Optimization of multi variant process parameters to reduce emissions of high sulfur coal during combustion in fluidized bed facility

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Abstract
Pakistan has 185 billion tonnes reserves of coal but unfortunately the quality of 80% of this coal is not good. As the country has a shortage of energy so it is necessary to refine the coal before it can be used to produce electricity. In this sense, this research is very important as it enables indigenous coal to meet the increasing energy demand. This study is focused on the control of sulfur emissions during the combustion of high sulfur Pakistani coal from the Trans Indus Range and the Salt Range. Parameters like Ca/S molar ratios (MRs), limestone particle size, bed temperature, and different percentages of biomass in co-firing with coal have been studied. It is showed that desulfurization of coal was maximum with the fine-sized particles of limestone. Co-firing of moderate quantity biomass exhibited a considerable decrease in SO₂ emissions. Results achieved are presented in the form of tables and plots. This study for control of the gaseous emissions from combustion in FBC facility has great potential for new coal based power projects, especially in Pakistan.

Keywords
Fluidized bed combustion, coal, combustion emissions, sulfur control, multi variant process

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Introduction
Coal is the world’s most abundant and widely distributed fossil fuel, with global proven reserves of nearly 1000 billion tonnes and has been a key component of electricity generation mix worldwide.¹,² Coal provides more than 40% of the world’s electricity³,⁴ and this share is predominant for some countries such as South Africa (93%), Poland (92%), China (79%), India (69%), and the United States (49%). Moreover, the growing energy needs of the developing world are leading to the fact that coal will remain a key component of the power regardless of climate change policy.⁵

In Pakistan, power generation from coal is currently gaining more interest, relative to oil and gas.⁶ Coal in Pakistan is of low grade such as lignite in Sindh (Thar and Lakhra), sub-bituminous in Baluchistan and Punjab (the Salt Range and Trans Indus Range).⁷ The coal reserves of the Salt Range and Trans Indus Range are approximately 500 million tons.⁸ In general

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Pakistani coal contains high sulphur (ranging from 3% to 11%) except Thar coal. Coal based power generation could represent an economic solution by using low-grade Pakistani coals and their blends with better quality imported coals, which would ensure high performance and compliance with environmental regulations. FBC technology could prove a better option to utilize such low-grade high sulfur coals in coal fired steam as well as power generation systems with controlled emissions of nitrogen oxides (NOx) and sulfur oxides (SOx).

In the literature, desulfurization of high sulfur Pakistani coals during combustion has not been fully investigated. Zaidi\textsuperscript{13} performed the study of coal reactivity as well as char formation of different coal samples collected from Punjab, Sindh, and Baluchistan. Iqbal et al.\textsuperscript{14} studied the effect of particle size on different combustion parameters of the coal from Islamkot Thar. Naveed et al.\textsuperscript{15} investigated the gasification perspectives of the Eastern Salt Range coals and found the coal suitable for gasification. Daood et al.\textsuperscript{16} performed the study of gaseous emissions of Thar coal such as SO\textsubscript{2}, NO\textsubscript{x}, CO, and CO\textsubscript{2} in pilot-scale facility. Rehman et al.\textsuperscript{9} have already studied the combustion perspectives along with geology and coalfield stratigraphy of the coals from Salt Range and Trans Indus Range Punjab and concluded that the coal has good combustion characteristics, low slagging, and medium fouling Indices. However, the work on desulfurization of high sulfur Pakistani coal has seldom been addressed in the open literature and thus needs to be studied comprehensively using different methods including limestone addition and co-firing with biomass.

In this paper authors have attempted to present the results of SO\textsubscript{2}, NO\textsubscript{x}, CO, and CO\textsubscript{2} concentration measurements obtained in a pilot-scale FBC, with and without continuous limestone addition. The results of SO\textsubscript{2} were also presented for co-firing of coal with different percentages of biomass, and to offer some qualitative interpretation relevant to the understanding of the SO\textsubscript{2} behavior during coal combustion. The main parameters of the desulfurization process in FBC such as different MRs, adsorbent (limestone) particle size, bed temperature, and co-firing with biomass were analyzed on the basis of the higher initial concentration of SO\textsubscript{2} in combustion furnace.

