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CFD MODEL OF NO_X REDUCTION BY SNCR METHOD

CFD MODEL REDUKCE EMISÍ NO_x METODOU SNCR

Abstract

The paper deals with numerical simulation of SNCR method. For numerical modelling was used CFD code Ansys/CFX. SNCR method was described by dominant chemical reaction, which were look up NIST Chemical database. The reactions including reduction of NO_x and concentration change of pollutants, like N_2O and CO in flue gas too. Proposed chemical kinetics and CFD model was applied to two boilers. Both simulations were compared with experimental measurements. First simulation was used to validation of chemical mechanism. Second simulation was based on first simulation and it was used to verification of compiled SNCR chemical mechanism

Abstrakt

Článek se zabývá numerickým modelováním SNCR metody. K simulacím byl využit CFD program Ansys/CFX. SNCR metoda byla definována základními dominantními reakcemi, které byly vyhledány v mezinárodní chemické databázi NIST. Samotné reakce zahrnovaly nejen redukci NO_x, ale také změnu koncentrace polutantů N_2O a CO ve spalinách. Navržený chemický mechanizmus zahrnutý v CFD simulaci byl aplikován na dva kotle. První simulace sloužila k upřesnění fyzikálních konstant a validaci chemických reakcí. Druhá simulace již sloužila k verifikaci sestaveného reakčního schématu.

1 INTRODUCTION

Emission of NO_x can be reduced by two different approaches. These approaches are called primary and secondary caution. Primary caution is based on prevention of production of NO_x, e.g. low emission burner etc. Secondary caution is based on chemical destruction of NOx in flue gas. Chemical principle of secondary caution is reduction of NO_x by NH_2^- radical. Products of this reduction are molecular nitrogen N_2 and water H_2O. Several modification of basic chemical principle exists, and we will deal with method called NO_xOUT.

Basic principle of NO_x reduction by NH_2^- radical is known from 70 years of 20 century. Dominant chemical reaction is very simply and is defined by following chemical formula

$$NH_2^- + NO \rightarrow N_2 + H_2O \tag{1}$$

This reaction is dominantly occurred without catalyser in temperature window 850-1050°C, this reaction is called Selective Non-Catalytic Reduction (SNCR). Reaction is occurred with catalyser in temperature window 150-300°C too, this reaction is called Selective Catalytic Reduction (SCR).

Three materials (reagents) are source of NH2- radical. First material is Urea $(NH_2)_2CO$. Business name of SNCR method with Urea is NO_xOUT. Second material is Ammonia water NH_4OH. Business name of SNCR method with Ammonia water is DeNOx. Relatively new third material is Cyanouric acid $(HNCO)_3$. Business name of SNCR method with Cyanouric acid is RAPRENO$_x$. Reaction scheme, which is shown in Fig. 1, illustrates individual reaction and relation between several materials. This SNCR scheme is strongly simplified and about 150 reactions occur in reality.

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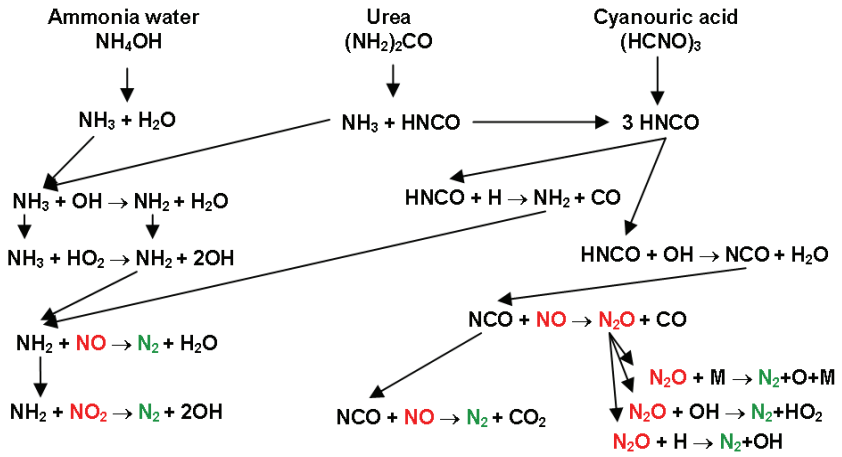

Fig. 1 Chemical mechanism of SNCR method

Main problem of SNCR method is relatively narrow temperature window (850-1050°C). If the reagent is injected into colder temperature zone of furnace, the nitrogen oxides NO_x do not react with NH_2^- radical, because the reaction rate is slow and high concentration of Ammonia HN_3 is in flue gas. If the reagent is injected into warmer temperature zone of furnace, the NH_2^- reacts with oxygen O_2. So that the nitrogen oxides are not reduced but they are produced by combustion of ammonia.

