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The origin of N_2O formation in the selective catalytic reduction of NO_x by NH_3 in O_2 rich atmosphere on Cu-faujasite catalysts

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Abstract

The selective catalytic reduction (SCR) of NO_x ($NO + NO_2$) by NH_3 in O_2 rich atmosphere has been studied on Cu-FAU catalysts with Cu nominal exchange degree from 25 to 195%. NO_2 promotes the NO conversion at $NO/NO_2 = 1$ and low Cu content. This is in agreement with next-nearest-neighbor (NNN) Cu ions as the most active sites and with N_xO_y adsorbed species formed between NO and NO_2 as a key intermediate. Special attention was paid to the origin of N_2O formation. CuO aggregates form 40–50% of N_2O at ca. 550 K and become inactive for the SCR above 650 K. NNN Cu ions located within the sodalite cages are active for N_2O formation above 600 K. This formation is greatly enhanced when NO_2 is present in the feed, and originated from the interaction between NO (or NO_2) and NH_3. The introduction of selected co-cations, e.g. Ba, reduces very significantly this N_2O formation. ©1999 Elsevier Science B.V. All rights reserved.

Keywords: Selective catalytic reduction; DeNO$_x$; Cu-faujasite; NH_3

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1. Introduction

N_2O is now considered as a pollutant which takes part in the greenhouse effect and depletion of the ozone layer. It was decided at the Kyoto Conference (1997) to reduce the emissions of N_2O issued from anthropogenic activities. These emissions mainly comes from both mobile and stationary sources (cars, power plants, nitric and adipic acid plants, etc.). For stationary sources, the selective catalytic reduction (SCR) by NH_3 is the preferred technology for NO_x removal. Most industrial SCR systems use vanadium-based formulations, efficient at medium temperatures. However, zeolite-based materials are excellent candidates for high temperature applications (> 400°C), where the oxidation of NH_3 competes with the SCR reaction on vanadium-based catalysts [1–4]. In particular Cu-faujasite (Cu-FAU) are low cost and very efficient catalysts for applications in acid nitric plants [5]. However, N_2O can be formed in the SCR process, in particular in the high temperature region (>350°C), and improved Cu-FAU formulations with exchanged co-cations were recently claimed to drastically reduce the N_2O formation [6]. From studies on model Cu-FAU with various Cu loadings it was suggested that the sites responsible for N_2O formation in the SCR of NO could be located in the sodalite cavities of Cu-FAU [7].

In industrial applications, the NO_2 content in the NO_x mixtures varies from 5% (power plant, total NO_x 300–1000 ppm) to 50% (acid nitric plant, total No_x 2000–3000 ppm). The presence of NO_2 promotes

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the activity of total NO_x reduction by NH_3 on both V_2O_5-TiO_2 catalysts [2,8], zeolite-based catalysts [9,10] and alumina-supported CuO and NiO catalysts [11]. It was thus shown that a synergistic effect exists between NO and NO_2 for the SCR of NO_x by NH_3 on Cu-mordenite, the activity goes through a maximum value for $NO/NO_2 \approx 1$ [10]. Beside this promotional effect of NO_2 on activity, an additional formation of N_2O due to the presence of NO_2 in the feed could occur [10]. The main goal of this work was then to identify the origin of N_2O formation in the SCR of $NO + NO_2$ mixtures on Cu-FAU catalysts, and to look at the influence of NO_2 on the SCR reaction.

2. Experimental

The Cu-FAU samples prepared for this study were labelled as follows: Cu(*x*)-FAU, *x* being the theoretical percent of Cu exchanged in the zeolite. Details of the preparation are given elsewhere [7]. In short, Cu(25)-, Cu(31)-, Cu(56)- and Cu(76)-FAU are prepared by ion-exchange of Na-FAU (Süd Chemie, Si/Al = 2.55, $S_{BET} \approx 700\,m^2\,g^{-1}$) with $Cu(NO_3)_2$, then filtration. A Cu(195)/FAU sample was prepared by impregnation of Na-FAU with $Cu(NO_3)_2$, then evaporation. All the samples were dried in an oven at 353 K for 2 h, then calcined in nitrogen at 723 K overnight. A Cu(35)-Ba(58)-FAU was prepared by a first ion-exchange of Na-FAU with $Ba(NO_3)_2$, then calcination at 773 K. A subsequent ion-exchange of Ba-FAU with $Cu(NO_3)_2$ was then carried out. The final thermal treatment was the same as above mentioned.

