Model research on NO$_X$ emission of an ultra-supercritical circulating fluidized bed boiler

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Abstract. It requires that coal-fired generator units to meet ultra-low emission standards by 2020 in policy of environmental protection in China. Compared with pulverized coal boiler, circulating fluidized bed (CFB) boiler technology has the natural advantage of low NO$_X$ emission, which has the ability to directly realize ultra-low NO$_X$ emission inside the furnace. To accurately obtain NO$_X$ emission characteristics in an ultra-supercritical CFB boiler under construction, model research and experimental verification of NO$_X$ emission prediction were carried out in this paper. The results indicate that three models could all predict NO$_X$ emission in the CFB boiler, and the deviation among the predicted values calculated by these models can be controlled within 15%. When an ultra-supercritical CFB boiler burns existing engineering coal, NO$_X$ emission would be higher than 50 mg/Nm$^3$, which requires structural improvement and operation parameters optimization to ensure that NO$_X$ emission inside the furnace of the CFB boiler could directly reach the ultra-low emission standard.

1. Introduction
Circulating fluidized bed (CFB) boiler technology, which has the outstanding advantages of wide coal adaptability and low cost pollutant control inside the furnace, is recognized as one of the clean coal power generation technologies in commission [1]. In recent years, the CFB boiler technology has developed rapidly in China, and begins to enter ultra-supercritical era [2]. However, it is also facing an unprecedented new situation of environmental protection for CFB units, requiring coal-fired units to achieve ultra-low emissions standard by 2020. Generally, NO$_X$ emission in a new CFB unit is obtained by trial burning in the real boilers or model prediction [3]. Model prediction is of great significance for coal characteristics adjustment and operation parameter optimization.

The study on NO$_X$ emission characteristics in CFB boilers mostly focuses on the mechanism of NO$_X$ generation and reduction. However, mechanism models are always complex [4-11]. Because prediction accuracy of mechanism models is affected by selection of boundary conditions and operation parameters, so it is difficult to be directly applied in the real CFB boilers. On the basis of a large number of operation and test data from the real boilers, some scholars have obtained some empirical formula to predict NO$_X$ emission in the CFB boiler. There are three typical empirical models. The first one [12] was a semi-empirical formula to obtain NO$_X$ emission in the boiler, based on the inherent characteristics of coal and fuel-N conversion rate inside the furnace. The second one [13] and the third one [14] are empirical formulas to obtain NO$_X$ emission characteristics, based on the inherent characteristics of the coal and operation parameters of a large number of actual CFB boilers. In
especial, it considered the effect of bed temperature and excess air coefficient on NO\textsubscript{X} emission more carefully in the third model.

On this basis, in order to obtain NO\textsubscript{X} emission characteristics of an ultra-supercritical CFB boiler, model research on an ultra-supercritical boiler burning engineering coal was carried out in this paper. First of all, three models were used to predict NO\textsubscript{X} emission of a 300MW CFB boiler burning two different kinds of coals, and then predicted values calculated by models were compared with measured values. At last, NO\textsubscript{X} emission of the 660MW ultra-supercritical CFB boiler was predicted, and design optimization of operation parameters such as bed temperature and excess air coefficient etc. were proposed. Further optimization suggestions of the boiler were put forward to realize NO\textsubscript{X} ultra-low emission directly inside the furnace.

2. Model

2.1. Model 1

In the design stage of the CFB boiler, based on coal characteristics and fuel-N conversion of different coals, NO\textsubscript{X} emission concentration could be reasonably predicted in Model 1 [12], but it could not accurately predict the condition deviated from design point. The formula for Model 1 is as follows:

\[
NO_x = NO_{x,pot} \times \frac{C_R}{100} \times \frac{N}{V_{DFG}} \times 10^4 \times \frac{C_R}{100}
\]  

where, \( NO_x \) is the NO\textsubscript{X} emission value, mg/Nm\textsuperscript{3} (6\%O\textsubscript{2}, dry). \( NO_{x,pot} \) is theoretical conversion of fuel-N to NO\textsubscript{X}, mg/Nm\textsuperscript{3}. \( C_R \) is fuel-N conversion rate, %. \( N \) is the coal received base nitrogen, %. \( V_{DFG} \) is the theoretical dry flue gas volume, seen in Formula (2), Nm\textsuperscript{3}/kg.

\[
V_{DFG} = 0.08955C + 0.2115H + 0.03343S + 0.00800N - 0.02665O
\]  

Where, \( C \) is coal received base carbon, %. \( H \) is coal received base hydrogen, %. \( O \) is coal received base oxygen, %. \( N \) is coal received base nitrogen, %. \( S \) is coal received base sulfur, %.