This research work is very important as the process and methodology given help to utilize the coal available domestically. It also helps to reduce emissions resulting from the burning of coal. A substantial amount of coal reserves can be used as a fuel alternative to meet energy demand for a country like Pakistan.

**Materials and methods**

The test facility is installed at the Low Carbon Combustion Center, University of Sheffield, UK and it comprises a fluidized bed combustion (FBC) section, having overall dimensions of (1 m × 1 m × 5 m) with an extended freeboard section. Bed of sand, Garside Leighton Buzzard 14/25 range with nominal particle size of 1.18-0.60 mm in this case, was fluidized by air flowing through a standpipe distributor. The internal dimensions of the fluidized bed and the combustion section above the fluidized bed are 0.42 m × 0.38 m and 0.58 m × 0.38 m, respectively. The combustion section is lined with castable refractory backed with ceramic fiber (having low thermal conductivity) based insulation. The bed having an area of 0.16 m\textsuperscript{2}, is capable of accepting bio-fuels/wastes feed rate up to 100 kg/h and mineral feed rate up to 500 kg/h depending on plant operating conditions and material type. The rig can be operated with fluidizing velocities of up to 3.5 m/s and fluidized bed heights of up to 700 mm, at combustion temperatures ranging from 750°C to 950°C.

There are three variable speed screws with feed hoppers to form the above–bed solid fuel feed system. All of them discharge into a high speed screw having 125 mm diameter which acts as a part-filled transfer screw and transfers the material onto the surface of the fluidized bed or into the splash zone. The system allows the feeding of three different materials simultaneously and capable to handle a variety of solid granular as well as powder materials.

The fluidized bed is instrumented with temperature and pressure ports for monitoring bed and freeboard conditions. Five thermocouples are arranged in a vertical array at equal space, the bottom and top ones are at 30 and 450 mm from the top of the air distributor. In the access door, an off-take port is provided which allows overflow of bed material when the nominal fluidized height reaches 0.35 m. To withdraw either a fraction of the selected coarse size or the whole bed, an air classifier of 68 mm diameter is given in the base of the fluidized bed.

The positions of all thermocouples and pressure transmitters, to record temperature and pressure respectively, are shown in Figure 1. The transmitters are connected to a National Instrument (NI) TBX-1303 module which is then linked to a NI SCXI-1102 (temperatures) and SCXI-1100 (pressures) and lab-view is used to read them via a NI PCI-6035E card in the computer.

Manual control valves at primary air and secondary air lines as shown in Figure 1 were used to regulate fluidizing air flow rate through the fluidized bed and above the bed, respectively. The Inverter control of the FBC exhaust fan enables the operator to maintain a negative pressure above the bed, in the mid-freeboard and subsequent sections. Euro-therms PID controllers enable automatic and manual control of the fuel flow of each screw metering feeders into the bed.
Lab view software was used for logging the data of all the temperatures and pressures. The oxygen concentration was measured by a Portable Oxygen Analyzer (Servomex 570). A forced draught fan supplies fluidizing air which enters the bed through a standpipe distributor. A manual damper controls the airflow rate, while start-up is achieved by lighting the premixed gas at the primary burner.

The FBC consists of an air-cooled heat exchanger to reduce the flue gases temperature, a bag filter, and a cyclone to clean the flue gas. The fluidized bed and freeboard sections are made of mild steel with interior refractory lining which results in a low outer casing temperature and consequently low heat losses.

The flue gases leaving the fluidized bed are cooled in the air-cooled heat exchanger using forced draught air. The cyclone is used to separate most of the suspended solids from the flue gases leaving the heat exchangers. The solid particles are discharged into a sealed drum and can be collected for further analysis. The flue gases leaving the cyclone are passed either through the bag filter or discharged directly to the atmosphere via the induced draft fan as shown in Figure 1. The temperature of the flue gases is reduced down to 500°C, depending on operating conditions. Figure 2 and 3 show the main parts of FBC, and process flow diagram of lab scale and pilot scale testing respectively.