The samples were characterized by N_2 sorption, X-ray diffraction (XRD), temperature programmed reduction (TPR) and oxidation (TPO), temperature programmed desorption (TPD), IR and UV–VIS spectroscopies [7].

The SCR of NO with NH_3 was studied in a flow reactor operating at atmospheric pressure. Catalyst aliquots (≈20 mg) were activated in situ at 773 K in helium (ramp = $10\,K\,min^{-1}$, flow = $50\,cm^3\,min^{-1}$), and cooled to room temperature. The reaction gas, a mixture of 2000 ppm NO_x ($NO + NO_2$) with 2000 ppm NH_3 (or 2000 ppm $^{15}NH_3$), and 3% O_2 in helium, was fed to the catalyst. The space velocity was $250{,}000\,h^{-1}$ (flow = $130\,cm^3\,min^{-1}$). For the catalytic tests the temperature was decreased from 723 to 353 K in steps of 25 K and for 1 h. The catalytic behavior was recorded at the steady state during the last 15 min of each stages. The effluent composition was monitored continuously by sampling on line to a quadruple mass spectrometer (Balzers QMS 421) equipped with a Faraday detector (0–200 amu). Sampling was done through a capillary at constant flow rate and pressure drop. Nine masses characteristic of NO (30), NO_2 (30,46), N_2O (28,30,44), $^{14}N_2$ (28), $^{15}N\,^{14}N$ (29), NH_3 (17,18), H_2O (17,18), O_2 (16,32) and He (4) were followed. The intensities of NH_3 (17), H_2O (18), $^{14}N_2$ (28), $^{14}N^{15}N$ (29), NO (30), and NO_2 (46) were determined by solving a linear system of equations. The concentrations were derived from intensities by using prior standardization procedures before and after each experiments.

3. Results

Table 1 reports some characteristics of the samples [7]. The main point is that Cu(*x*)-FAU ($x \leq 76$) and Cu(36)-Ba(58)-FAU mainly contain isolated Cu ions

Table 1
Some characteristics of the Cu-FAU catalysts

Sample	S_{BET} ($m^2\,g^{-1}$)	$V_{\mu pores}$ ($cm^3\,g^{-1}$)	wt% Cu	Proportion of Cu species (%)		Cu^{2+}(supercages)/Cu^{2+}(sodalite)
				CuO	Cu ions	
Cu(25)-FAU[a]	680	0.32	2.9	0	100	0.36
Cu(31)-FAU[a]	630	0.32	3.3	0	100	0.38
Cu(56)-FAU[a]	660	0.34	5.7	0	100	0.30
Cu(76)-FAU[a]	620	0.32	7.3	5	95	0.24
Cu(195)/FAU[a]	300	0.15	17.6	80	20	–
Cu(35)-Ba(58)-FAU	–	–	3.5	0	100	2.90

[a] from [6].

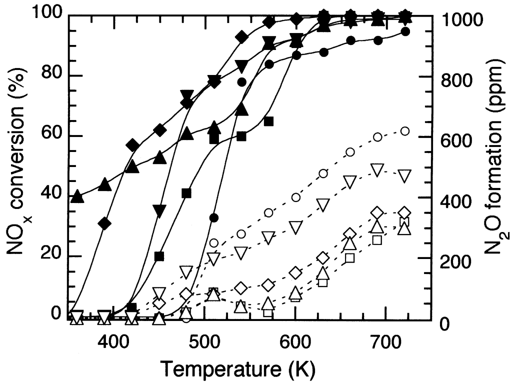

Fig. 1. SCR of NO_x by NH_3 on Cu(76)-FAU at various NO_x compositions: (■, □) NO, (▲,△) $NO/NO_2 = 3$, (◆, ◇) $NO/NO_2 = 1$, (▼, ▽) $NO/NO_2 = 0.33$, (●, ○) NO_2; full symbols = NO_x conversion, open symbols = N_2O formation.

