NO and O2 absorption in FeII(EDTA) solutions
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Chapter 8

Conclusions - A numerical, rate based steady state reactor model for the BiodeNOx absorber unit

Abstract
A numerical, rate based reactor model based on the film theory for mass transfer has been developed for the simultaneous absorption of NO and O₂ in aqueous Fe³⁺(EDTA) solutions in a counter current packed column reactor at isothermal conditions. The effect of process conditions (temperature, inlet Fe concentration and oxygen inlet concentration) on absorber performance (NO removal efficiency, selectivity, iron chelate conversion) have been assessed. Absorber performance is particularly depending on the absorber temperature and low temperature operation (298 K) results in a dramatic increase in NO removal efficiencies compared to the results obtained at 323 K. In addition, the selectivity for the NO reaction is also favored at lower temperatures. The model indicates that desorption of NO may occur at certain process conditions, suggesting that overdesign of the column might result in reduced column performance.
8.1 Introduction

The BiodeNOx process consists of two discrete stages, i.e. the reactive absorption of NO in an aqueous Fe\textsuperscript{II}(EDTA) solution to form Fe\textsuperscript{II}(EDTA)(NO) followed by regeneration of the Fe\textsuperscript{II}(EDTA)(NO) solution in a bioreactor using ethanol to form N\textsubscript{2} and CO\textsubscript{2} and Fe\textsuperscript{II}(EDTA) \cite{53}. A competing reaction in the absorber unit is the reaction between oxygen and Fe\textsuperscript{II}(EDTA) to form Fe\textsuperscript{III}(EDTA), which is not able to bind NO. In addition, the rate of oxidation is coupled to the rate of EDTA degradation, which is an important variable cost item. As such, it is of key importance to reduce the overall rate of the oxidation reaction and to enhance the overall rate of the reaction between NO and Fe\textsuperscript{II}(EDTA).

In this chapter, a rigorous, numerical, rate based, steady state reactor model for the absorber unit is presented. The model is based on the film theory for mass transfer. The objective is to determine the overall absorption rates of both NO and oxygen and their concentrations as a function of the position in the column. The model will be applied to predict the effects of various process variables on absorber performance and to select the optimum conditions for the absorber. The model also gives information on the overall rate of the undesired oxidation reaction compared to the overall rate of the reaction between NO and Fe\textsuperscript{II}(EDTA) and as such on the composition of the liquid outlet stream (C\textsubscript{Fe\textsuperscript{II}(EDTA)}, C\textsubscript{Fe\textsuperscript{III}(EDTA)} and C\textsubscript{Fe\textsuperscript{II}(EDTA)(NO)}). This is an important input variable for the design and operation of the bioreactor. In addition, the model may be applied for scale-up purposes and to get a better understanding of the results obtained in previous pilot plant scale studies. The experimental results reported in the previous chapters, like the kinetics of the various reactions and the physical properties of various components (solubilities, diffusivities) will be used to generate the reactor model and as such, the model integrates all results described in the previous chapters of this thesis.

8.2 Model considerations

The reactor model is developed for the treatment of a typical flue gas of a 600 MW coal fired power plant. The volumetric gas flow rate is set at 2 · 10\textsuperscript{6} Nm\textsuperscript{3}/h, the oxygen concentration in the flue gas is 5 vol\% and the NO concentration is 250 vppm. The desired removal efficiency for NO is set at 90\%. Various reactor configurations for reactive gas-liquid absorption have been developed and commercialized. We have selected a counter current packed tower absorber for the modeling study. This reactor appears to be particularly appropriate for the BiodeNOx process as it is capable of treating high gas flow rate with reasonably low pressure drops. In addition, empirical relations describing the hydrodynamics of packed tower reactors are well known in literature \cite{111}. A representation of the reactor configuration is given in Fig. 1. A gas mixture containing NO is fed at the bottom of the column. The aqueous phase containing Fe\textsuperscript{II}(EDTA) is fed to the top of the column.
8.3 Model development

The following assumptions were made when setting up the model:
- isothermal operation, axial temperature gradients due to the absorption of gases, water evaporation, and other heat effects are not taken into account;
- the flow pattern of the gas and liquid phase is assumed to be plug flow;
- mass transfer of components through the gas-liquid interface is described by the film theory;
- the volumetric flow rates of the gas and liquid phase are constant and not a function of the position;
- steady state operation.