2 BOILERS GEOMETRY

SNCR model was tested on two similar boilers with SNCR technology. The boilers are granulation type and as a fuel is used brown coal (lignite). Complete geometry of the both combustion chambers were created pursuant to drawing documentation. Ansys DesignModeler was used to 3D CAD design. The convection domain of combustion chambers were only used to model of SNCR process, and zone of super heater and heat exchanger was separated from all geometry of boilers respectively, see Fig. 2. Domain inlet (1) is located over zone of flame. Domain includes OFA (Over Flame Air) jet (2) too. The main heat transfer surfaces are plate super-heater (3) and pipe heat exchangers (4). Last heat exchanger is ECO (economiser) (5), which is located in second draught of boiler. Domain outlet (6) is located behind ECO.

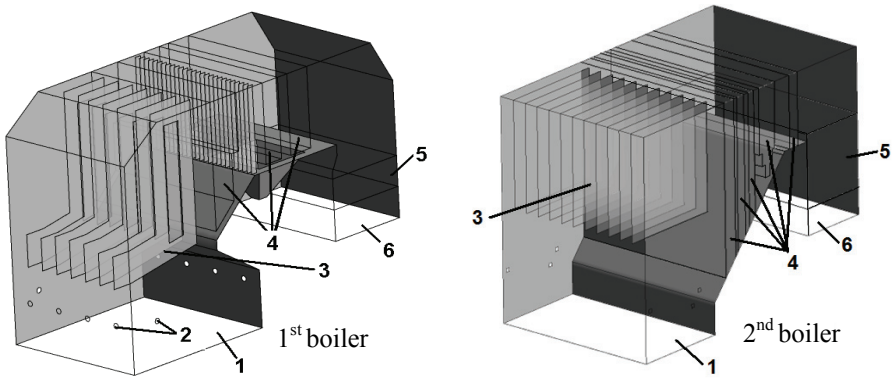

Fig. 2 Geometry of Boilers

This geometry was used to CFD simulation of SNCR process, because there are located the temperature Windows 850-1050°C and the reagent is injected into this zone of furnace. First geometry was used to validation of chemical mechanism. Second geometry was used to verification of compiled SNCR chemical mechanism.

3 CFD Model of SNCR and Chemical ReactionS

The basic equation set which describes the bought regimes of flow represents application of conservation law. . The law of mass, momentum and energy conservation is used to numerical modelling of flow. The law of momentum conservation represents Navier-Stokes equation. The Law of mass conservation represents continuity equation, and the law of energy equation represent energy equation.

Turbulent flow is stochastic, but it is statistic stable. Arbitrary physical quantity can be decompiled on sum of average value and fluctuation value $u = \bar{u} + u'$. Commonly used turbulent models are based on description of local turbulent intensity by length and velocity scale. Standard k – ε turbulent model was used to CFD simulation in this case [1]. The turbulent viscosity is specified by two transport equations. These equations solve turbulent kinetic energy k and eddy dissipation rate ε. [1],[2]

Chemical reactions included in CFD are based on Arrhenius equation, which describes reaction rate [2],[4].

$$k = A \cdot e^{\frac{-E}{RT}} \cdot T^{\beta}, \tag{2}$$

where:

k – reaction rate $\left[\frac{\left(\mathrm{mol}\cdot\mathrm{m}^{-3}\right)^{(1-n)}\cdot \mathrm{K}^{\beta}}{\mathrm{s}}\right]$,

A – pre-exponential factor $\left[\frac{\left(\mathrm{mol}\cdot\mathrm{m}^{-3}\right)^{(1-n)}}{\mathrm{s}}\right]$,

E – activation energy $[\mathrm{J}]$,

R – universal gas constant $\left[\frac{\mathrm{J}}{K\cdot mol}\right]$,

T – temperature $[\mathrm{K}]$,

β – temperature exponent $[-]$,

n – sum of the number of reaction orders $[-]$.

Arrhenius equation describes only rate of reaction. The chemic mechanism of every reaction must be specified by parent and children materials of reaction and stochiometric coefficients. The chemical mechanism for SNCR with Urea was specified by following reactions. The chemical mechanism includes basic components of NO_x pollutant, they are Nitric oxide NO, Nitrogen dioxide NO_2 and Nitrous oxide N_2O. Following reaction describes dominant reaction in SNCR process with Urea.