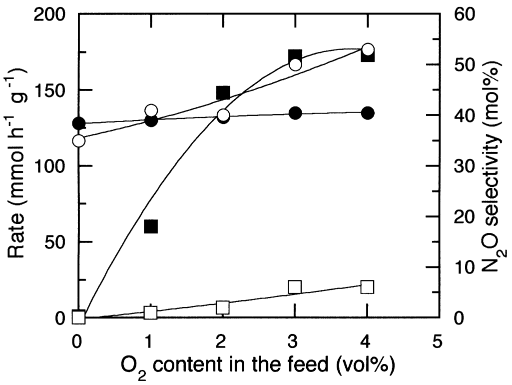

Fig. 2. NO_x conversion rate (full symbols) and N_2O selectivity (open symbols) at 723 K on Cu(56)-FAU as a function of O_2 content in the feed; (■, □) SCR of NO, (●, ○) SCR of NO_2; $NO = NO_2 = NH_3 = 2000$ ppm.

(>95%). In contrast, CuO aggregates are the main Cu species in Cu(195)/FAU, with an average size of 25 nm determined from XRD. This is the reason why S_{BET} and $V_{\mu\text{pores}}$ decrease in this sample due to pore blockage by CuO aggregates.

Fig. 1 presents the conversion of NO_x and the N_2O formation as a function of temperature at different $NO + NO_2$ compositions on the Cu(76)-FAU catalyst. The SCR profile of pure NO shows a first conversion wave at ca. 500 K, before reaching full NO conversion. The presence of NO_2 in the SCR of NO_x has three effects:

1. At $NO/NO_2 \geq 1$, the SCR activity is generally increased in agreement with previous reports on the SCR of NO_x on Cu-MOR [10], V_2O_5-TiO_2 [8], and CuO-NiO/Al_2O_3 [11].
2. At $NO/NO_2 < 1$ there is a shift of the light-off temperature (temperature of 50% NO_x conversion) to higher values when the NO_2 content increases in NO_x. In the SCR of pure NO_2, the activity thus falls abruptly from 80 to 0% at ca. 500 K. This actually corresponds to the formation of adsorbed NH_4NO_3 which inhibits the catalyst surface [12]. We shall recall that the catalytic tests were carried out by decreasing the temperature from 723 to 353 K. The thermal decomposition of FAU-supported NH_4NO_3 (ramp = 5 K min^{-1}) showed a sharp peak at 530 K with the concurrent release of N_2 (60%) and N_2O (40%) [12].
3. The N_2O formation increases with the concentration of NO_2 in the feed. At 700 K (full NO_x conversion), it is of 150 ppm for the SCR of pure NO, and steadily increases up to 600 ppm when processing pure NO_2.

Short parallel experiments have been performed on Cu(56)-FAU in order to examine the effect of the content in O_2, NO, NO_2 and NH_3 on the selectivity to N_2O. The content of these reactants were varied in the ranges: $1000 < NO < 3000$ ppm, $1000 < NO_2 < 3000$ ppm, $1000 < NH_3 < 3000$ ppm, $0 < O_2 < 4\%$. These experiments show that:

1. The O_2 content, which increases significantly the rate of NO reduction (Fig. 2), has no effect on the NO_2 reduction rate as previously reported for the SCR of NO_x on CuO/Al_2O_3 catalysts [11].
2. The selectivity to N_2O increases greatly with the NO_2 content, but slightly with the O_2 content, and decreases with the NH_3 content.

