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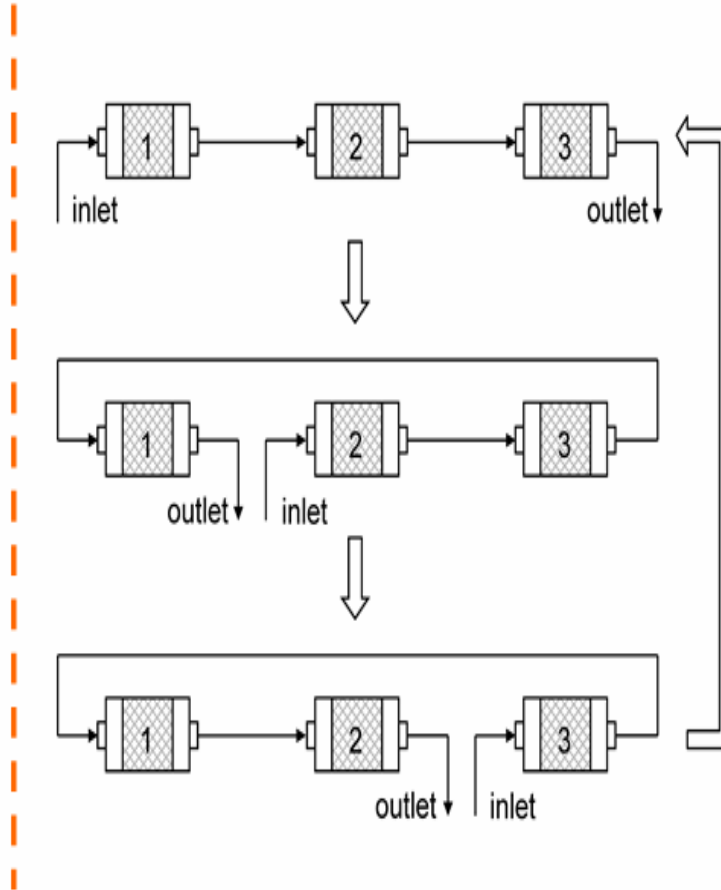
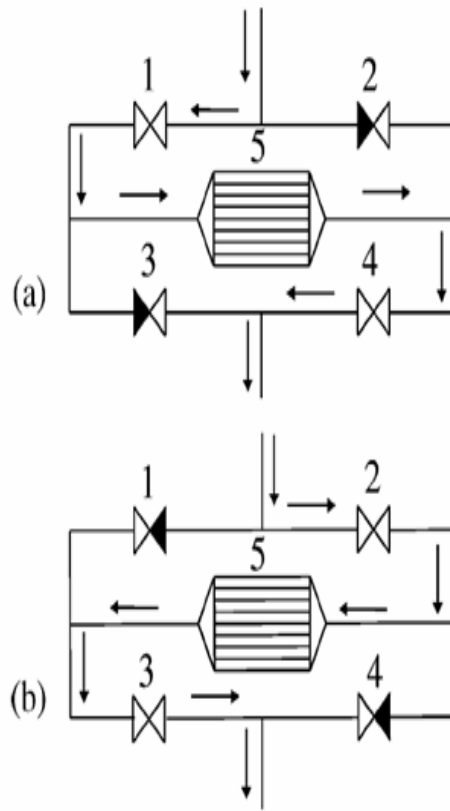
Selective Catalytic Reduction of NO_x with ammonia in unsteady-state reactors

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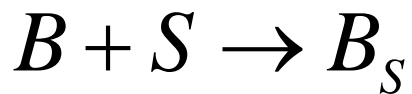
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Reverse Flow Reactor (RFR) vs. Reactor Network



Process mathematical model

Reaction mechanism:



Arrhenius type dependence of the kinetic constants with the temperature

Reactions kinetic:

$$\begin{cases} r_{red} = -k_{red} c_A^* \theta_B \\ r_{ads} = k_{ads} c_B^* (1 - \theta_B) \\ r_{des} = k_{des} \theta_B \end{cases}$$

$$\begin{cases} k_{red} = k_{0,red} e^{-\frac{E_{a,red}}{RT_S}} \\ k_{ads} = k_{0,ads} e^{-\frac{E_{a,ads}}{RT_S}} \\ k_{des} = k_{0,des} e^{-\frac{E_{a,des}}{RT_S}} \end{cases}$$

Process mathematical model

Gas phase mass balance:

Mass balance at the interface:

$$\frac{\partial c_A}{\partial t} = -v \frac{\partial c_A}{\partial x} + h_A a_v (c_A^* - c_A)$$

$$-h_A a_v (c_A^* - c_A) = r_{red} \cdot \Omega$$

$$\frac{\partial c_B}{\partial t} = -v \frac{\partial c_B}{\partial x} + h_B a_v (c_B^* - c_B)$$

$$-h_B a_v (c_B^* - c_B) = (r_{ads} - r_{des}) \cdot \Omega$$

Solid phase mass balance:

Gas phase energy balance:

$$\frac{\partial \theta_B}{\partial t} = r_{ads} - r_{des} - r_{red}$$

$$\frac{\partial T_G}{\partial t} = -v \frac{\partial T_G}{\partial x} + \frac{h_T a_v}{\rho_G c_{p,G}} (T_G - T_S)$$

Solid phase energy balance:

$$\rho_S c_{p,S} \frac{\partial T_S}{\partial t} = -h_T a_v (T_S - T_G) + r_{red} \cdot \Omega \cdot (-\Delta H_{red}) + r_{ads} \cdot \Omega \cdot (-\Delta H_{ads}) + r_{des} \cdot \Omega \cdot (-\Delta H_{des})$$

