furnaces and boilers [3, 4], but none addresses NH3 co-firing in a fluidized bed reactor. This work aims to contribute to this disruptive topic by developing a 2D Eulerian-Lagrangian numerical model set to describe the co-firing of coal with biomass and NH3 in a pilot-scale fluidized bed reactor. The numerical model is validated against experimental data on coal and biomass combustion to assess the model's accuracy. The study delves into coal-NH3 and coal-biomass-NH3 blends' co-firing to assess its effect on carbon

and NO emissions reduction from coal-fired power systems.

II. EXPERIMENTAL SETUP

The experiments were conducted in a thermally insulated pilot-scale bubbling fluidized bed reactor measuring 0.25 m wide by 2.3 m tall. Figure 1 shows a detailed schematic of the combusting facility.

Quartz sand was used and bed material with a density of 2650 kg/m3 and particle size around 500 µm. Bituminous coal and forest residues were fed into the reactor at a rate of 17-50 g/min from 0.35 m above the distribution plate. Bituminous coal held a density of 1346 kg/m^3 and was sieved to a particle size of 500 to 4000 µm, while forest residues had a density of 478 kg/m3 and particle size range from 1000 to 4000 µm. Table 1 shows the properties of the feedstocks. Regarding the airflow, the first stream was heated and supplied from the bed's bottom, keeping a fluidization speed between 0.28 and 0.32 m/s depending on the operating condition. At the same time, the second stream was fed about 0.30 m above the bed surface. Thus, both streams provided a total airflow of 250 NL/min. Eight water-cooled probes arranged along the bed maintained the bed temperature within the desired range The excess air and air staging were established by adjusting the coal feed and secondary airflow rates. Air staging was used, with the primary air at the bottom accounting for 100, 80, and 60% of the total combustion air. Experiments were carried out under steady-state conditions with three surplus air levels (10, 25, and 50%) and three operating temperatures (1023, 1098, and 1173K). Further details concerning the experimental setup and combustion facility can be found in [5, 6].

Material	Bituminous coal	Forest residues
Proximate analysis (wt.%, wet basis)		
Moisture	4.4	9.0
Ash	13.8	2.0
Volatile matter	28.5	70.0
Fixed carbon	53.3	19.0
Ultimate analysis (wt.%, dry basis)		
Ash	13.8	2.5
C	65.97	49.8
H	4.27	6.7
N	1.67	3.0
S	0.88	n.d.
O (by difference)	13.41	38.0

Table 1. Chemical composition of coal and biomass.

