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### UDC 004.02 RESEARCH ON METHODS TO REDUCE HARMFUL EMISSIONS DURING THE COMBUSTION OF SOLID MUNICIPAL WASTE IN CIRCULATING FLUIDIZED BED FURNACES дослідження методів зниження шкідливих викидів при спалюванні твердих побутових відходів у топках з циркулюючим киплячим шаром

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Abstract. Combustion in circulating fluidized bed (CFB) furnaces is one of the most advanced technologies for the thermal utilization of organic solid fuels, ensuring high combustion efficiency while adhering to environmental standards. It has been established that the typical nitrogen oxide  $(NO_x)$  emissions during solid fuel combustion in CFB boilers range from 100 to 200 mg/m<sup>3</sup> under standard conditions, with oxygen content in the flue gases of 6%. For fuels with high volatile content, NOx concentrations can rise to 250 mg/m<sup>3</sup>. Various methods to reduce harmful compound formation are discussed, with a key advantage being the effective removal of sulfur dioxide (SO<sub>2</sub>) through the introduction of limestone into the bed. The ability to achieve efficient thermal destruction of different fuels in CFB boilers also enables their use for co-firing biomass and fossil fuels.

*Keywords:* combustion; circulating fluidized bed; harmful emissions; flue gases; solid municipal waste.

#### Introduction.

With increasing demands for environmental protection, particularly for reducing harmful gas emissions such as sulfur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>), one of the promising solutions for the thermal destruction of organic solid fuels is combustion in circulating fluidized bed (CFB) furnaces. Emissions from the combustion of solid municipal waste (SMW) are influenced not only by the fuel properties but also by operational parameters, such as excess air ratio, combustion temperature, mixing quality, and residence time.

#### Main text

In the field of optimizing combustion methods, numerous studies have been conducted. For example, it has been found that during combustion in a CFB furnace, the share of nitrogen converted to  $NO_x$  significantly decreases with an increase in the nitrogen content of the combustible material (Figure 1) [1].



Figure 1: Dependence of nitrogen conversion to NO<sub>x</sub> on the nitrogen content (CN) in the combustible mass at a bed temperature of 800 °C (5, 6) and 850 °C (7), with oxygen concentrations of 10% by volume (5, 7) and 8% by volume (6). 1 - Grate furnace; 2 - Fuels of various types (Skreiberg, 1996); 3 - Fluidized bed; 4 - Furnace (Hofbauer, 1994)

It has also been observed that the molar ratio of calcium to sulfur (Ca/S) affects  $NO_x$  emissions. When the Ca/S ratio increases from 1.5 to 4.0,  $NO_x$  emissions rise from 150 to 250 mg/m<sup>3</sup>. This process is complex and not yet fully understood. During volatile release, the parent nitrogen in the fuel transitions to the gas phase as  $NH_3$  and HCN. In the presence of CaO and coke, these compounds partially convert to NO through the intermediate formation of N<sub>2</sub>O, and partially reduce to N<sub>2</sub>. As the bed height increases, the concentration of N<sub>2</sub>O decreases, while the  $NO_x$  concentration rises. Additionally, an increase in excess air leads to a consistent rise in  $NO_x$  concentration, normalized to 6% oxygen. However, studies [2] have noted a decrease in N<sub>2</sub>O concentrations with increasing excess air.

N<sub>2</sub>O emissions from CFB boilers do not exceed 100 mg/m<sup>3</sup> and decrease with rising temperatures. Nitrous oxide is 200 times more harmful as a greenhouse gas compared to CO<sub>2</sub>, but its concentration in CFB boilers is 10000 times lower than CO<sub>2</sub>. Consequently, N<sub>2</sub>O contributes minimally compared to other greenhouse gases, and interest in detailed studies on N<sub>2</sub>O formation and suppression is relatively low.