8.3.1. Chemistry

In the column, NO (A) reacts reversible with Fe$^{II}$(EDTA) (B) to form Fe$^{II}$(EDTA)(NO) (P), see eq (1).

$$A + B \rightleftharpoons P$$ (1)

The reaction is known to be first order in substrates and product. The rate constant for the for- and backward reaction were measured by Schneppensieper et al [46]. The equilibrium constant for the reaction as a function of the temperature was measured previously in our group work [106]:

$$K = \exp\left(\frac{4702}{T} - 8.53\right) \text{ (m}^3/\text{mol)}$$ (2)
Oxygen (O) is also absorbed and reacts fast and irreversible with B:

$$ O + v_B B \rightarrow v_B Q $$ \hspace{1cm} (3)

All kinetic studies indicate that the reaction is first order in oxygen. However, the order in iron chelate appears to be more complex and values between 1 and 2 have been reported [96, 97]. Recent studies have indicated that the order in iron is a function of the iron chelate concentration [45, 57]. At low concentrations (< 10 mol/m³), the reactions is first order in iron whereas it becomes second order at higher concentrations. At typical BiodeNOx absorption conditions (T = 50 - 55 °C, $C_{FeII(EDTA)} = 10 - 50$ mol/m³, $C_{NO} = 0 - 250$ vppm, $C_{O2} = 10 - 20$ vol%, pH = 7), the reaction was proven to be first order in oxygen and second order in iron chelate [107].

The rate constant for the oxidation reaction as a function of the temperature was measured previously in our group and may be expressed as [107]:

$$ k_{12} = \exp \left( 10.156 - \frac{4615.4}{T} \right) \text{ (m}^6/\text{mol}^2 \text{ s)} $$ \hspace{1cm} (4)

8.3.2. Reactor model

The gas phase component balances, assuming plug flow, read:

$$ \phi_{n,G} \frac{dC_{n,G}}{dh} = -J_{n,G} aS_a \hspace{1cm} (n = A, O) $$ \hspace{1cm} (5)

where $J_{n,G}$ is the absorption flux through the gas liquid interface, with the positive direction from the gas towards the liquid. The boundary conditions to eq (5) simply state that at the bottom of the column, the gas phase concentrations equal the gas feed concentrations:

$$ (C_{n,G})_{h=0} = C_{n,G}^{in} \hspace{1cm} (n = A, O) $$ \hspace{1cm} (6)

For the liquid phase, in the bulk, we have the balances:

$$ \phi_{n,L} \frac{dC_{n,L}}{dh} = -J_{n,L} aS_a + \varepsilon LS_a R_{n,L} \hspace{1cm} (n = A, O, B, P, Q) $$ \hspace{1cm} (7)

where $J_{n,L}$ denotes the diffusion flux through the interface between the liquid film and the liquid bulk, with the positive direction from the liquid film towards the bulk. $R_{n,L}$ denotes the reaction rate in the liquid bulk, positive for reactants, negative for reaction products.

The boundary conditions to eq (7) read:

$$ (C_{n,L})_{h=H} = C_{n,L}^{in} \hspace{1cm} (n = A, O, B, P, Q) $$ \hspace{1cm} (8)

The system of equations (5) - (8) constitutes a two point boundary value problem and was solved using a relaxation method. The equations contain flux terms, $J_{n,L}$ and $J_{n,G}$. 
These fluxes are related to the concentration gradients in the gas and liquid film layers, which were obtained from volume element modeling activities (*vide infra*).