Fuel-N conversion rate \( C_R \) recommended values is as shown in Table 1.

| Coal type              | Unit | Recommend value |
|------------------------|------|-----------------|
| Lignite                | %    | 5.0             |
| Sub-bituminous        | %    | 4.0             |
| High volatile bituminous | %    | 6.0             |
| Middle volatile bituminous | %    | 4.0             |
| Low volatile bituminous | %    | 3.0             |
| Anthracite             | %    | 2.0             |

2.2. Model 2

Based on coal inherent characteristics and field test operation parameters of the actual CFB boilers, Model 2 [13] was proposed. However, due to the strong experience of the data from the boiler, it was difficult to predict NO\textsubscript{X} emission deviated from design condition. In Model 2, there is an empirical formula to obtain NO\textsubscript{X} emission directly, which is as follows:

\[
NO_x = 8909N_{zs} \left(1 - \frac{\eta_{self}}{100}\right)
\]  

where, \( N_{zs} \) is the coal conversion nitrogen content, g/MJ. \( \eta_{self} \) is the self-denitration efficiency of the boiler, %.

Table 2 shows empirical data on self-denitration efficiency of real CFB boilers. In Table 2, it can be seen that when the coal conversion nitrogen content is between 0.2g/MJ and 0.4g/MJ, the self-denitration efficiency of the boiler is chosen as 96%. When the coal conversion nitrogen content is
between 0.4 g/MJ and 0.6 g/MJ, it is chosen as 97%. When the coal conversion nitrogen content is between 0.6 g/MJ and 0.8 g/MJ, it is chosen as 98%.

2.3. Model 3
A number of variables obtained from field test showed that NO\textsubscript{X} emission was highly related to coal type, especially volatile content, bed temperature and excess air coefficient. Model 3 was established by introducing the parameters mentioned above [14]. The correlation could eliminate the effect of boiler capacity and make a good prediction. The formula for Model 3 is as follows:

\[
NO_{X} = \frac{38174}{V_{g}^{0.2441}} V_{\text{daf}}^{0.8375} \alpha^{2.0641} \exp\left(-\frac{3215.4}{T_{b}}\right)
\]  

(4)

Where, The \(V_{g}^{0}\) is theoretical dry flue gas volume, see Formula (5), mg/Nm\textsuperscript{3}. \(V_{\text{daf}}\) is coal drying ash-free volatile matter, %. \(\alpha\) is boiler excess air coefficient. \(T_{b}\) is bed temperature in dense phase zone, K.

\[
V_{g}^{0} = 1.866 \frac{C}{100} + 0.7 \frac{S}{100} + 0.8 \frac{N}{100} + (1.4 - 0.21)V_{\text{air}}^{0}
\]  

(5)

The \(V_{\text{air}}^{0}\) in the formula is theoretical air volume, see Formula (6), mg/Nm\textsuperscript{3}.

\[
V_{\text{air}}^{0} = \frac{1}{0.21} \left(1.866 \frac{C}{100} + 5.85 \frac{H}{100} + 0.7 \frac{S}{100} - 0.7 \frac{O}{100}\right)
\]  

(6)

2.4. Comparative analysis of the empirical models
When three models are all used to predict NO\textsubscript{X} emission in the boiler, because fuel-N conversion rate and self-denitration efficiency of the boiler in Models 1 and 2 are both selected according to the coal characteristics, so the predicted values represents the best value of the theory, while Model 3 fully considers the influence factors such as coal volatile matter, bed temperature and excess air coefficient on NO\textsubscript{X} emission, so the predicted NO\textsubscript{X} emission was closer to the measured value from the real boiler.

3. Results and discussions

3.1. Model verification
To verify the prediction accuracy of the three models, field measurements were carried out on a 300 MW CFB boiler at full boiler load. The boiler, designed with a 1025 t/h CFB steam generator, had
been in commercial operation since 2011. At that time, it belongs to the first batch of 300MW self-developed CFB boilers in China. The boiler is consisted by a pant-leg furnace, four hot cyclones and four loop seals. Tables 3 and 4 give coal characteristics and operation parameters during field test, respectively.