All reported values of gaseous emissions like SO₂, NOₓ, etc. are corrected to 6% O₂ in the flue gas using the following equations (1) and (2).

\[
SO_2 \text{ at } 6 \% O_2(\%) = \frac{SO_2}{(20.9\% - 6\%)} \\
NO_x \text{ at } 6 \% O_2(\%) = \frac{NO_x}{(20.9\% - 6\%)}
\]

The bag filter houses ceramic elements to enable the plant to be operated with a range of flue gas exit temperatures, and also to provide information on cleaning and cake properties for full-scale plant proposing to use ceramic filters.

In this experimental work, two different types of coals were used for desulfurization and emission study of high sulfur coal from Trans Indus Range (coal A) and Salt Range (coal B), Punjab, Pakistan. Limestone for desulfurization of the said coals was also taken from the Salt Range area. Coal and limestone were continuously added to the bed, keeping control of the bed height. The feed rate of limestone was in the range of 12–60 g/min, while that of coal was from 150 to 300 g/min. The bed was continuously monitored during the experiments. All the measurements (temperature and gas composition) were taken with the FBC reactor.
Figure 2. Actual testing facility and main parts of FBC.
1. Coal storage, 2. Coal feeder, 3. Limestone feeder, 4. Screw feeder, 5. Air plenum, 6. Bed, 7. Temp. sensors, 8. Above bed, 9. Free board, 10. Mid free board, 11. Top free board, 12. Heat exchanger, 13. Primary air fan, 14. Primary air line, 15. Gas line, 16. Lab view, 17. SO2 analyzer, 18. NOx, CO, CO2 analyzer.

Figure 3. Process flow diagram of lab scale and pilot scale testing.
operating at pre-set steady conditions. At specified feeding conditions of air, coal, and limestone, the steady state condition was evaluated by monitoring the temperature and the flue gas composition (in terms of O2 and CO2). The steady state condition was considered when the temperature and the flue gas composition were stable, and then a complete set of measurements was obtained.

Coals were sieved from 5 to 15 mm sizes and their properties are shown in Table 1. Limestone was used as an adsorbent and sieved to different size ranges (0.1–5.6 mm) and its properties are given in Table 2.

White wood was used as biomass for co-firing with coal. The results of proximate analysis, ultimate analysis, and semi-quantitative XRF analysis are given in Table 3.

Table 1. Results of GCV, proximate, and ultimate analyses of coal samples.

| Coal | Moisture ARB (wt%) | VM ADB (wt%) | Ash ADB (wt%) | N ARB (wt%) | C ARB (wt%) | H ARB (wt%) | S ARB (wt%) | GCV ADB (kcal/kg) |
|------|--------------------|--------------|---------------|-------------|-------------|-------------|-------------|------------------|
| A    | 5.46               | 35.5         | 34.71         | 1.17        | 44.00       | 3.46        | 7.23        | 4826             |
| B    | 7.27               | 33.32        | 34.04         | 1.25        | 45.82       | 3.41        | 6.75        | 4949             |

ARB: as received basis, ADB: air dry basis, VM: volatile matter, N: nitrogen, C: carbon, H: hydrogen, S: sulfur, GCV: gross calorific value.

Table 2. Chemical analysis of limestone.

| Parameters | SiO2 | Al2O3 | Fe2O3 | CaO | MgO | K2O | Na2O | SO3 | Cl |
|------------|------|-------|-------|-----|-----|-----|------|-----|----|
| Analysis results (%) | 4.28 | 0.53  | 0.39  | 51.14 | 1.59 | 0.09 | 0.04 | 0.06 | 0.004 |

Table 3. Results of proximate, ultimate and XRF analysis of biomass.

| Proximate and ultimate analysis of biomass | Semi-quantitative XRF Results |
|------------------------------------------|-------------------------------|
| Constituents | wt% | Elements | % |
| N | 0.17 | SiO2 | 40 |
| C | 47.05 | K2O | 8.81 |
| H | 5.39 | P2O5 | 1.3 |
| S | <0.1 | MgO | 4.79 |
| O | 47.39 | CaO | 22.9 |
| Ash | 1.14 | Al2O3 | 8.46 |
| Moisture | 6.6 | Na2O | 2.2 |
| volatile matter (VM) | 76.32 | Br | 3.21 |
| Fixed carbon (FC) | 11.83 | Cl | 0.0912 |
| | | SO3 | 0.292 |
| | | Fe2O3 | 4.92 |
| | | MnO | 1.72 |
| | | ZnO | 0.174 |
| | | N2O | 0.0146 |
| | | TiO2 | 0.502 |