### 8.3.3. Volume element modeling

In the scrubber, across the gas liquid interface we have absorption of NO (A) and O\(_2\) (O):

\[ A_G \leftrightarrow A_L \]  \hspace{1cm} (9)
\[ O_G \leftrightarrow O_L \]  \hspace{1cm} (10)

In the liquid, the mass transfer is accompanied by chemical reactions, according to (1) and (3). The reaction rates of reactions (1) and (3) are given by:

\[ r_1 = k_{11}(C_A C_B - C_p / K) \]  \hspace{1cm} (11)
\[ r_3 = k_{12}C_O C_B^2 \]  \hspace{1cm} (12)

According to the film model, the material balances for diffusion with parallel reaction in the liquid film read [73]:

\[ D_A \frac{d^2 C_A}{dx^2} = R_A = r_1 \]  \hspace{1cm} (0 ≤ x ≤ δ)  \hspace{1cm} (13)
\[ D_O \frac{d^2 C_O}{dx^2} = R_O = r_3 \]  \hspace{1cm} (0 ≤ x ≤ δ)  \hspace{1cm} (14)
\[ D_B \frac{d^2 C_B}{dx^2} = R_B = r_1 + v_B r_3 \]  \hspace{1cm} (0 ≤ x ≤ δ)  \hspace{1cm} (15)
\[ D_P \frac{d^2 C_P}{dx^2} = R_P = -r_1 \]  \hspace{1cm} (0 ≤ x ≤ δ)  \hspace{1cm} (16)
\[ D_Q \frac{d^2 C_Q}{dx^2} = R_Q = -v_P r_3 \]  \hspace{1cm} (0 ≤ x ≤ δ)  \hspace{1cm} (17)

The boundary condition at the liquid bulk side of the film reads for all components:

\[ (C_n)_{x=\delta} = C_{n,L} \]  \hspace{1cm} (18)

At the interface, a distinction is made between transferable and non-transferable components. For the dissolved gasses the interface condition reads:

\[ -D_n \left( \frac{dC_n}{dx} \right)_{x=0} = k_{G,n} \left( C_{n,G} - \frac{(C_n)_{x=\delta}}{m_g} \right) \]  \hspace{1cm} (n = A, O)  \hspace{1cm} (19)

while for non-transferable components we have:

\[ \left( \frac{dC_n}{dx} \right)_{x=0} = 0 \]  \hspace{1cm} (n = B, P, Q)  \hspace{1cm} (20)

For the fluxes across the gas liquid interface we have:
\[ J_{n,G} = k_{G,n} \left( C_{n,G} - \frac{C'_{n,L}}{m_n} \right) \quad (n = A, O) \]  

(21)

For the fluxes from the liquid film into the liquid bulk at \( x = \delta \), \( J_{n,L} \), we have:

\[ J_{n,L} = -D_n \left( \frac{dC_n}{dx} \right)_{x=\delta} \quad (n = A, O, B, P, Q) \]  

(22)

The concentration profiles of the components were calculated solving the system of equations (13) - (18), using a relaxation method to obtain finite difference equations and solving the system with the method of Newton. Subsequently, the fluxes were calculated using equations (21) and (22). To obtain the gradient of the concentration at \( x = \delta \), a Taylor series expansion of the correspondent concentration was used.

**8.3.4. Model output**

The NO removal efficiency of the packed column reactor, \( \eta \), is defined as:

\[ \eta = \frac{C_{A,in} - C_{A,out}}{C_{A,in}} \cdot 100\% \]  

(23)

The selectivity of the gas absorption process in the absorber is defined as:

\[ \sigma = \frac{C_P}{C_P + C_Q} \cdot 100\% \]  

(24)

It is a measure of the desired overall rate of NO absorption compared to the overall rate of absorption of both oxygen and NO.