A key advantage of CFB combustion is the effective removal of SO<sub>2</sub> by introducing limestone into the bed. This process involves the following reactions:

Calcination:  $CaCO_3 = CaO + CO_2$ 

Sulfation:  $CaO + SO_2 = CaSO_3$ 

Oxidation:  $CaSO_3 + \frac{1}{2}O_2 = CaSO_4$ 

The conditions in CFB boiler furnaces are favorable for efficient SO<sub>2</sub> adsorption by the alkaline components of ash. A detailed analysis of the mechanism of sulfur binding by limestone is presented in [3]. Calcium sulfate forms a molten crust on the surface of calcium oxide, which significantly reduces the continuation of the reaction. As a result, the utilization of CaO is typically limited to about 40%. Optimal furnace temperatures for this process are near 850 °C. Increasing the Ca/S ratio beyond 2.5 has little effect on further reducing SO<sub>2</sub> emissions .

Figure 2 shows the dependence of sulfur binding efficiency on the Ca/S ratio. In conditions without limestone addition, the Ca/S ratio is at the level of 0.2–0.3, and the sulfur binding efficiency is 4–9%. The highest sulfur binding efficiency is observed in modes with reduced loads and bed temperatures [4].



Figure 2: Dependence of sulfur binding efficiency on the Ca/S ratio, recorded in April 2018 (1) and November 2017 (2) [4].

Reducing sulfur oxide (SO<sub>2</sub>) emissions is achieved by improving the quality of bed material, with particle sizes of fly ash smaller than 0.05 mm and bottom ash smaller than 0.2 mm. Limestone particles should also be finer to increase the reaction surface area. This improves cyclone capture efficiency, leading to longer particle residence time in the furnace and greater limestone utilization efficiency. Since 2015, certain regions in China have implemented requirements for new power plants to ensure that harmful emissions from coal-fired boilers are at the same level as those from natural gas-fired systems. The permissible emission levels are as follows (in mg/m<sup>3</sup>): nitrogen oxides (NO<sub>x</sub>) – 50, sulfur oxides (SO<sub>2</sub>) – 35, and particulate matter – 5. These standards have already been implemented in one of the CFB boilers in China [5].

Analysis of existing data shows that the desulfurization process using limestone is most effective (minimum observed SO<sub>2</sub> concentrations in flue gases) at bed temperatures of 840–870°C. These temperatures align with the operating temperatures of most CFB furnaces, ensuring the required efficiency of solid fuel combustion and slag-free bed operation. As the temperature of the reacting limestone closely matches the bed temperature, the optimal temperature for SO<sub>2</sub> capture with limestone is 840–870°C. Since the sulfur dioxide binding process using active ash sorbents occurs on the surface or inside the burning coal particle, the temperature level for SO<sub>2</sub> binding differs significantly from the overall bed temperature. It is known that the temperature of a burning coal particle in the bed exceeds the observed bed temperature by several degrees to several tens or even hundreds of degrees, depending on both the combustion conditions of the coal particle and the type of fuel being burned.

Thus, during the combustion of high-ash Ekibastuz coal, for example [6], the optimal desulfurization temperature should differ significantly from the optimal temperature for SO<sub>2</sub> capture using limestone, shifting toward a lower value.

Figure 3 shows the dependence of SO<sub>2</sub> capture efficiency during the combustion of Ekibastuz coal with varying ash content on bed temperature. As seen in the figure, the maximum SO<sub>2</sub> capture efficiency ( $\eta$ SO<sub>2</sub>) by coal ash with ash content of 37.9% and 41.7% is observed at a temperature of 750–770°C. For higher-ash coal ( $A_r = 61.8\%$ ), the optimal temperature is less distinct, and for coal material with  $A_r = 61.8\%$ , no clear optimum is observed, as  $\eta$ SO<sub>2</sub> increases with decreasing bed temperature. At a bed temperature of approximately 800°C, the SO<sub>2</sub> capture efficiency, depending on coal ash content, ranges from 60% to 90%. Notably, the highest SO<sub>2</sub> capture efficiency is observed for coal material with an ash content of 68.2%.



Figure 3. Dependence of SO<sub>2</sub> capture efficiency by Ekibastuz coal ash on bed temperature without limestone addition [6]

As shown in Figure 3, the optimal temperature during the combustion of pure coal with a molar ratio of Ca/S = 0.88 was 750–770°C, with a maximum SO<sub>2</sub> capture efficiency ( $\eta$ SO<sub>2</sub>) of 65%. When the same coal was mixed with 5% limestone (Ca/S = 3.63),  $\eta$ SO<sub>2</sub> increased to 85%, and the optimal temperature rose to 825–830°C. Increasing the limestone content in the mixture to 10% (Ca/S = 6.65) resulted in a slight improvement in  $\eta$ SO<sub>2</sub> to 87%, maintaining the same optimal temperature of 825–830°C.