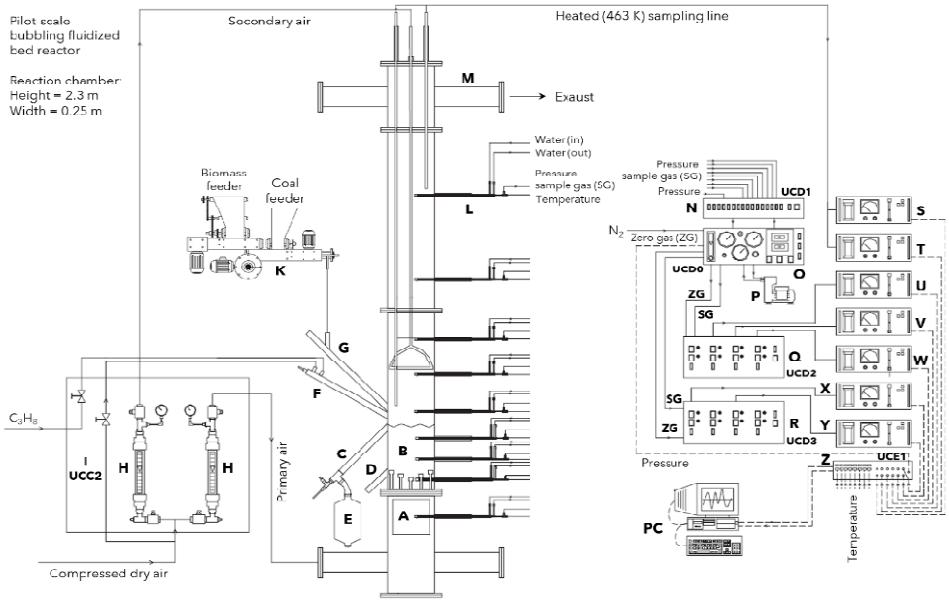


Figure 1. Detailed schematic of the experimental combustion unit: Dashed line - Electric circuit; Continuous line - Pneumatic circuit; A - Primary air heating system; B - Sand bed; C - Bed solids level control; D - Bed solids discharge; E - Bed solids discharge silo; F - Propane burner system; G – Coal addition port; H - Airflow meter; I - Control and command unit (UCC2); K – Coal and biomass feeder; L - Gas sampling probe; M - Exhaust duct; N, O, Q, R - Command and gas distribution units; P - Gas sampling pump; PC - Computer data acquisition and control system; S, T, U, V, W, X, Y – Automatic online gas analysers for determination of O2, CO2, CO, N2O, NO, SO2, HC; Z – Electronic command unit; UCD0, UCD1, UCD2, UCD3 - Electro-pneumatic command and gas distribution units; UCE1 - Electronic command unit.

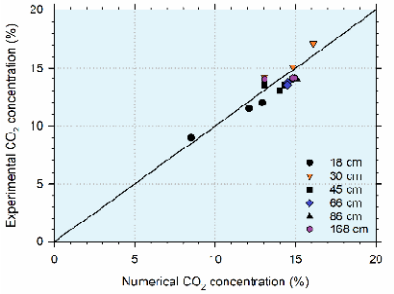
III. MATHEMATICAL MODEL

The developed 2D numerical model is based on an Eulerian-Lagrangian method, with the gas phase set within the Eulerian framework, while the solid phase is monitored separately through the Lagrangian approach. The Lagrangian scheme assumes that solid fuel particles are volatile materials, char, and ash, represented by spherical particles with a given size distribution. The direct interactions between the gas and solid phases were defined considering mass exchange (due to heterogeneous chemical processes), momentum (due to drag between stages), and energy. The k-ε realizable model was selected for the turbulence model as it offers increased prediction to the standard k-ε model, which is already resilient and relatively accurate. Table 2 briefly describes the model's governing equations. The chemical reactions model and corresponding rate coefficients used in the simulations are summarized in Table 3.

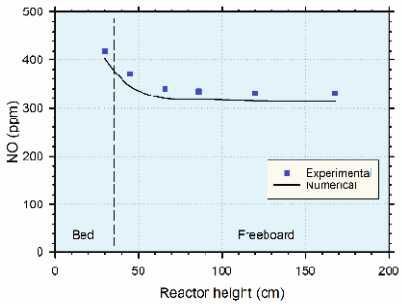
IV. RESULTS AND DISCUSSION

4.1 Numerical model validation

Figures 2a and b compare the experimental and numerical results for CO2 and NO concentrations at various coal and biomass combustion heights, respectively. The simulations were carried out for bituminous coal and forest residues following the experimental results attained from [5, 6]. The results show that the mathematical model could predict the CO2 and NO concentrations at several positions within the reactor with very reasonable agreement. Concerning the CO2 concentration, the tight dispersion from the line means decreased error variance, while a close resemblance between experimental and numerical values can be seen for NO concentration. After validating the model for both coal and biomass combustion in a pilot-scale reactor, the co-firing of coal, biomass, and NH3 in the same system was then investigated. Since the numerical model could predict the combustion process reasonably, it is safe to assume that it can also simulate the same conditions using coal and biomass with NH3 implementation. Furthermore, the research group already has extensive experience developing mathematical models describing thermochemical processes for various feedstocks, experimental conditions, and reactor scales [7].



(a)



(b)

Figure 2. Experimental and numerical results for (a) CO2 concentration from coal combustion and (b) NO concentration from biomass combustion.