The total iron conversion is defined as:

\[ X_B = \left( 1 - \frac{C_{B,out}}{C_{B,in}} \right) \cdot 100\% \]  

(25)

**8.3.5 Physical constants and model parameters**

The diffusion coefficient of NO in water is given by Wise and Houghton [79]. The diffusion coefficient of oxygen in Fe\(^{III}\)(EDTA) solutions at various concentrations was calculated using the diffusivity of oxygen in water [82] and corrected for the presence of electrolytes [80]. The diffusion coefficients of the iron chelate, \( D_B \), was taken from O’Conner et al. [103]. The diffusion coefficients of NO and oxygen in the gas phase were calculated using an expression given by Reid et al. [83].

The solubilities of NO and O\(_2\) in Fe\(^{III}\)(EDTA) solutions were determined from physical absorption experiments using N\(_2\)O and the results are reported in previous studies [106, 107].

The relative viscosity of the Fe\(^{III}\)(EDTA) solutions with respect to water at various concentrations and temperatures were measured experimentally. The results are reported in a previous study [119].
The viscosity of the gas phase is assumed to be equal to the viscosity of N₂, which is given by Daubert and Danner [121]. The density of the iron chelate solutions was measured experimentally and the results are provided in reference [119]. An overview of all physical constants applied in the model ($T = 323$ K and $C_{FeII(EDTA)} = 10$ mol/m³) is given in Table 1.

| DNO,L (m²/s) | 8.65 · 10⁻⁹ |
| D₂O₂,L (m²/s) | 4.07 · 10⁻⁹ |
| DFeII(EDTA),L (m²/s) | 1.08 · 10⁻⁹ |
| DNO,G (m²/s) | 2.79 · 10⁻⁴ |
| DO₂,G (m²/s) | 2.40 · 10⁻⁴ |
| HeNO (Pa m³/mol) | 7.2 · 10⁴ |
| HeO₂ (Pa m³/mol) | 1.08 · 10⁵ |
| $µ_{FeII(EDTA)} / µ_W$ | 1.02 |
| $ρ_{FeII(EDTA)}$ (kg/m³) | 992 |

The geometric area ($a_p$), the packing factor ($FP$) and the characteristic dimension of the packing ($DP$) were taken from Trambouze et al. [111]. For 1” metal Pall rings, $a_p$ is 240 m²/m³, $FP$ is 150 m²/m³ and $DP$ is 25 mm. The interfacial area of the column ($a$) was considered equal to the geometric area of the packing ($a_p$). At flooding conditions, the superficial gas velocity ($V_{SG}$) is equal to the flooding value, $V_{SGe}$. For this situation, the following expressions hold [111]:

\[
\ln \pi_w = 0.1117 - 4.012 \pi_2 \frac{1}{3} \tag{28}
\]

with:

\[
\pi_1 = \frac{F_p}{g} \left( \frac{ρ_G}{ρ_L} \right) \left( \frac{ρ_w}{ρ_L} \right) \left( \frac{µ_L}{µ_w} \right)^{0.2} V_{SG}^2 \tag{29}
\]

\[
\pi_2 = V_{SL} \sqrt{\frac{ρ_L}{ρ_G}} \tag{30}
\]

The minimum liquid velocity in order to guarantee full wetting of the column material was calculated using the following expression [111]:

\[
\ln \pi_w = 0.1117 - 4.012 \pi_2 \frac{1}{3} \tag{28}
\]
\[
\frac{V_{sl}}{a_p} \geq 3.5 \cdot 10^{-3} \left( \frac{\Omega_w}{\Omega_C} \right)^2 \frac{m^3}{m \cdot h}
\]

(31)

where \(\Omega_C\) is the critical surface tension of the packing material, which is equal to 25 \(\cdot 10^{-3}\) N/m in case of Pall metal rings packing [111] and \(\Omega_w\) is the surface tension of water [124].