Switching time influence

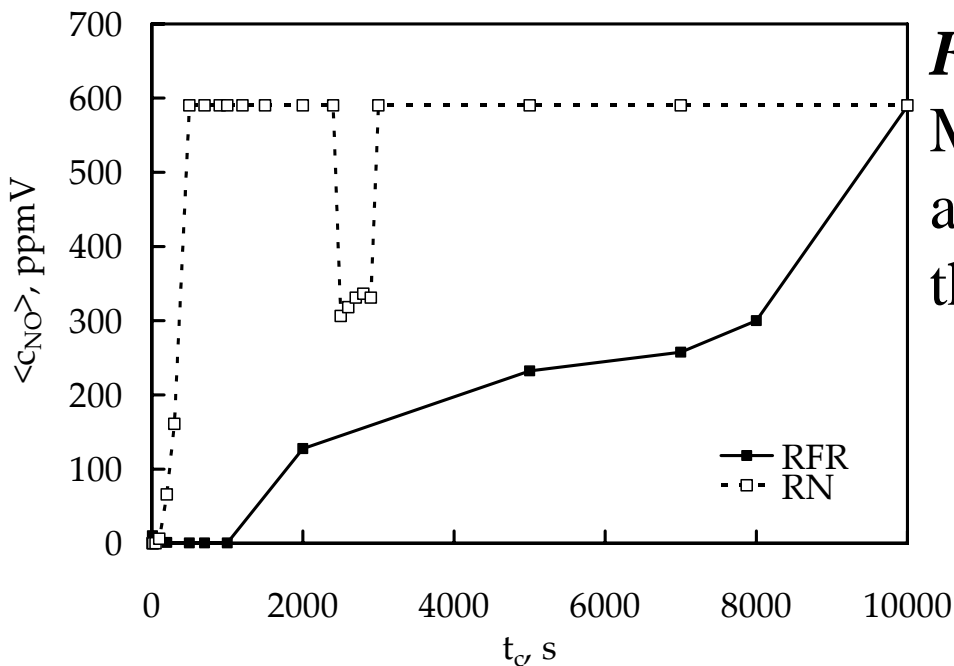
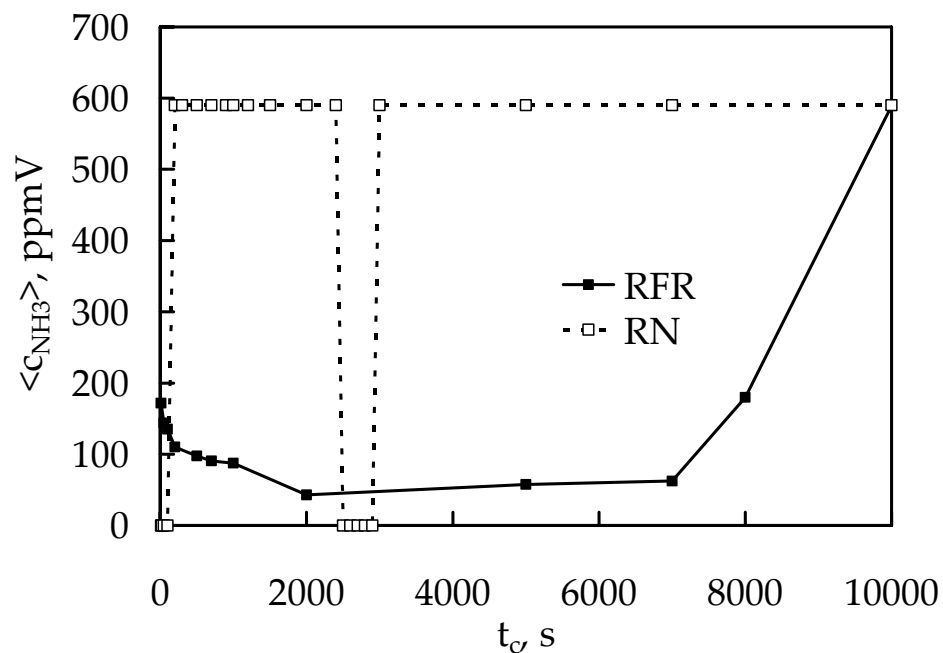


Figure 1.

Mean outlet concentration of NO as a function of the switching time in the RFR and in the RN.

Figure 2.

Mean outlet concentration NH₃ as a function of the switching time in the RFR and in the RN.



Influence of the surface velocity (RFR)