Results and Discussion

**Effect of limestone quantity on SO2** Figure 4 shows the influence of different quantities of limestone (added in the bed) in terms of Ca/S molar ratios (MRs) on SO2 emissions during combustion of coal A from the Trans Indus Range and coal B from the Salt Range.

Different values of MR were used depending on limestone particle size and operational parameters. It is clear from Figure 4 that with an increase in MR, SO2 emissions decrease significantly for both coal A and...
coal B, reaching up to 40% and 41% at MR = 2 and 49% and 52% at MR = 3, respectively. However, a maximum decrease of 72% for coal A and 78% for coal B (i.e. maximum desulfurization) was observed at MR = 3.5. With a further increase in MR to 4, sulfur capture level decreased to 69% for coal A and 70% for coal B. It shows that an increase in limestone quantity beyond MR = 3.5, is not feasible for SO2 retention, and thus 3.5 is the optimum value of MR to get the maximum reduction in SO2 in the case of both coals. It can also be observed from Figure 4 that at MR = 2, a significant reduction in SO2 emission occurred (40% for coal A and 41% for coal B). With the further addition of 50% in limestone quantity (at MR = 3), only 9% and 11% were added to initial SO2 reduction for coal A and coal B respectively. Although overall SO2 emissions are decreased with the increase in MR values but desulfurization efficiency decreased at higher MR values. Therefore, it is evident from the results that desulfurization efficiency strongly depends on the initial concentration of SO2 in the bed.

The above results are in accordance with the investigation of Bragança and Castellan17 which reveals that SO2 retention efficiencies are 48%, 60%, 68%, and 77% at MR values of 1, 2, 3, and 4 respectively during coal combustion in FBC. Their results also show that the amount of limestone utilization lies in the MR range from 1 to 4 to meet the environmental norms and further rise in MR value will not be economically feasible. According to the study of Tarelho et al.,18 the range of MR is from 1 to 5 for various developed/developing countries to meet the SO2 emission norms and high SO2 removal efficiencies can be attained at MR = 3.5.

Limestone reacts with SO2 in presence of O2 to form calcium sulfate (CaSO4).

A number of reactions are possible including the following (equations (4) and (5)):

\[
CaCO_3 + SO_2 + \frac{1}{2} O_2 \rightarrow CaSO_4 + CO_2 \Delta H = -303 \text{ kJ/mole} \tag{3}
\]

\[
CaO + SO_2 + \frac{1}{2} O_2 \rightarrow CaSO_4 \Delta H = -481 \text{ kJ/mole} \tag{4}
\]

Stoichiometric calculations show that 1 mol of limestone feed can reduce 1 mol of sulphur.19 The molar volume of CaSO4 is greater than that of either CaO or CaCO3 which leads to the plugging of pores and therefore, complete conversion of the adsorbent particle is impossible. Also, sulfation only proceeds at the outer surface of the CaO particle. The sulfation process continues until external pores are blocked significantly and an impenetrable CaSO4 shell is formed leaving a considerable amount of unreacted CaO core. The increase in SO2 emissions for both coals beyond MR of 3.5 value may be due to the equilibrium of sulfation and reduction of CaSO4.20 According to Cheng et al.21 operational parameters including MRs, furnace temperature, residence time, and SO2 partial pressure affect the sulfation reaction.

Furthermore, the lower SO2 emissions at all MR values in the case of coal B relative to its counterpart coal

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**Figure 4.** Comparison of different MRs and SO2 emissions for coal A and coal B.
A is attributed to its lower inherited sulphur as shown in Table 1.