An expression to calculate the pressure drop for a packed column was taken from Trambouze [111]:

\[
\frac{\Delta P}{H} = \frac{98\pi_t}{\pi_{1v}} \left( A_1 + A_2 \frac{\tau_{1v}}{\tau_{1e}} \right) \text{(Pa/m)}
\]

(32)

with:

\[
A_1 = 21.79 - 36.19\pi_{2\frac{1}{2}} + 16.60\pi_{2\frac{1}{2}}
\]

(33)

\[
A_2 = 7.06 - 10.30\pi_{2\frac{1}{2}} + 10.36\pi_{2\frac{1}{2}}
\]

(34)

The liquid hold-up in the column was calculated according a relation given by Buchanan [125]:

\[
\varepsilon_L = 2.2 \left( \frac{\mu_L V_{sl}}{g \rho_L d_p^2} \right)^{\frac{1}{3}} + 1.8 \left( \frac{V_{sl}^2}{g d_p} \right)^{\frac{1}{2}}
\]

(35)

8.4 Results and discussion

8.4.1 Definition base case and results

The reactor model has been applied to assess the effects of various process conditions on the NO removal efficiency using a (hypothetical) flue gas from a 600 MW coal fired power plant. The input, process conditions and design variables for the base case are provided in Table 2. Typical superficial gas- and liquid velocities for packed columns were applied. The liquid velocity was selected to guarantee full wetting of the column. The gas phase velocity was always less than the flooding value, \(V_{Sge}\), and typically between 55 and 60% of \(V_{Sge}\).

The inlet iron chelate concentration was selected on the basis of earlier simultaneous absorption studies of NO and O2 in Fe(II)(EDTA) solutions in a stirred cell reactor [118]. Here, the selectivity of the gas absorption process was highest for Fe(II)(EDTA) concentrations in the range 30 – 40 mol/m³. The flue gas temperature is expected to be significantly higher than 400 K. When brought into intimate contact with the liquid phase, the gas phase will be cooled and water will evaporate until the vapor pressure of water is reached. As such, temperature gradients are expected to be present in the column. This aspect has not been taken into consideration and it was assumed that flue gas of 328 K saturated with water enters the column.
Table 2. Base case input, process conditions and design variables.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>T (K)</td>
<td>323</td>
</tr>
<tr>
<td>$P_{\text{inlet}}$ (bar)</td>
<td>1.1</td>
</tr>
<tr>
<td>$\Phi_G$ (m$^3$/s)</td>
<td>556</td>
</tr>
<tr>
<td>$C_{\text{CO}_2}^{\text{in}}$ (vol%)</td>
<td>5%</td>
</tr>
<tr>
<td>$C_{\text{NO}}^{\text{in}}$ (vppm)</td>
<td>250</td>
</tr>
<tr>
<td>$C_{\text{NO}}^{\text{out}}$ (vppm)</td>
<td>25</td>
</tr>
<tr>
<td>$V_{\text{SL}}$ (m/s)</td>
<td>0.01</td>
</tr>
<tr>
<td>$V_{\text{SG}}$ (m/s)</td>
<td>1</td>
</tr>
<tr>
<td>$C_{\text{Fe}^{II}(EDTA)}^{\text{in}}$ (mol/m$^3$)</td>
<td>30</td>
</tr>
<tr>
<td>$k_{G,NO}$ (m/s)</td>
<td>$3.55 \cdot 10^{-2}$</td>
</tr>
<tr>
<td>$k_{G,O_2}$ (m/s)</td>
<td>$3.30 \cdot 10^{-2}$</td>
</tr>
<tr>
<td>$k_{L,NO}$ (m/s)</td>
<td>$3.88 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>$k_{L,O_2}$ (m/s)</td>
<td>$1.83 \cdot 10^{-4}$</td>
</tr>
<tr>
<td>$a$ (m$^2$/m$^3$)</td>
<td>240</td>
</tr>
<tr>
<td>% flooding</td>
<td>54</td>
</tr>
<tr>
<td>$\varepsilon_L$</td>
<td>0.058</td>
</tr>
</tbody>
</table>

Model calculations for the base case indicate that a column height of 0.91 m is required to obtain 90% NO removal efficiency. When considering the selected column diameter (26.6 m), this means that a large number of smaller columns have to be placed in a parallel mode. The total iron chelate conversion in the absorber is about 6.3%. The selectivity of the gas absorption is equal to 45%, meaning that 45% of the Fe$^{II}$(EDTA) is converted to Fe$^{III}$(EDTA)(NO) and 55% is oxidized to Fe$^{III}$(EDTA). The pressure drop for the base case is 290 Pa.