Fig. 3 presents the SCR profiles of NO_x on Cu(25)-FAU. The SCR activity is very low for pure NO, with the light-off temperature at ca. 650 K. In contrast, the SCR activity is boosted in the presence of NO_2 and the light-off temperature for the SCR of NO/NO_2 (1/1) falls to 525 K. This promotion of SCR activity by NO_2 is much greater than observed on Cu(76)-FAU. The N_2O formation is low on Cu(25)-FAU whatever the feed composition, and increased from 0 to 150 ppm for the SCR of NO and NO_2 at 700 K, respectively.

Fig. 4 presents the SCR profile of NO_x on Cu(195)/FAU, sample which mainly contains CuO aggregates. Whatever the NO_x composition the con-

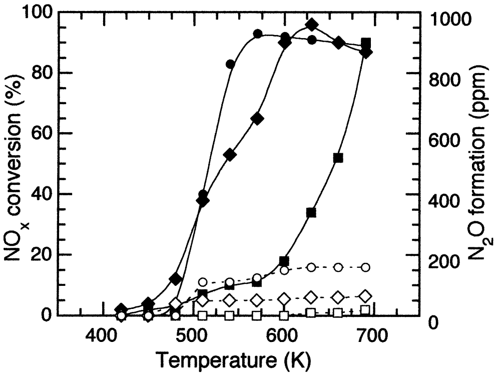

Fig. 3. SCR of NO_x by NH_3 on Cu(25)-FAU at various NO_x compositions; same symbols as in Fig. 1.

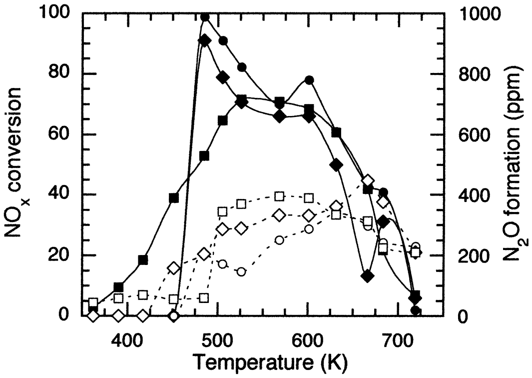

Fig. 4. SCR of NO_x by NH_3 on Cu(195)/FAU at various NO_x compositions; same symbols as in Fig. 1.

version of NO_x goes through a maximum value of 60–100% at ca. 500–520 K. In contrast, the NH_3 conversion steadily increases and reaches 100% at above 600 K. On CuO/Al_2O_3 Blanco et al. [11] reported the same behavior in the SCR of NO_x ($NO/NO_2 \leq 1$), with a maximum at 480–550 K. The decrease of the SCR activity above 500–550 K on Cu(195)/FAU was attributed to the onset of the side reaction of NH_3 oxidation by O_2 which competes with the main reaction of the oxidation of NH_3 by NO_x. The oxidation of NH_3 on Cu(195)/FAU yields N_2 mainly with increasing amounts of N_2O and NO at high temperature [13]. The formation of N_2O in the SCR of NO_x on Cu(195)/FAU is significant (up to 400–450 ppm) and little sensitive to the NO_x composition.

4. Discussion

The characteristics of the samples, as well as their redox and catalytic properties in the SCR of NO are detailed elsewhere [7,12]. To sum up:

1. In the samples prepared by ion exchange ($Cu \leq 8$ wt.%, $x \leq 76\%$), Cu is mainly in the form of ionic species. In contrast, the heavily impregnated Cu(195)/FAU sample ($Cu \approx 17.6$ wt.%) mainly contains CuO aggregates.
2. The TPR profiles of Cu(x)-FAU ($x \leq 76$) present two peaks of H_2 consumption in the low temperature region at 495–535 and 600–645 K (see Fig. 5 for Cu(25)-FAU as an example). They were assigned to the reduction of Cu^{2+} to Cu^{+} located in supercages and sodalite cavities, respectively. A small contribution could even be identified at 770 K likely due to reduction of Cu^{2+} in the hexagonal prisms. The proportion of Cu in supercages has been determined [7], and decreases when the exchange degree increases (Table 1). The reduction of Cu^{+} to Cu^{0} occurred above 1100 K.