**Effect of limestone on NOx** Figure 5 displays the effect of different MRs on NOx emissions for coal A and coal B. It is clear from the graph that there is an increase in NOx emission with the increase in MR from 2 to 3.5 for coal A and coal B and the maximum increase occurs at MR = 3.5. Further increase in the MR beyond the value of 3.5 results in the decrease of NOx emission. The results are in line with the study of Zhou et al. explaining that the NOx emission increases with the addition of limestone during coal combustion. Some previous studies also show that taking the coal combustion without limestone as a reference, the addition of limestone results in the increase of NOx emission. An increase in MR to control SO2 emissions has a prominent impact on NOx emissions. There are three major mechanisms that explain the reason of this increase in NOx emission during limestone addition: the catalytic oxidation of NH3 to NO by CaO surfaces, the catalytic oxidation of CO to CO2 over CaO, and the catalytic oxidation of HCN to NO over CaO. The reaction scheme summarizing the selectivity of CaO toward NH3, HCN, and CO is given below in equations (5)–(7).

\[
\begin{align*}
    NH_3 + O_2 \xrightarrow{CaO} NO + N_2 \\
    HCN + O_2 \xrightarrow{CaO} NO + N_2 \\
    NO + CO \xrightarrow{CaO} \frac{1}{2} N_2 + CO_2
\end{align*}
\]

The addition of limestone may result in the shift from the homogeneous HCN oxidation mechanism to the catalytic one, which results in higher selectivity for NO formation and a lower one for N2O.

A decrease in NOx emission beyond the MR = 3.5 at which maximum desulfurization occurred, is supported by the fact that catalytic conversion of NH3 to NO is affected by the desulfurization. Limestone (CaO based sorbent) is an active catalyst for the oxidation of NH3 to NO but pore plugging of limestone occurs at higher MRs when the maximum conversion of CaO to CaSO4 is reached, which reduces NO formation from NH3.

**Effect of limestone on CO**

Figure 6 shows the influence of MR on CO emissions. It is clear from the figure that CO emissions decrease with an increase in the value of MR up to 3, for both of the coals. As the MR is further increased to 3.5, CO emissions increase but decrease again as the MR is increased to 4. The maximum decrease in CO emissions is observed at MR = 3.

Decrease in CO emissions upto MR = 3, is in accordance with the study of Liu and Gibbs who investigated the effect of limestone addition on gaseous emissions in a CFB combustor and concluded that with an increase of MR, the concentration of CO decreases during coal combustion in CFB. Tarelho et al. also found that CO and N2O emissions decrease and NO emissions increase with the addition of limestone during coal combustion in FBC. But for higher MRs, CO emissions increase and maximum concentration of CO is observed at MR = 3.5 at which maximum desulfurization occurred. There is a significant decrease in CO emissions with further increase in MR beyond 3.5 for both coal A and coal B. Armesto et al. reported that CO emissions are related to the concentration of O2.
temperature, reaction time/phase, and mixing of fuel with air. Amand et al.\textsuperscript{30} studied the effect of lime used for desulfurization in 12 MW CFB research plant. Their results reveal that the influence of SO\textsubscript{2} on NO/ N\textsubscript{2}O chemistry is not dependent on the catalytic effect of the lime surface and CO acts as an important intermediary species in these reactions. They also concluded that increase in SO\textsubscript{2} results in a decrease of NO\textsubscript{x} accompanied by an increase in CO emissions. The relation between SO\textsubscript{2} and NO\textsubscript{x} has already been discussed and reasoned in the previous section 3.2 of this article. However, the increase of CO with a rise in SO\textsubscript{2} has not been fully understood and needs further investigation. Authors are of the view that the rise in CO emission at and beyond MR = 3.5, might be due to the fact that as NO formation from NH\textsubscript{3} reduces at higher MRs and the rate of catalytic reduction of NO by CO also decreases thereby reducing the consumption of CO. This may result in an increase of CO emissions at higher MRs, however, this phenomenon needs further investigation.