Table 3. Coal characteristics of a 300MW CFB boiler.

| Item       | Unit | Coal A   | Coal B   | Item       | Unit | Coal A   | Coal B   |
|------------|------|----------|----------|------------|------|----------|----------|
| $M_{ar}$   | %    | 25.10    | 26.60    | $C_{ar}$   | %    | 53.31    | 50.88    |
| $A_{daf}$  | %    | 6.92     | 8.13     | $H_{ar}$   | %    | 2.40     | 2.25     |
| $V_{daf}$  | %    | 32.75    | 33.00    | $O_{ar}$   | %    | 11.26    | 11.22    |
| $N_{ar}$   | %    | 5.55     | 0.55     | $S_{ar}$   | %    | 0.46     | 0.45     |
| $Q_{ar,net}$ | MJ kg$^{-1}$ | 18.87 | 21.62 | | |

Table 4. Main operation parameters of the boiler at full boiler load.

| Item                      | Unit      | Coal A   | Coal B   |
|---------------------------|-----------|----------|----------|
| Main steam flow           | t·h$^{-1}$ | 1004     | 1025     |
| Main steam pressure       | MPa       | 17.1     | 16.9     |
| Main steam temperature    | °C        | 536      | 536      |
| Reheater steam pressure   | MPa       | 3.5      | 3.4      |
| Reheater steam temperature| °C        | 536      | 537      |
| Coal mass flow            | t·h$^{-1}$ | 230      | 206      |
| Pressure drop inside plenum| kPa    | 14.2     | 15.2     |
| Total air volume flow     | Nm$^3$/h | 862000   | 891000   |
| Feed water flow           | t·h$^{-1}$ | 279      | 280      |
| Bed temperature           | °C        | 940      | 930      |
| Excess air coefficient    |           | 1.14     | 1.18     |

Table 5 gives the boundary conditions which are needed by model calculation. Parameters such as coal rank, Fuel-N conversion rate, self-denitration efficiency are chosen based on coal characteristics.

Table 5. Boundary conditions of model calculation.

| Item                      | Unit  | Coal A         | Coal B         |
|---------------------------|-------|----------------|----------------|
| Coal rank                 |       | Sub-bituminous | Sub-bituminous |
| Fuel-N conversion rate    |       | 0.4            | 0.4            |
| Convert nitrogen          | %     | 0.29           | 0.22           |
| Self-denitration efficiency| %   | 96             | 96             |
| Bed temperature           | °C    | 940            | 930            |
| Excess air coefficient    |       | 1.14           | 1.18           |

Figure 1 represents a comparison of NO$_X$ emission calculated by the three models and measured values in a 300MW CFB boiler. It can be seen from the figure that compared with the measured value, the calculation results of the three models are generally smaller. The NO$_X$ prediction values calculated by Models 1 and 2 are relatively closer, but the deviation from the measured values is larger, while the predicted values calculated by Model 3 are closer to the measured value. The reason may be that there are non-uniform combustion and the catalysis of limestone desulfurizer inside the furnace under the real condition which made NO$_X$ emission be higher than that under design condition in Models 1 and 2. Thus, Models 1 and 2 can be used to predict NO$_X$ emission of the CFB boiler under design condition, while Model 3 can be used to predict that of actual boiler. When the operation parameters such as bed temperature, excess air coefficient etc. are adjusted to the design values, the predicted NO$_X$ emission
of the three models will be almost the same, providing an important improvement direction for the structure and operation optimization of the CFB boiler.

In addition, it can also be seen from Figure 1 that deviations of the models are not the same under the two conditions in the boiler. The reason is that there are obvious change of coal characteristics and operation parameters under the two conditions, which has a great influence on the predicted NO\textsubscript{X} emission calculated by the models.

![Figure 1. A comparison of NO\textsubscript{X} emission calculated by the three models and measured values in a 300MW CFB boiler.](image)

### 3.2. NO\textsubscript{X} emission prediction of an ultra-supercritical CFB boiler

According to the coal characteristics and design operation parameters of a 660MW ultra-supercritical CFB boiler under construction, NO\textsubscript{X} emission of the boiler will be predicted. The boiler is consisted by a furnace, four steam cooled cyclones and external heat exchangers. Tables 6 and 7 show coal characteristics and operation parameters of the CFB boiler.