4.2 Coal-NH3 co-firing: effect on the emissions reduction

A decreasing trend of CO2 emissions at the reactor's outlet with the increase in NH3 co-firing ratio can be seen in Figure 3a. NH3 co-firing ratio simulations were performed from 0% to 80% (by mass), where 0% portrays pure coal firing. Since NH3 is free of any carbon, its replacement in coal combustion directly leads to CO2 emissions reduction by eliminating coal carbon ($CxHyOz+O2 \rightarrow CO2+H2O$). Furthermore, NH3 injection in the flue gas is known for inhibiting the CO oxidation to CO2 ($CO+0.5O2 \rightarrow CO2$) by consuming all the OH radicals needed for the oxidation to occur [8]. At a 20% co-firing ratio, CO2 emissions decreased up to 26% compared to pure coal firing, meeting the 20% range addressed in the literature [9].

The water-gas shift reaction ($CO+H2O \rightarrow CO2+H2$), as well as NH3 combustion ($NH3+O2 \rightarrow NO+H2O+0.5H2$, and $NH3+NO \rightarrow N2+H2O+0.5H2$), are likely to have a substantial role in reducing CO emissions (see Figure 3b). Due to the increased H2 content of the overall feed, the H2O concentration in the flue gas rises significantly, pushing the CO to react with H2O and so decreasing its concentration [9]. In Figure 3c, the NO concentration is almost identical to pure coal firing until a co-firing ratio of 10% is attained. At a 20% ratio, a modest rise in NO level is seen, which is typically ascribed to increased combustion intensity and an increase in fuel-NOx from the NH3 injection [3, 10]. When the ratio of unreacted NH3 exceeds 20%, NO emissions steadily decrease due to the DeNOx effect of the rising quantity of unreacted NH3. The NO reduction reaction with NH3 ($NH3+NO \rightarrow N2+H2O+0.5H2$) plays a critical role in this process since NH3 interacts with NO to create mainly N2 and H2O, reducing the amount of NO formed by other NH3 consumption routes. Furthermore, the air staging significant influence on NO formation is the restricted airflow created by the bed, which acts in a fuel-rich/O2 deficient, preventing NO formation. Therefore, the higher the air staging level, the less the fuel-NH3 proportion transformed into NO will be. Besides, air staging produces a more extended reduction zone upstream of the second

airflow input, giving more time for the NO reduction reaction with NH3. A 40% decrease in NO levels was achieved, which is consistent with experimental trends reported in the literature for coal-NH3 co-firing in fluidized bed reactors operating under comparable circumstances [11].

In Figure 3d, for 10% and 20% co-firing ratios low NH3 emissions are predicted (<5 ppm); as the NH3 co-firing ratio increases, the amount of unreacted NH3 at the reactor outlet progressively increases as well. The slippage of NH3 is a significant problem that must be addressed while conducting NH3 combustion, and such levels must be monitored and minimized. NH3 emissions standards are extremely variable, but some reports state that an average of acceptable NH3 emission factors for coal fluidized bed reactors coupled with SNCR (Selective Non-Catalytic Reduction) systems are set around 20 to 30 ppm [12]. Increasing NH3 injection may benefit NO reduction, but it may also increase the risk of unburned NH3.