**Effect of limestone on CO\textsubscript{2}**

Figure 7 shows the influence of limestone in terms of MR on CO\textsubscript{2} emissions. It is evident from the Figure 7 that the CO\textsubscript{2} emissions increased at MRs 2, 3 and decreased for higher MRs for coal A. Similarly, CO\textsubscript{2} increased at MRs 2, 3, and 3.5 and decreased for higher MRs for coal B. At high temperatures, CO\textsubscript{2} can be separated by the calcium oxide (sorbent) and high-temperature CO\textsubscript{2} capture systems are based on carbonation reaction when coupled with a calcination step. Grasa and Abanades\textsuperscript{31} experimentally investigated the evolution with the cycling of the capture capacity of CaO. Stanmore and Gilot\textsuperscript{19} examined some aspects of using lime from limestone to sequester CO\textsubscript{2} from combustion systems.

**Effect of bed temperature on SO\textsubscript{2}**

Figure 8 displays the effect of bed temperature on SO\textsubscript{2} emissions for coal B. This coal has been selected here owing to its better reduction of SO\textsubscript{2} emissions as demonstrated in the previous case. In this case, MR of 3.5 is kept which has proved to be optimum value for maximum desulfurization of both coals from the Salt Range and Trans Indus Range. It is clear from the figure that there is a slight increase in SO\textsubscript{2} emission against the bed temperature range from 700\textdegree C to 750\textdegree C at the beginning of the combustion but there is a decrease in SO\textsubscript{2} emission for the bed temperature window of 750\textdegree C–800\textdegree C. For bed temperature beyond 800\textdegree C, SO\textsubscript{2} emissions increase significantly revealing that further increase in temperature is not prone to control of SO\textsubscript{2} emissions. Therefore, the bed temperate of 800\textdegree C is the optimum temperature for maximum desulfurization of the coal under study at MR = 3.5. This behavior is also in accordance with previous research findings that the efficiency of SO\textsubscript{2} removal by limestone appears to decrease with an increase in the bed temperature.\textsuperscript{18} According to Bragança and Castell,\textsuperscript{17} the optimum bed temperature is 850\textdegree C for maximum desulfurization, while Tarelho et al.\textsuperscript{18} conducted their tests in FBC pilot plant facility and investigated that SO\textsubscript{2} removal efficiency decreases with the increase in
bed temperature ranging from 825°C to 900°C, and thereby a maximum rise in SO₂ removal efficiency occurred around 825°C which has been reported as optimum temperature. Anthony and Granatstein⁰² are of the view that there is still no consensus over the explanation of maximum temperature in FBC boiler for optimum sulfur retention, and its value depends on the operational parameters, the adsorbent used for desulfurization and types of coal, etc. The seize in SO₂ reduction beyond 800°C in the current study may be attributed either to one or more possible reasons including physical properties of limestone such as solid sintering and choking of porous structure, reductive decomposition of CaSO₄, or a net balance between limestone sulfation and CaSO₄ decomposition, at high bed temperatures.¹⁷

**Effect of limestone particle size on SO₂**

Figure 9 shows the influence of the particle size of limestone on sulfur reduction efficiency. It is evident from the figure that the reduction efficiency is higher for fine particle sizes ranging 0.1–2.0 mm and 2.0–2.8 mm relative to coarse particle size ranging 2.8–4.0 mm and 4.0–5.6 mm, at the same MR of 3.5. Fine particle-size ranges have a higher surface area for reaction than coarse particle-size ranges, as the surface area is the most important factor for better performance of finer
particle sizes for SO2 retention. Maximum sulfur retention efficiency of 72% is achieved at limestone particle-size range of 2.0–2.8 mm instead of finer particle-size range of 0.1–2.0 mm. The lower retention efficiency of finer limestone is accounted for the excessive elutriation occurring at this particle size which is a potential limitation for the investigation of finer sorbents. The particle size of the ground limestone is inversely related to its specific surface area; decreasing particle size increases the available surface area thereby improving the reactivity of the limestone. However, if the fragments generated from the attrition are too fine, unreacted sorbent would elutriate from the reactor, thus reducing the overall performance of the sorbent. Bragancxa and Castell17 investigated two different dolomite particle sizes of 0.47 and 0.32 mm during desulfurization of coal with high oxidizing conditions in FBC pilot-scale facility at bed temperature = 850°C. According to their results, finer particle size achieved 60% while the coarser gained only 30% SO2 retention efficiency at same MR.