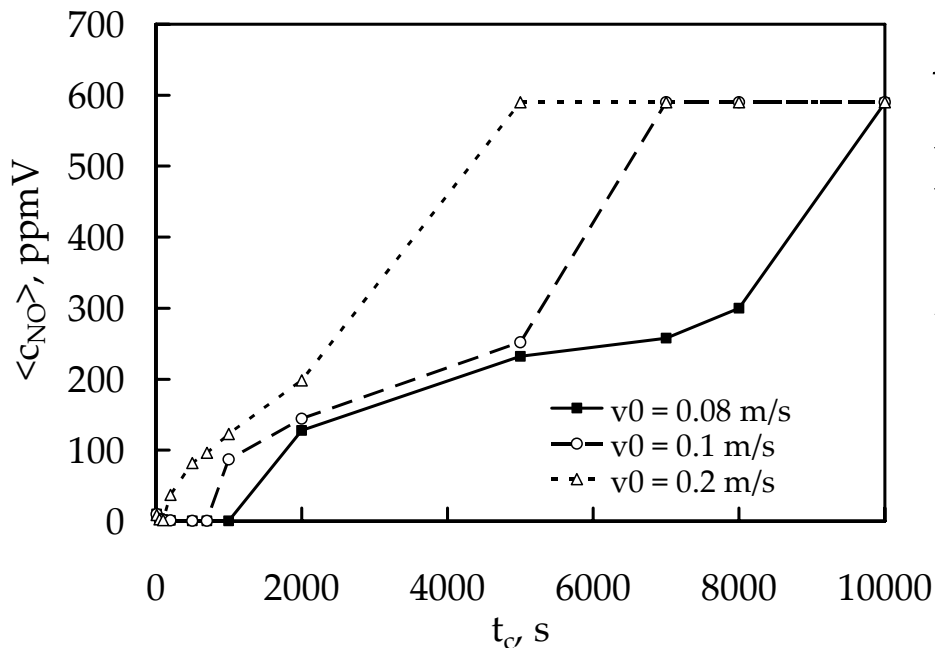
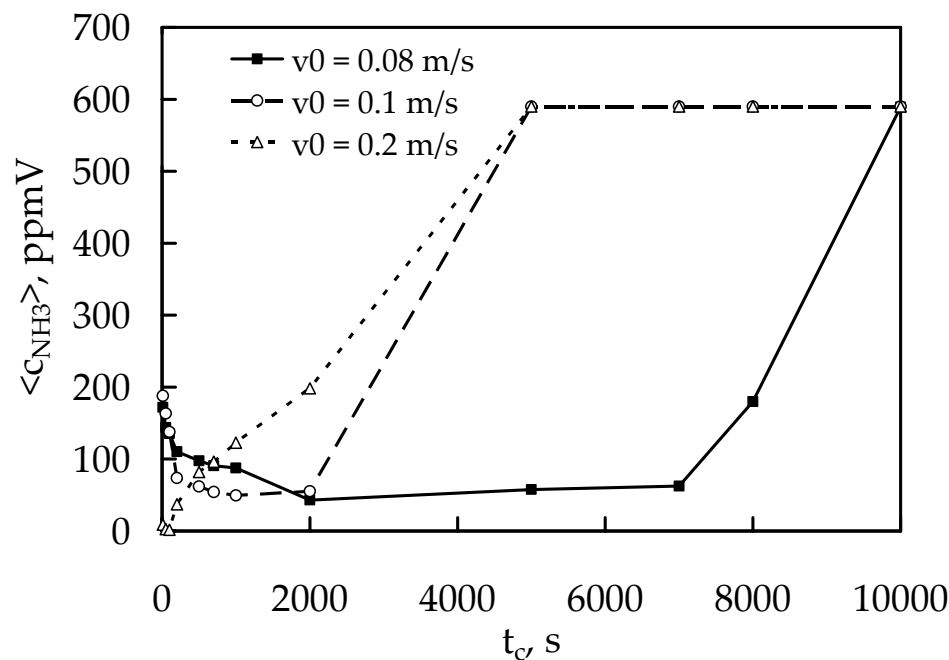


Figure 4.

Mean outlet concentration of NH₃ as a function of the switching time, for various values of the inlet flow rate.

Figure 3.

Mean outlet concentration of NO as a function of the switching time, for various values of the inlet flow rate.



Influence of the surface velocity (RN)