Conservation equations
Energy (gas phase):
$\frac{\partial}{\partial t}(\alpha_g\rho_g h_g) + \nabla\cdot(\alpha_g\rho_g\vec{v}_g h_g) = \alpha_g\left(\frac{\partial p}{\partial t} + \vec{v}_g\cdot\nabla p_g\right) + \phi - \nabla\cdot(\alpha_g q) + \dot{Q} + S_h + \dot{q}_D$
Mass (gas phase):
$\frac{\partial}{\partial t}(\alpha_g\rho_g) + \nabla\cdot(\alpha_g\rho_g\vec{v}_g) = \delta\dot{m}_s$
Momentum (gas phase):
$\frac{\partial}{\partial t}(\alpha_g\rho_g\vec{v}_g) + \nabla\cdot(\alpha_g\rho_g\vec{v}_g\vec{v}_g) = -\alpha_g\cdot\nabla p_g + F + \alpha_g\rho_g g + \nabla\cdot(\alpha_g\tau_g)$
Energy (discrete solid phase):
$m_i c_i\frac{dT_i}{dt} = h_i A_{pi}(T_g - T_i) + \frac{\varepsilon_i A_{pi}}{4}(G - 4\sigma T_i^4) - H_{evap}\mathrm{d}m_{vapor} - H_{O_2/CO_2/H_2O}\,dm_{C-O_2/CO_2/H_2O}$
Momentum (discrete solid phase):
$\frac{dv_i}{dt} = \frac{3\mu_g C_D Re_p}{4\rho_i d_i^2}(u_g - v_i) + g\left(1 - \frac{\rho_g}{\rho_i}\right)$, $C_D = \begin{cases}\frac{24}{Re_p}\left(1 + \frac{1}{6}Re_p^{\frac{2}{3}}\right) & Re_p \le 1000 \\ 0.424 & Re_p > 1000\end{cases}$, $Re_p = \frac{\rho_g d_i\lvert u_g - v_i\rvert}{\mu_g}$
Mass (discrete solid phase):
$\frac{\mathrm{d}m_i}{dt} = \frac{\mathrm{d}m_{vapor}}{dt} + \frac{\mathrm{d}m_{devol}}{dt} + \frac{\mathrm{d}m_{C-O_2}}{dt} + \frac{\mathrm{d}m_{C-CO_2}}{dt} + \frac{\mathrm{d}m_{C-H_2O}}{dt}$
Turbulence model
Kinetic energy:
$\frac{\partial}{\partial t}(\rho k) + \frac{\partial}{\partial x_j}\left(\rho k_{u_j}\right) = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\sigma_k}\right)\frac{\partial k}{\partial x_j}\right] + G_k + G_b - \rho\varepsilon - Y_M + S_k$
Dissipation rate:
$\frac{\partial}{\partial t}(\rho\varepsilon) + \frac{\partial}{\partial x_j}\left(\rho\varepsilon_{u_j}\right) = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_t}{\sigma_\varepsilon}\right)\frac{\partial\varepsilon}{\partial x_j}\right] + \rho C_1 S_\varepsilon - \rho C_2\frac{\varepsilon^2}{k + \sqrt{v\varepsilon}} + C_{1\varepsilon}\frac{\varepsilon}{k}C_{3\varepsilon}G_b + S_\varepsilon$

Table 2. Mathematical model governing equations [1].

Reactions	**Arrhenius reaction rates**
Volatiles combustion	
$R_{1(coal)} = C_xH_yO_z + (x + y/4 - z/2)O_2 \rightarrow xCO_2 + (y/2)H_2O$	$r_1 = 657000\,T\,exp\left(\frac{-8.02\times10^7}{T}\right)[C_xH_yO_z]^{0.5}[O_2]$
$R_{2(biomass)} = C_xH_yO_z + (x + y/4 - z/2)O_2 \rightarrow xCO_2 + (y/2)H_2O$	$r_2 = 9.2\times10^8\,T\,exp\left(\frac{-1.3576\times10^8}{T}\right)[C_xH_yO_z]^{0.5}[O_2]$
Homogeneous reactions	
H_2 combustion:	
$R_3 = H_2 + 0.5O_2 \leftrightarrow H_2O$	$r_3 = 5.69\times10^{11}\,exp\left(\frac{-1.47\times10^8}{T}\right)[H_2][O_2]^{0.5}$
CO combustion:	
$R_4 = CO + 0.5O_2 \rightarrow CO_2$	$r_4 = 1.93\times10^{13}\,T^{-2}exp\left(\frac{-1.26\times10^8}{T}\right)[CO][O_2]^{0.5}$
Water-gas shift:	
$R_5 = CO + H_2O \leftrightarrow CO_2 + H_2$	$r_5 = 2.75\times10^9\,exp\left(\frac{-8.36\times10^7}{T}\right)[CO][H_2O]$
NH_3 combustion:	
$R_6 = NH_3 + O_2 \rightarrow NO + H_2O + 0.5H_2$	$r_6 = 350\,T^{7.65}\,exp\left(\frac{-5.24\times10^8}{T}\right)[NH_3][O_2]$
$R_7 = NH_3 + NO \rightarrow N_2 + H_2O + 0.5H_2$	$r_7 = 4.24\times10^5\,T^{5.3}\,exp\left(\frac{-3.5\times10^8}{T}\right)[NH_3][NO]$
NH_3 pyrolysis:	
$R_8 = NH_3 \rightarrow 0.5N_2 + 1.5H_2$	$r_8 = 0.185\,T^{1.25}\,exp\left(\frac{-6.9\times10^7}{T}\right)[NH_3]$
Heterogeneous reactions	
Char combustion:	
$R_9 = C(s) + 0.5O_2 \rightarrow CO$	$r_9 = 0.052\,exp\left(\frac{-6.1\times10^7}{T}\right)$
Boudoir reaction:	
$R_{10} = C(s) + CO_2 \rightarrow 2CO$	$r_{10} = 0.0732\,exp\left(\frac{-1.125\times10^8}{T}\right)$
Water-gas reaction:	
$R_{11} = C(s) + H_2O \rightarrow CO + H_2$	$r_{11} = 0.0782\,exp\left(\frac{-1.15\times10^8}{T}\right)$