Primary breakdown process of urea solution droplets (thermal dissociation of Urea) is specified by two chemical reactions. Urea is injected into furnace as water solution. First the Urea solution is evaporated by reaction (4) so that Urea reacts with water. Than water is evaporated from droplet and only solid Urea is sublimated. Urea is thermal dissociated by reaction (3) respectively [2].

$$\left(NH_2\right)_2 CO \rightarrow NH_3 + HNCO, \tag{3}$$

$$\left(NH_2\right)_2 CO + H_2O \rightarrow 2NH_3 + CO_2, \tag{4}$$

Nitric oxide NO and Nitrogen dioxide NO2 reduction process are described by following reaction (5),(6). These reactions specify the conversion of Nitric oxide NO and Nitrogen dioxide NO_2 into Nitrogen N_2 and Water H_2O. These reactions represent positive influence of SNCR method on NO_x pollutant concentration in flue gas. Reactions rate are significant above temperature 850°C. Reactions are relatively slowly below temperature 850°C that means the concentration of Ammonia NH_3 increase in flue gas.

$$NH_3 + NO \rightarrow N_2 + 1{,}056H_2O\,, \tag{5}$$

$$NH_3 + NO_2 \rightarrow N_2 + 1{,}994H_2O\,, \tag{6}$$

Next dominant reaction describes oxidation of Ammonia NH_3. This reaction is significant above temperature 1050°C and it is faster than reaction (5) and (6). Products of reaction are Nitric oxide NO and Water. This reaction represents higher level of temperature window for SNCR.

$$NH_3 + O_2 \rightarrow NO + 1{,}05H_2O\,, \tag{7}$$

One problem of SNCR is second channel of primary breakdown (3). Urea seems to involve a significant emission of the pollutants, such as Carbon monoxide CO and Nitrous oxide N_2O. Productions of pollutants are described by following reactions (8), (9).

$$HNCO + 0{,}25O_2 \rightarrow NCO + 0{,}5H_2O\,, \tag{8}$$

$$NCO + NO \rightarrow N_2O + CO\,, \tag{9}$$

Production of Carbon monoxide CO and Nitrous oxide N2O is partially reduced by following reactions (10), (11). First reaction describes spontaneous thermal decomposition of Nitrous oxide N_2O. Second reaction describes combustion of Carbon monoxide CO.

$$N_2O \rightarrow N_2 + 0{,}5O_2\,, \tag{10}$$

$$CO + 0{,}5O_2 \rightarrow CO_2\,, \tag{11}$$

Simplified SNCR reaction mechanism is described by previous reactions. Proposal mechanism does not include Ammonia radical NH_2^- that means the problematic non-stable transitional material is not necessary to specified. Reaction mechanism with NH_2^- radical is complicated. Proposed chemical mechanism for SNCR with Urea solution is relatively precise and simply. Complexity and computational intensity of CFD simulation is depended on number of materials and reaction. Reaction mechanism was proposed with reference to simulation speed and practise usability.

4 CFD SIMULATION PROCESS

CFD simulation was divided into two basic steps. First the CFD simulation without SNCR reaction was performed. This simulation was used to verification of concentration of all pollutants without SNCR, temperature field in furnace and mass flow of flue gas. This simulation represents initial data file for simulation with SCNR. Next the CFD simulation with SNCR reaction mechanism was performed.

5 RESULTS

CFD simulations were compared with results of experimental measurement. The CFD simulations and experimental measurements were performed for boiled load 60, 80, 100 %. Location of Urea solution injection lancets were designed with to view to measured temperature window. But the location is not optimum, it is due to design of boiler and manipulation restriction. The main results are well-arranged in Tab. 1.

Tab. 1 Comparison of results, experimental measurement vs. CFD simulation

		Concentration	[mg.m_N^{-3}], 6% O_2, t=0°C					[%]
		Material	CO	NO_x	NO_2	N_2O	NH_3	O_2
Validation of the SNCR model	Without SNCR	Measurement	23,0	375,0	28,1	13,5	0	6,4
		CFD	23,7	392,3	27,2	16,0	0	5,9
	With SNCR	Measurement	41,3	192,0	16,4	31,8	1,2	6,4
		CFD	50,5	187,7	15,2	42,5	9,2	5,9
Verification of the SNCR model	Without SNCR	Measurement	19,4	300,8	68,5	1,6	0	5,4
		CFD	20,9	298,0	67,2	2,1	0	3,8
	With SNCR	Measurement	45,8	127,5	64,8	42,3	25,2	5,4
		CFD	46,2	138,7	56,0	21,3	26,2	3,8

The measured data was defined as half hour time average. The CFD results were primarily computed as mass flow average of mass fraction. Both results (measured and CFD) were specified for normal condition, it is temperature 0°C, pressure 101325 Pa and 6% of O_2 concentration in flue gas. Most significant mistake of CFD simulation is value of oxygen concentration in flue gas. This value is probably impressed with false air, which penetrates into furnace per wall leakage. It is problematic to specified sources of leakage and it is problem to include leakage to CFD simulation.