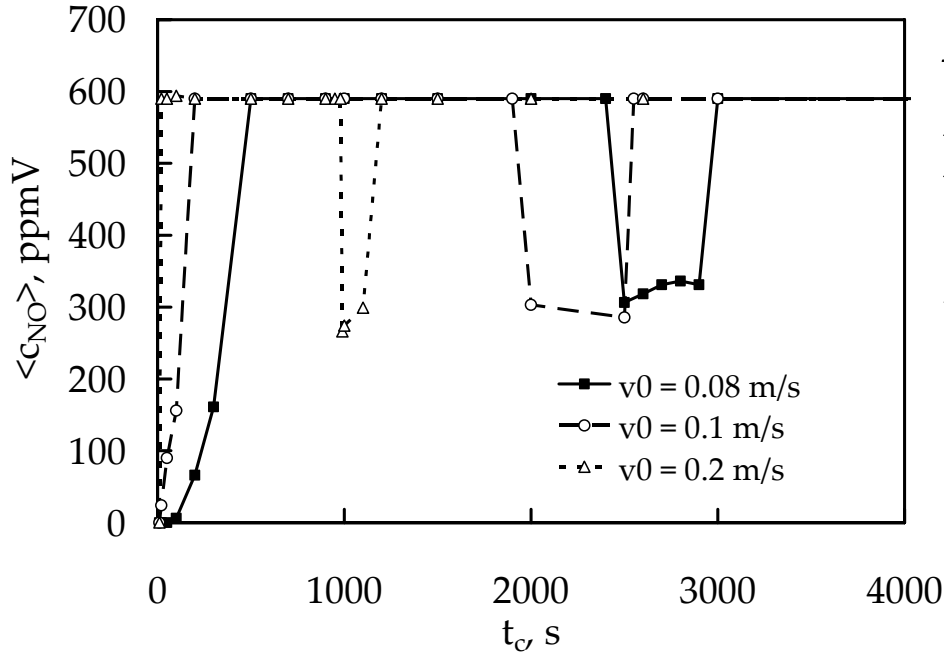
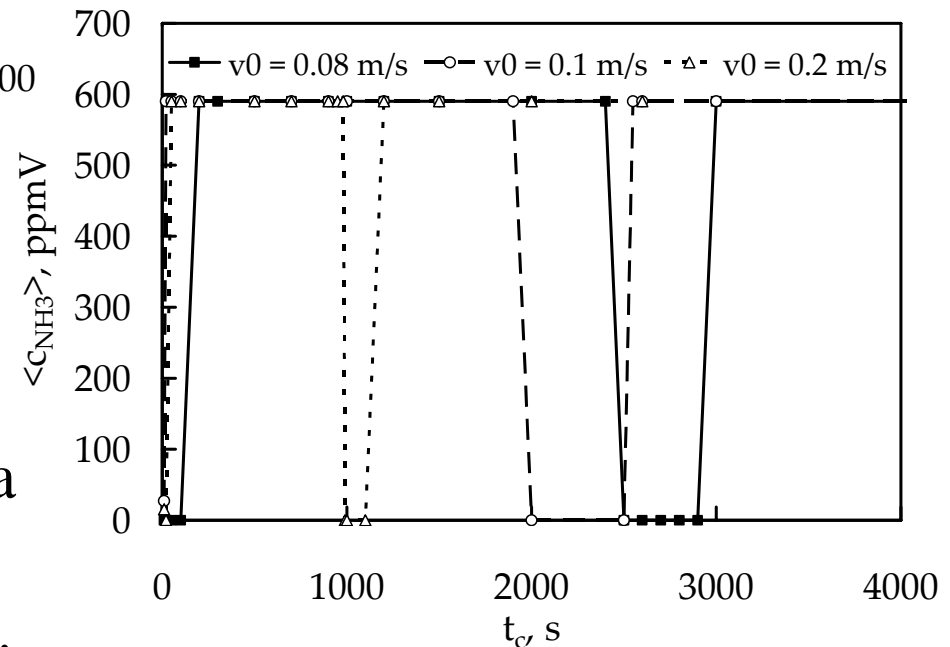


Figure 6.

Mean outlet concentration NH₃ as a function of the switching time, for various values of the inlet flow rate.

Figure 5.

Mean outlet concentration of NO as a function of the switching time, for various values of the inlet flow rate.