**Table 6.** Coal characteristics of the 660MW ultra-supercritical CFB boilers.

| Item     | Unit | Value | Item     | Unit | Value |
|----------|------|-------|----------|------|-------|
| \(M_{ar}\) | %    | 24.48 | \(C_{ar}\) | %    | 38.67 |
| \(A_{ar}\) | %    | 31.03 | \(H_{ar}\) | %    | 2.14  |
| \(V_{daf}\) | %    | 36.86 | \(O_{ar}\) | %    | 2.18  |
| \(N_{ar}\) | %    | 0.58  | \(Q_{ar,net}\) | MJ kg\(^{-1}\) | 14.35 |

**Table 7.** Main operation parameters of the boiler at full boiler load.

| Item                      | Unit    | Value |
|---------------------------|---------|-------|
| Main steam flow           | t·h\(^{-1}\) | 1910  |
| Main steam pressure       | MPa     | 29.4  |
| Main steam temperature    | °C      | 605   |
| Reheater steam temperature| °C      | 623   |
| Feed water temperature    | °C      | 300   |
| Bed temperature           | °C      | 860   |
| Excess air coefficient    | /       | 1.15  |
In addition, boundary conditions needed by model calculation contain Fuel-N conversion rate (0.4) and self-denitration efficiency of the boiler (96%).

Figure 2. Predicted NO\textsubscript{X} emission of the ultra-supercritical CFB boiler.

Figure 2 shows the predicted NO\textsubscript{X} emission of the ultra-supercritical CFB boiler calculated by the three models. From Figure 2, it can be seen that NO\textsubscript{X} emission calculated by Models 1 and 2 would be from 139mg/Nm\textsuperscript{3} to 142mg/Nm\textsuperscript{3} under design condition, while that calculated by Model 3 is 159 mg/Nm\textsuperscript{3}, which is 12.8% higher than that under the design condition. Taking into account boiler combustion performance, the design parameters such as bed temperature and excess air coefficient etc. are further optimized, that is, bed temperature is 850°C, and excess air coefficient is 1.1, at this time NO\textsubscript{X} emission prediction calculated by Model 3 will be 138mg/Nm\textsuperscript{3}. Thus, predicted NO\textsubscript{X} emission calculated by the three models are nearly the same, indicating that operation inside the furnace has reached the best condition. However, NO\textsubscript{X} emission of this boiler could not directly meet the ultra-low emission standard inside the furnace, that is, NO\textsubscript{X} emission is higher than 50mg/Nm\textsuperscript{3}.

To make further NO\textsubscript{X} emission of the ultra-supercritical CFB boiler be as low as possible, or even less than 50mg/Nm\textsuperscript{3}, it should be under the strict control of bed temperature and excess air coefficient of boiler, and measures such as combustion uniformity design, flow regime reconstruction and structural optimization should also be adopted. Generally, the average particle size of the circulating ash in the boiler is from 150μm to 250μm [15]. According to the design theory of flow regime steady of the CFB boiler, NO\textsubscript{X} emission could be obviously reduced by controlling appropriate coal particle size, improving the efficiency of cyclone and raising the height of secondary air duct, which has been reported in CFB boilers burning similar coal at boiler capacity of 150t·h\textsuperscript{-1}, 260t·h\textsuperscript{-1} and 560t·h\textsuperscript{-1} [16, 17].

4. Conclusions

Three models were used to predict NO\textsubscript{X} emission of a 660MW ultra-supercritical CFB boiler burning engineering coal in this paper. The main findings include:

1) Three models are all feasible to predict NO\textsubscript{X} emission of the CFB boiler. Models 1 and 2 could be used to predict NO\textsubscript{X} emission under design condition, while Model 3 can be used to predict the actual NO\textsubscript{X} emission of the CFB boiler under different operation conditions.

2) The calculation results indicate that NO\textsubscript{X} emission of an ultra-supercritical CFB boiler is from 139mg/Nm\textsuperscript{3} to 142mg/Nm\textsuperscript{3} under design condition. According to real operation parameters such as bed temperature and excess air coefficient etc., NO\textsubscript{X} emission of the boiler will reach 159mg/Nm\textsuperscript{3}.

3) When operation parameters of 660MW ultra-supercritical CFB boiler such as bed temperature and excess air coefficient etc. are optimized to the best condition, the optimal NO\textsubscript{X} emission is still
138mg/Nm³, which can not reach directly ultra-low emission standard. So further important improvement direction would be combustion uniformity design, fluid regime reconstruction and structural optimization.

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