Table 3. Chemical reactions model [1].

Therefore, a balance in the NH3/NO ratio is required to enhance NH3 reactivity and thus diminish NH3 slip in the unburned streams [13].

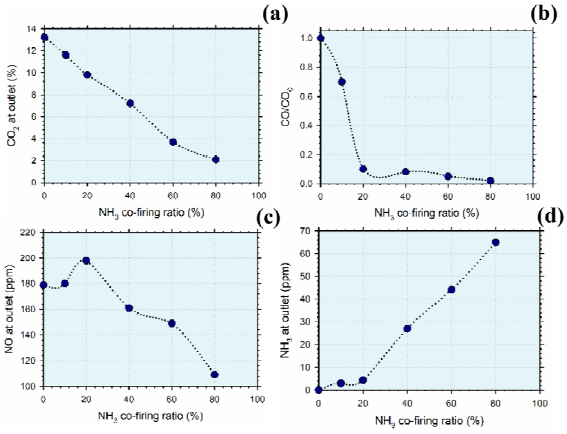


Figure 3. Effect of coal-NH_3 co-firing on (a) CO_2, (b) normalized CO, (c) NO, and (d) NH_3 emissions.

4.3 Coal-biomass-NH_3 co-firing: effect on the emissions reduction

The combined effect of biomass and NH_3 co-firing with coal maintained identical trends to the ones attained from coal-NH_3 co-firing, with CO_2, normalized CO, and NO gradually decreasing with the biomass-NH_3 ratio increase, while normalized NH_3 emissions increase (Figures 4a, b, c, and d). The co-firing of coal and biomass, in particular, has already been long established and demonstrated as an effective measure to reduce greenhouse gas emissions and environmental pollution [14]. Here, biomass and NH_3 co-firing ratios were raised to 50 and 40% (by mass), respectively, while coal was gradually phased out from the mixture. Lessons learned from coal-NH_3 co-firing showed that 40% NH_3 ratio operation (<30ppm) was possible regarding NH_3 emissions, although some emissions restraints could be applied depending on local environmental regulations and policies [15]. Whereas at 60 to 80% ratios, substantial abatement measures for NH_3 emissions would be required for feasible operation. Thus, given these assumptions, a maximum of 40% NH_3 ratio was kept in the fuel mix.

