Theoretical evidence of the observed kinetic order dependence on temperature during the N_2O decomposition over Fe-ZSM-5

Hazar Guesmi,^{*a}[†] Dorothee. Berthomieu,^{*a} Bryan Bromley,^b Bernard Coq^a and Lioubov Kiwi-Minsker^{*b}

a: MACS, Institut Charles Gerhardt, UMR 5253 CNRS/ENSCM/UM2/UM1, Ecole Nationale Supérieure de Chimie de Montpellier, France emails: hazar.guesmi@upmc.fr; dorothee.berthomieu@enscm.fr †present address: Laboratoire de Réactivité de Surface, UMR 7197 Université Pierre et Marie Curie, 4 place Jussieu, 75252 Paris, France b: Ecole Polytechnique Fédérale de Lausanne, GGRC-ISIC-EPFL, Station 6, CH-1015, Lausanne, Switzerland email lioubov.kiwiminsker@epfl.ch

Supplementary Information

Experimental determination of the reaction order.

To determine the reaction order in respect to N₂O, the N₂O decomposition was studied varying concentrations of N₂O (2.0, 1.3, 1.0 and 0.75 %) at the total gas flow rate constant. Finally, the concentration was adjusted again to 2.0% to check the reproducibility of the experiment and the catalyst stability. The result for 573 K is presented in Figure S1 showing little influence of N₂O concentration on the rate. The amount of catalyst has been adjusted to obtain a conversion < 15% allowing to consider the reactor as differential.

$$r = \frac{-dC_{N_2O}}{dt} \cong \frac{C_{N_2O,in} - C_{N_2O,out}}{\tau}$$
(eq. S1)

$$r = kC_{N,O}^n$$
 (eq. S2)

$$C_{N_2O,in} - C_{N_2O,out} = k\tau \cdot C_{N_2O}^n$$
 (eq. S3)

$$\ln(C_{N_2O,in} - C_{N_2O,out}) = \ln(k\tau) + n\ln(C_{N_2O})$$
(eq. S4)

Reaction order has been determined using (eq. S1) by a linear fitting of $\ln(C_{N2O,in} - C_{N2O,out})$ as function of $\ln(C_{N2O})$ [mol.s⁻¹].

SI Figure captions

Figure S1: N_2O , N_2 , O_2 and Ar inlet and outlet concentrations monitored during the N_2O decomposition over HZSM-5_{1200Fe;40Si/Al} catalyst at 573 K under steady state conditions

Figure S2: Rate of N_2O decomposition as a function of N_2O concentration at 573 K on HZSM-5_{1200Fe;40Si/Al}

Figure S1



Figure S2