Response of the system to the disturbances

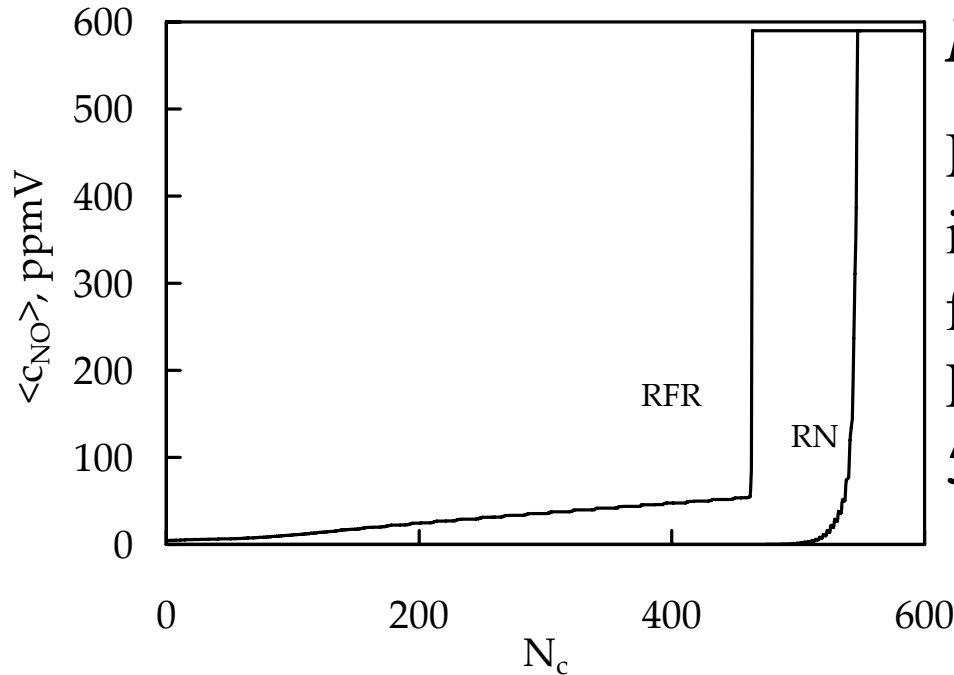
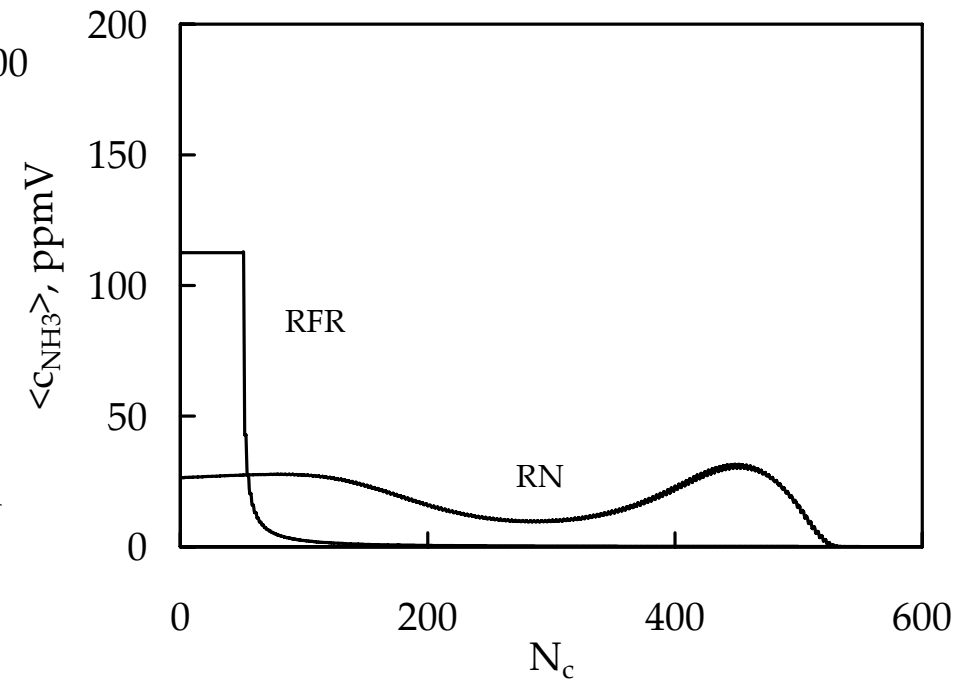


Figure 7.