Both biomass and coal are carbon-bearing fuels, yet CO_2 emissions are directly related to the fuel's carbon content [16]. The bituminous coal used in this study has higher fixed carbon and carbon elements than forest residues. Hence, fossil CO_2 emissions were

continuously reduced by displacing coal and increasing the share of biomass and carbon-free NH_3 (Figure 4a). The same reasoning applies to CO emissions, as they decrease with both coal feed reduction and the increase in biomass and NH_3 ratio (Figure 4b) [13]. Ultimately, biomass is regarded as a carbon-neutral resource; therefore, CO_2 emission should decline proportionally to the amount of coal offset by biomass [17].

Apart from the previously discussed effect of NH_3 on NO abatement during coal co-firing, several advantages are addressed to biomass in reducing NO emissions (Figure 4c). Biomass contains lower nitrogen content than coal leading to reduce NO emissions. Plus, during combustion, most of the fuel nitrogen in biomass is converted to NH radicals (mainly NH_3), NH_3 on the other hand, it reduces NO to N_2, hence lowering NO emissions [18]. Also, both biomass and NH_3 are known for decreasing peak flame temperature within the reactor [19]. Biomass's lower calorific value and NH_3's low flammability and reluctance to combustion contribute to NO emissions abatement, as higher temperatures promote NO formation [20]. This situation occurs as lower flame temperature leads to more char being generated. NO formation is reduced by char. Thus, as the flame temperature decreases, NO reduction by the char increases, leading to lower NO emissions [9]. Lastly, the high volatile content of biomass can also be used as a reburn fuel for NO reduction, providing biomass with additional potential in NO emissions abatement from coal combustion [18]. Similar to coal-NH_3 co-firing, NH_3 emissions showed to build up with the biomass-NH_3 ratio increase. As aforementioned, this increase is mainly attributed to the NH_3 ratio increase in the fuel mix. Most of the nitrogen in biomass is converted to NH_3 products during combustion, yet NH_3 destruction occurs to be converted into N_2 and H_2 [3]. Given the numerical results, the analysed fluidized bed reactor could operate with coal-biomass-NH_3 up to 20% NH_3 with no NH_3 slip.

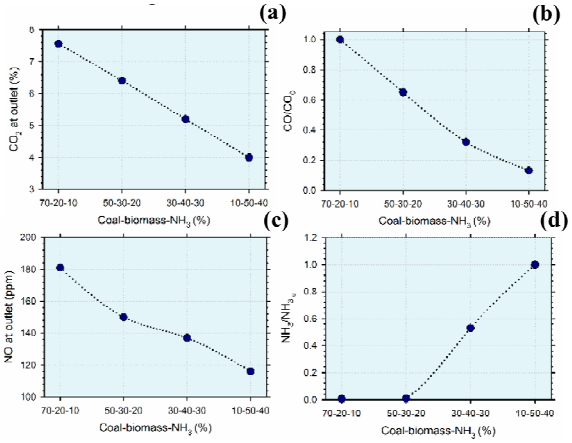


Figure 4. Effect of coal-biomass-NH_3 co-firing on (a) CO_2, (b) normalized CO, (c) NO, and (d) normalized NH_3 emissions.

V. CONCLUSIONS

This numerical study showed that through biomass and NH_3 co-firing in a pilot-scale fluidized bed reactor, one can reduce carbon and NO emissions from coal combustion. Co-firing with NH_3 reduces CO_2 emissions by up to 26% compared to pure coal combustion. CO emissions decreased when the flue gas H_2O content increased due to NH_3 combustion, which caused CO to react with H_2O, increasing CO abatement. UC concentrations were reduced by NH_3 co-firing. However, NO emissions remained at the same level as pure coal combustion at a 10% ratio but fell by up to 40% at 20%-80% ratios owing to the $DeNO_x$ effect and the NH_3 catalytic action of reducing NO to create N_2 and H_2O. Small concentrations of NH_3 emissions were predicted at 10%-20% ratios, while at 40% co-firing and above, the amount of unreacted NH_3 increased. Overall, the results support existing evidence for the use of NH_3 as a viable energy carrier for decarbonizing energy-intensive sectors while meeting previous experimental demonstrations sustaining that the co-firing of NH_3 in coal-fired units is achievable with minor modifications to the existing systems within a 20% NH_3 range.

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