**Effect of co-firing on SO2**

Figure 10 shows the influence of different quantities of biomass (added in to the bed) on SO2 emissions during combustion of coal A and coal B. It is clear from the graph that with the increase in biomass percentage from 30% to 60%, SO2 emissions decrease from 47.5% to 68.3% for coal A, and 36.3% to 67.8% for coal B. There is no considerable difference in SO2 emissions at 40% and 50% biomass for coal A but for coal B, SO2 emissions are 58% and 53% respectively at 40% and 50% biomass. A decrease in SO2 emissions at 50% biomass in comparison to 40% for coal B could be due to an error in measurement during experimental work. A maximum decrease of 68.3% in SO2 emissions for coal A and 67.8% for coal B achieved at 60% biomass fractions. This is in accordance with the study of Narayanan and Natarajan,32 they got 50% reduction in SO2 with the addition of 60% biomass. During their study, it was also found that the decrease in SO2 emissions remained upto 16% and 36% with cofiring of 20% and 40% biomass, respectively. Study of Demirbas33 also showed a decrease in SO2 with an increase in the proportion of the biomass.

Hein and Bemtgen34 explained the results of 2-years research project launched by the European Commission in which the co-combustion, in laboratory, pilot and full-scale units was studied with the objective to investigate the effect of biomass addition on the gaseous emissions. They confirmed that co-combustion of biomass with coal has a significant effect on SO2 reductions reaching up to 75%. The reduction in SO2 with the increase in biomass is ascribed to the dilution effect, as the biomass contains a negligible amount of sulfur. CaO and K2O present in the biomass, have the ability to capture SO2 by forming CaSO4 and K2SO4 in the presence of O2 just like SO2 capture using limestone.35

**Conclusion**

In the current study, the potential of limestone and biomass for the reduction in gaseous emissions like SO2, NOx, CO, and CO2 has been demonstrated on two different coal samples sourced from Pakistan using a pilot-scale FBC facility. Using coal as the primary fuel, limestone and biomass were mixed in different proportions to investigate the impact on desulfurization during combustion. The SO2 reduction was influenced by fuel properties, firing methodology (pure coal/cofiring with biomass), different values of MR, fine/coarse particle sizes of limestone, and the bed temperature. It was observed that maximum desulfurization of coal A and
coal B remained 72% and 78% respectively at the optimum MR value of 3.5. There was a predominant impact of bed temperature in the window of 750°C–800°C on SO₂ reduction, revealing the maximum reduction of 70% at 800°C with the same MR of 3.5.

Moreover, the effect of limestone particle size was studied on SO₂ emission which exhibited that maximum desulfurization of 72% occurred at a fine particle size range of 2.0–2.8 mm, relative to finer and coarse mode particles owing to excessive elutriation and small surface area in the former and latter cases, respectively. Also, there was a significant decrease in SO₂ emissions with the increase in the percentage of biomass (30%–60%) during co-firing with coal. Consequently, a remarkable decrease of 68% in SO₂ was noted with 60% biomass relative to other proportions.

This study for control of the gaseous emissions from combustion of high sulfur Pakistani coal in FBC facility has great potential for new coal based power projects, especially under China Pakistan Economic Corridor (CPEC) program. The study would also provide useful knowledge to the international scientific and engineering community for optimization, design, and development of thermal power plants based on high sulfur coal.

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Author contributions

All the authors actively participate in the research work. Muhammad Mahmood Aslam Bhutta conceived and designed the analysis, contributed data, or analysis tools. Shaqiq ur Rehman collected the data, contributed data or analysis tools, performed the analysis, and wrote the paper. Asad Naeem Shah conceived and designed the analysis. Muhammad Sajid Kamran contributed data or analysis tools. Ahmad Naveed contributed data or analysis tools. Syed Muhammad Sohail Rehman contributed data or analysis tools, wrote the paper, redraft the paper layout.

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Ethical approval/patient consent

Research involves no ethical concerns. However, all applicable international, national, and/or institutional guidelines for conduction of research followed. No harm to nature or its habitants is caused.

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