Mean outlet concentration of NO in the RFR and in the RN as a function of the number of cycles. Feeding of NH₃ is stopped after 50 cycles.

Figure 8.

Mean outlet concentration NH₃ in the RFR and in the RN as a function of the number of cycles. Feeding of NH₃ is stopped after 50 cycles.



Response of the system to the disturbances

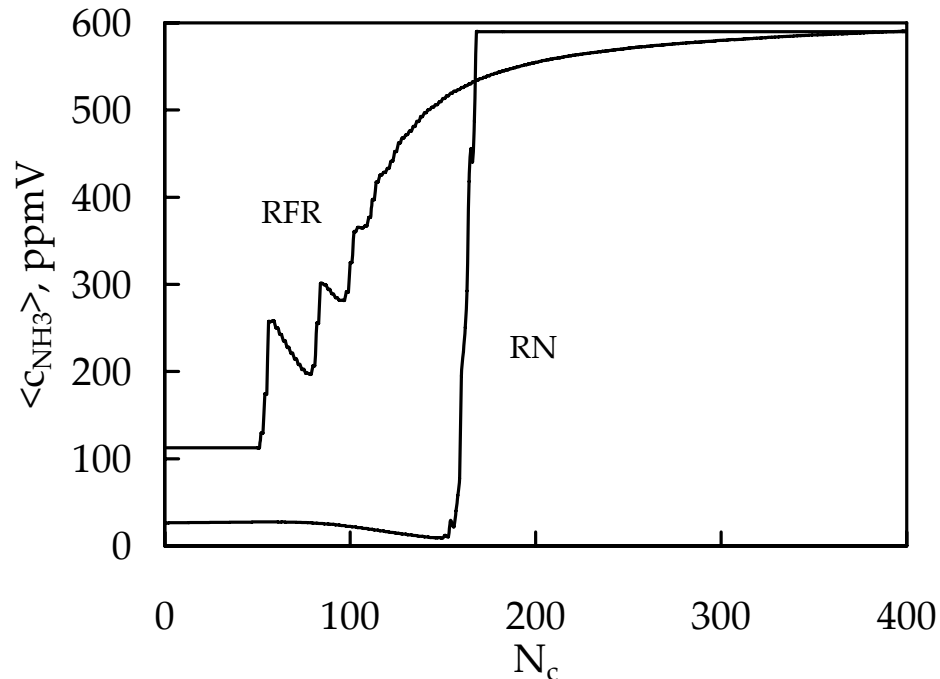


Figure 9.

Mean outlet concentration of NH₃ in the RFR and in the RN as a function of the number of cycles. Feeding of NO is stopped after 50 cycles.

Conclusions

The feasibility of the Selective Catalytic Reduction (SCR) of NO_x with ammonia in unsteady-state reactors has been demonstrated by means of numerical simulations.

A reactors network with periodical change of the feeding position was shown to be an alternative to the well investigated reverse-flow reactor, proving to be effective in avoiding the emission of ammonia (whose level is subject to stricter limits than for NO_x) that occurs in the RFR at every switch.

Non-isothermal operation was considered as even if the adiabatic temperature rise in NO_x removal is usually very low, the temperature rise in a RFR is a multiple of this value, thus allowing autothermal operation when low temperature gas is fed to the reactor.

Conclusions

In this case the choice of the switching time is affected not only by the dynamic of the trapping of one reactant, but also by the dynamic of the heat wave, as too long switching time will lead to reaction extinction, due to the heat removal from the catalyst.

The higher is the inlet flow rate, the narrower is the range of switching time where autothermal operation with high conversion is obtained.

The response of the RFR and of the RN to disturbances in the feed composition has been investigated, evidencing that the robustness of the RN is higher, even if the controllability is poorer than in the RFR

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