8.4.2. Effect of the column temperature

Previous experimental studies in stirred cell contactors have shown that the temperature has a profound effect on the overall absorption rates of NO and oxygen. Lower temperatures favored the overall rate of NO absorption compared to the overall rate of oxygen absorption [118].

The effect of the column temperature on column performance has been modeled in the range 298 - 323 K. At lower temperatures, the calculated height of the packed column to achieve 90% removal efficiency decreased considerably. As shown in Fig. 2, the calculated height at 298 K is 30% lower than at 323 K. These findings are in line with earlier experimental studies in a stirred cell contactor [106], where it was shown that the overall NO absorption rate is significantly higher at lower temperatures.

The selectivity of the gas absorption (eq. (26)) is also strongly temperature dependent and it increases from 45% at 328 K up to 66% at 298 K, see Fig. 3. Hence, the undesired oxidation reaction and the associated EDTA degradation rates are suppressed considerably at low temperatures. The pressure drops at lower temperatures are lower than the value calculated for the base case (328 K). These modeling results imply that the absorber unit should be operated at temperatures as low as possible.
8.4.3 Effect of the inlet $C_{FeII(EDTA)}$

The inlet $C_{FeII(EDTA)}$ has shown to be an important process variable in simultaneous absorption studies in stirred cell contactors. Model calculations with inlet $C_{FeII(EDTA)}$ in the range between 10 - 60 mol/m$^3$ were applied to study the effects on the absorption process in the packed tower reactor. All other conditions were equal to the base case as given in Table 2. The inlet $C_{FeII(EDTA)}$ has a clear effect on the column performance, see Fig. 4 for details. The required height of the reactor for $\eta = 90\%$ decreases exponentially with the $C_{FeII(EDTA)}$. The required height of the packed tower for 90% removal efficiency is 85% lower at 60 mol/m$^3$ when compared to 10 mol/m$^3$. 

Figure 2. Calculated column height for $\eta = 90\%$ as a function of the temperature. See Table 2 for the other conditions.

Figure 3. Calculated $\sigma$ (solid line) and $X_B$ (dotted line) as a function of the temperature for 90% NO removal. See Table 2 for the other conditions.
Figure 4. Required column height for 90% removal efficiency as a function of the inlet $C_{\text{Fe(II)(EDTA)}}$. Solid line: base case ($T = 323$ K); dashed line: $T = 298$ K; see Table 2 for other conditions.

The selectivity as a function of the $C_{\text{Fe(II)(EDTA)}}$ is represented in Fig. 5. The selectivity initially increases with the $C_{\text{Fe(II)(EDTA)}}$, it reaches a maximum value for $C_{\text{Fe(II)(EDTA)}} = 30 - 40$ mol/m$^3$ and subsequently decreases when $C_{\text{Fe(II)(EDTA)}}$ exceeds 40 mol/m$^3$. To suppress Fe oxidation at high temperatures, it appears beneficial to work in the range of 30 - 40 mol/m$^3$. As expected, $X_B$ decreases when the $C_{\text{Fe(II)(EDTA)}}$ increases. $X_B$ is 3.4% when the highest concentration of iron chelate is applied in the simulation.