Main aim of CFD simulation is possibility to study aerodynamic in zone of Urea solution injection. Especially it can be monitored droplets trajectory, time of droplet live, concentration of each materials and reaction rate. Results of the CFD simulation can be used to prediction of SNCR technology efficiency too.

We can see that reduction of NO_x is dependent on production of Ammonia HN_3. The Urea solution droplets are evaporated very quickly and the time of droplets live is approximately 1.5s, and the thermal breakdown of Urea solution is finished before first pipe heat exchanger. The increasing of the pollutants CO and N_2O can be predicted too. The Ammonia slip is low, the value is practically immeasurable, that means the injection lancet are suitable dislocated. In other words the Urea solution is injected into temperature window 850-1050°C. Low increase of pollutant CO and N_2O predicate of high efficiency of NO_x reduction too.

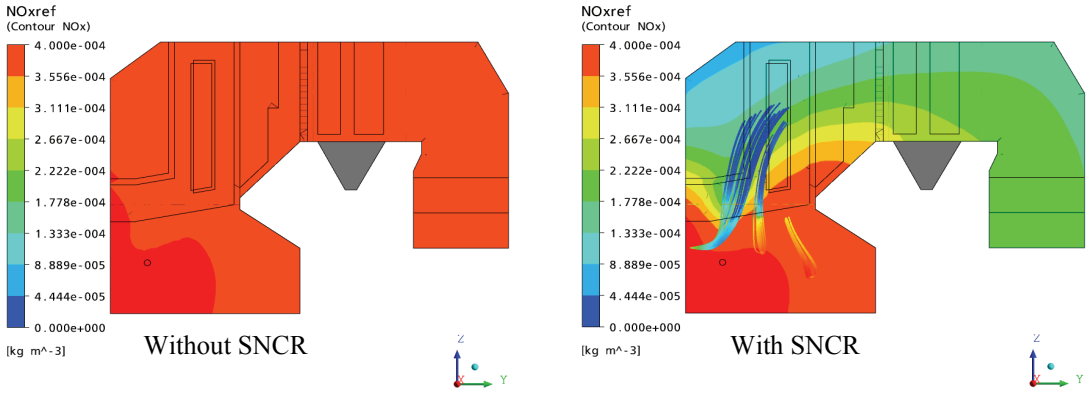

Fig. 3 Comparison of NO_x concentration for simulation of SNCR model validation

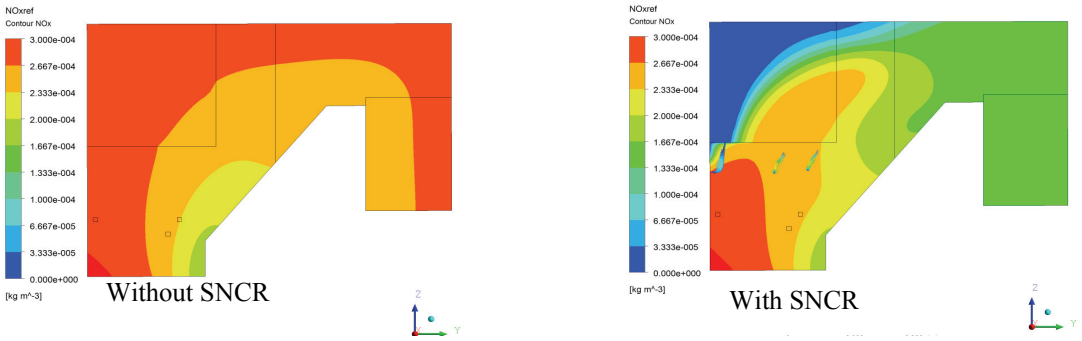

Fig. 4 Comparison of NO_x concentration for simulation of SNCR model validation

6 Conclusions

The goal of this study is proposition of chemical reaction scheme of SNCR method with Urea solution, which can be included into CFD simulation. Proposed model can be used to prediction of efficiency of SNCR technology, and it can be verified the design of injection lancet dislocation. Experimental measurement must be performed for specification of temperature window and boundary condition specification. In the simulation is not included leakage. This model can be used to study of local aerodynamic and droplets trajectory too. The significant materials CO, CO_2, NO, NO_2, N_2O a NH_3 are included in the result of simulation. Proposed model is verified only for two cases of realisations, so the model must be verified repeatedly. Than the SNCR model can be consider as credible and universal significant.

The next work will be focused on verification of actual SNCR with Urea solution on other boilers. The new model of SNCR with Ammonia water will be compiled on the basis of present model in the future.

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