Y. Sun, V. Morgunov, A.G. Chmielewski

MODELING STUDY OF NOX REMOVAL IN FLUE GAS IN THE PRESENCE OF C_2H_6 UNDER ELECTRON BEAM IRRADIATION

Electron Beam Flue Gas Treatment (EBFGT) technology has demonstrated its efficiency in purification flue gases from SO_x and NO_x for coal and oil fired boilers [1]. High removal efficiency of SO_2 (> 95%) and NO_x(>70%) has been demonstrated and industrial plant applying this process has been built in Poland [2]. However SO_2 removal from off-gases by using EB is relatively easy, but NO_x removal needs higher energy consumption. It demands a new method to remove NOx with lower energy consumption. Our previous work showed that NO_x removal efficiency was improved with the presence of alcohol [3]. In this work we theoretically studied NO_x removal with presence of C_2H_6 with the aid of computer simulation.

The computer simulation of NO_x removal in flue gas under EB-irradiation was carried out by using self-developed computer code "ELO", GEAR method was used. 883 reactions involving 94 species were considered for NO_x+ (air +CO_2 + H_2O) + C_2H_6 (0 – 400 ppm) system, and 998 reactions involving 137 species were considered for NO_x + (air +CO_2 + H_2O) + 400 ppm C_2H_6 + 700 ppm SO_2. Five main groups of reactions were included, the rate constants of reactions were mostly taken from the literatures [4, 5, 6]. The units of rate constant are /s, m^3/mole s and m^6/mole^2 s for first-, second and third- order reactions, respectively.

When fast electrons from electron beams are absorbed in the carrier gas, they cause ionization and excitation process of the nitrogen, oxygen, CO_2 and H_2O molecules in the carrier gas. Primary species and secondary electrons are formed.

The generation of active species under the electron beam is described by [6]:

$$\frac{dn_i}{dt} = G_{n_i} \dot{D} x_i \rho, \qquad (1)$$

where n_i - concentration of i-th component, mole/m^3; G_{n_i} - radiation yield of the i-th component of the gas, mole/J; x_i - mole fraction of i-th component; $\dot{D}$ - dose rate, J/(kg·s); ρ - gas density, kg/m^3.

Kinetics of chemical reactions of species formed during the gas irradiation with molecules of the gas medium and with one another is described by differential equations:

$$\frac{dn_i}{dt} = n_i \sum_n k_i^{(n)} \prod_{k=1}^{n} n_k \qquad (2)$$

For given initial concentrations:

$$n_i(0) = n_{i0} \qquad (3)$$

where, n_i - concentration of i-th component, mole/m^3; $k_i^{(n)}$ - the rate constant for n-order chemical reaction between the i-th and the k-components of gas; n_k - concentration of k-th component, n_{i0} - the initial concentration of the i-th component. Calculations were made in the following conditions:

- NO = 494 ppm, NO_2 = 38 ppm, CO_2 = 7 %, H_2O = 10-11 % (v/v), O_2 = 10 %, N_2 as balance, T = 70 ^{0}C (no any additives);
- NO = 494 ppm, NO_2 = 38 ppm, CO_2 = 7 %, H_2O = 10-11 % (v/v), O_2 = 10 %, N_2 as balance, T = 7 0 ^{0}C (with presence of 100 ppm and 400 ppm C_2H_6, respectively);
- NO = 494 ppm, NO_2 = 38 ppm, CO_2 = 7 %, H_2O = 10-11 % (v/v), O_2 = 10 %, N_2 as balance, T = 70 ^{0}C (with presence of 700 ppm SO_2 and 400 ppm C_2H_6).

Fig.1 presents calculation and experimental results of NO_x removal in flue gas vs. dose under EB-irradiation. Calculation results agree with the experimental results [3] to some extent. NO_x removal under influence of additives is presented in Fig.2. It is seen that NO_x removal efficiency is slightly improved with the presence of C_2H_6. The key reactions are listed below:

$OH + C_2H_6 = C_2H_5\bullet + H_2O$ (R1)

$O_2 + C_2H_5\bullet = C_2H_5O_2\bullet$ (R2)

$2C_2H_5 O_2\bullet = 2C_2H_5O\bullet + O_2$ (R3)

$C_2H_5O_2\bullet + NO = C_2H_5O\bullet + NO_2$ (R4)

$C_2H_5O_2\bullet + NO + M = C_2H_5ONO_2 + M$ (R5)

$C_2H_5O_2\bullet + NO_2 + M = C_2H_5O_2NO_2 + M$ (R6)

$NO_2 + OH + M = HNO_3 + M$ (R7)

The oxidation–reduction cycle between NO_2 and NO is toward the oxidation path and an increase in NO_x removal efficiency is favored.

From calculation results, following conclusions are drawn:

1. Removal efficiency of NO_x is increased by 3 % at a dose of 10.9 kGy with the presence of C_2H_6 when concentration of C_2H_6 is in the range of 100 ppm to 400 ppm.
2. Removal efficiency of NO_x is decreased by 23.84 % at a dose of 10.9 kGy with the presence of

400 ppm C_2H_6 and 700 ppm SO_2. SO_2 presence decreases removal efficiency of NO_x when ammonia is not added.

References

[1]. Basfar A.A. et al: Fuel, 87, 8-9, 1446-1452 (2008).
[2]. Chmielewski A.G. et al.: Radiat. Phys. Chem.71, 1-2, 439-442 (2004).
[3]. Chmielewski A.G. et al.: Radiat. Phys. Chem. 65, 4-5, 397-403 (2002).
[4]. Albritton, DL: At. Data Nucl. Data, 22, 1-101(1978)
[5]. http://kinetics.nist.gov/kinetics/index.jsp
[6]. Mätzing H.: Advances in Chemical Physics Volume LXXX, John Wiley & Sons, Inc., New Jersey 1991, 315-402.

List title of figures