In the previous paragraph, it was shown that it is favorable to operate the column at low temperatures. The effect of the $C_{\text{Fe(II)(EDTA)}}$ in the inlet on column performance was also evaluated for $T = 298$ K. The results are given in Fig. 4 and 5. As expected, the required column height for 90% removal efficiency is lower when applying higher $C_{\text{Fe(II)(EDTA)}}$ (Fig. 4). The required height at 298 K is always lower as for 323 K, irrespective of the $C_{\text{Fe(II)(EDTA)}}$. The selectivity of the process as a function of the $C_{\text{Fe(II)(EDTA)}}$ shows a different pattern at the two temperatures. At 323 K, the selectivity-versus the $C_{\text{Fe(II)(EDTA)}}$ shows an optimum at about 30 - 40 mol/m$^3$. This optimum is absent at 328 K and a small decrease of $\sigma$ was observed when increasing the $C_{\text{Fe(II)(EDTA)}}$. 


Figure 5. $\sigma$ and $X_B$ as a function of $C_{Fe^{II}(EDTA)}$ and temperature for $\eta = 90\%$. $T = 323$ K: $\sigma$ (solid line), $X_B$ (dotted line); $T = 298$ K: $\sigma$ (dashed line), $X_B$ (dashed-dotted line). See Table 2 for the other conditions.

Simulations performed for the base case ($T = 323$ K) and a relatively low $C_{Fe^{II}(EDTA)}$ (10 mol/m$^3$) reveal a very remarkable feature (Fig. 6). It appears that the column efficiency lowers when the height of the column is increased. This suggests that NO desorption may occur at certain conditions within the column and particularly when increasing the column length. In case the $C_{Fe^{II}(EDTA)}$ exceeds 10 mol/m$^3$, NO desorption was not observed. For instance, at 30 mol/m$^3$, efficiency drops were not observed when increasing the column height up to 12 m.

This behavior is likely the result of the mutual interactions between the reaction of Fe$^{II}$(EDTA) with NO and oxygen. Gambardella et al. [106] reported that the reaction between NO and Fe$^{II}$(EDTA) is an equilibrium reaction, whereas the reaction with oxygen is irreversible [107]. Both reactions occur simultaneously in the column and it is well possible that at certain conditions (high temperature and low $C_{Fe^{II}(EDTA)}$), the NO concentration in the gas phase is lower than the equilibrium value. Consequently, NO will desorb and the enhancement factor for NO absorption will become negative. These findings indicate that overdesign of the absorber unit may result in reduced column performance.
8.5 Conclusions

A rate based, steady state based reactor model has been developed for the simultaneous absorption of NO and oxygen in an aqueous Fe(II)(EDTA) solutions in a counter current packed (1” metal Pall rings) column reactor at isothermal conditions. When applying typical BiodeNOx conditions \( C_{Fe(II)(EDTA)} = 30 \text{ mol/m}^3, T = 323 \text{ K}, C_{NOin} = 250 \text{ vppm}, C_{O2in} = 5 \% \text{ vol} \) combined with a \( V_{SL} \) of 0.01 m/s and a \( V_{SG} = 1 \) m/s, the modeled height of the scrubber to achieve 90% NO removal efficiency is 0.91 m.

A remarkable improvement in absorber performance may be expected when operating the column at lower temperatures. Not only the required column height for 90% removal efficiency is reduced considerably, but also the overall rate of oxidation is significantly reduced. As such, the rate of EDTA degradation, which is known to be coupled with the rate of oxidation, will be reduced as well at low temperatures.

Variations of the inlet \( C_{Fe(II)(EDTA)} \) at 328 K, indicate that the selectivity of the absorption process shows a maximum value (45%) at \( C_{Fe(II)(EDTA)} \) between 30 and 40 mol/m\(^3\). Lowering the temperature to 298 K results in higher selectivities at 323 K (62%) and the observed maximum of the selectivity as a function of the \( C_{Fe(II)(EDTA)} \) is absent.

At certain process conditions, desorption of NO may occur, leading to a lowering of the NO removal efficiency at increasing column lengths. These findings indicate that overdesign of the absorber unit may result in reduced column performance.