In [1], the issue of HCl formation, which promotes corrosion of superheaters and deposit formation, is discussed in detail. Chlorine in the fuel primarily oxidizes to HCl, but other significant factors can lead to the formation of molecular chlorine  $(Cl_2)$ , including reactions with aluminosilicates. Figure 4 illustrates the possible interactions of sulfur with chlorines, heavy metals, and alkaline elements, as reported

# in [1, 7].

Chlorines, especially Cl<sub>2</sub>, can contribute to the formation of dioxins and furans. Copper and iron may act as catalysts in furan formation. In CFB boilers, these emissions are significantly reduced even at relatively low combustion temperatures, provided the residence time in the reaction zone exceeds 3 seconds at temperatures above 800°C. During the combustion of municipal waste in CFB boilers, the addition of coal has been shown to drastically reduce dioxin and furan concentrations.



Figure 4. Possible interactions of sulfur with chlorines, heavy metals, and alkaline elements [7]

In [8], the impact of primary air supply conditions on nitrogen oxide (NO<sub>x</sub>) emissions was studied. Experiments were conducted on a 235 MW boiler at the Turow power plant in Poland. Modeling of primary air distribution revealed uneven distribution across the bed surface. Achieving greater uniformity could theoretically be accomplished by increasing the resistance of the air distribution grid. However, this solution was deemed impractical as it would increase the energy consumption of the power unit. Instead, modifications were made to the design of the primary air distribution chamber. Profiles were installed at the chamber inlet, dividing the airflow into three zones with equal distribution. The results showed that temperature distribution in the furnace became more uniform, bed temperatures decreased by approximately  $50^{\circ}$ C, and NO<sub>x</sub> emissions were reduced by an average of 10%.

# Summary and conclusions.

For CFB boilers, typical NO<sub>x</sub> concentrations during the combustion of solid fuels range from 100 to 200 mg/m<sup>3</sup> under standard conditions with 6% oxygen content in flue gases. For fuels with very high volatile content, NO<sub>x</sub> concentrations can increase to 250 mg/m<sup>3</sup>.

One of the main advantages of fluidized bed combustion is the effective removal of sulfur dioxide (SO<sub>2</sub>) by introducing limestone into the bed. However, calcium sulfate forms a molten crust on the surface of calcium oxide, significantly reducing the continuation of the reaction. As a result, CaO utilization is limited to approximately 40% of its content in limestone. The optimal furnace temperature for this process is approximately 850°C. Increasing the molar ratio of Ca/S by 2.5 times has little effect on further reducing SO<sub>2</sub> emissions.

The ability of CFB boilers to efficiently combust various fuels makes them suitable for co-firing biomass and fossil fuels. This flexibility can be a decisive factor for many European countries, considering biomass is carbon-neutral. Co-firing up to 10% biomass by heat input can reduce CO<sub>2</sub> emissions by 18–22%.

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Анотація. Спалювання в топках з циркулюючим киплячим шаром (ЦКШ), є однією з передових технологій термічної утилізації органічного твердого палива, яка забезпечує високу ефективність згоряння при дотриманні екологічних показників. Виявлено, що типовими для котлів з ЦКШ під час спалювання твердого палива є концентрації NOx у викидах, що дорівнюють 100-200 мг/м3 за нормальних умов і вміст кисню в димових газах, 6%. Для палив з дуже великим виходом летких можливе збільшення концентрації NOx до 250 мг/м3. Розглянуто методи, що сприяють зниженню утворення шкідливих сполук, де однією з основних переваг є можливість ефективного видалення діоксиду сірки SO2 шляхом подачі шару вапняку. Можливість ефективної термічної деструкції різних палив у котлах з ЦКШ дозволяє використовувати їх для спільного спалювання біомаси та викопного палива. **Ключові слова:** спалювання; циркулюючий киплячий шар; шкідливі викиди; димові гази; тверді побутові відходи.

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