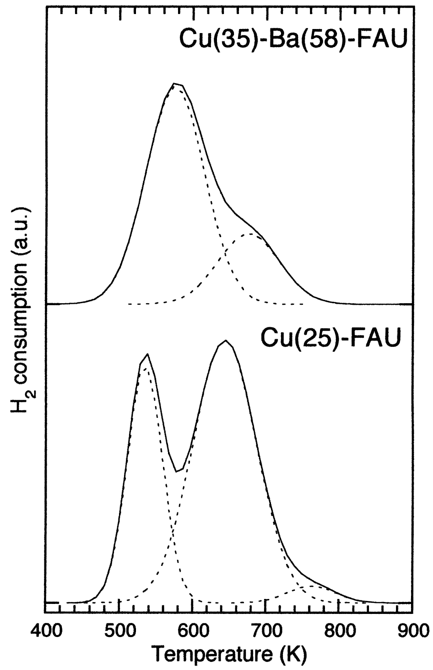

Fig. 5. Temperature programmed reduction profiles of Cu(25)-FAU and Cu(35)-Ba(58)-FAU; $m \approx 50$ mg, gas = H_2/Ar (3/97, v/v), ramp = 10 K min^{-1}, flow rate 18.6 cm^3 min^{-1}.

3. The TPD and IR spectroscopy of adsorbed NH_3, NO and NO_2 show that (i) Cu ions form a complex with 3–4 NH_3 ligands at room temperature (RT) and the last ligand desorbs at ca. 600 K (ramp = 5 K min^{-1}), (ii) NO forms weakly bonded nitrosyl species which desorb at RT in He flow, and (iii) NO_2 adsorbs as nitrate and nitrite species at RT. These observations are in general agreement with literature data (see [14] for a review). One should only remark that NO is much more weakly bonded on exchanged Cu-FAU than on Cu-MOR [15] and Cu-MFI [16].
4. The TPO (ramp = 5 K min^{-1}, VVH = 100,000 h^{-1}) of $Cu^+(x)$-FAU shows that NO/He (0.2/99.8) cannot oxidize Cu^+ to Cu^{2+} below 773 K, but O_2/He (0.2/99.8) and better ($NO + O_2$)/He ((0.2 + 0.2)/99.8) can do it. The TPO profiles with ($NO + O_2$)/He mixture present two peaks of O_2 and NO consumption at ca. 500 K ($NO/O_2 \approx 3$) and 580 K ($NO/O_2 \approx 1$) with the concurrent release of N_2 and H_2O.
5. In the SCR by NH_3 on Cu(*x*)-FAU ($x \leq 76$) two waves of NO conversion occur in the range 400–500 K and above 600 K. This behavior was assigned to NO conversion on two different Cu sites. It was proposed that in the range 400–550 K NO is reduced on specific Cu^{n+} sites located in the supercages and stabilized by NH_3 ligands. The plateau of activity observed at ca. 550 K, and delimiting the first wave of NO conversion, was assumed to be related with the migration of some reactive Cu species from supercages to sodalite cavities [7]. Above 600 K, undifferentiated Cu^{n+} sites in both sodalite cavities and supercages of FAU are active.

The catalytic cycle can be then proposed as in Fig. 6, in which Cu^{2+} is reduced to Cu^+ by $NO + NH_3$, Cu^+ is in turn oxidized to Cu^{2+} by $NO + O_2$. In this scheme, $n = 0$, 1 and $x = 2$, 3 very likely, x being the number of NNN Cu ions in the active sites [7,12]. The originality of this SCR mechanism is that reduction of NO to N_2 is involved in both the oxidation and reduction steps of Cu ions. Based on this catalytic cycle and the TPO data, a global reaction was proposed for the SCR process below 550 K [7]:

$$13NO + 10NH_3 + O_2 \rightarrow 15H_2O + \left[\frac{23}{2}\right]N_2 \qquad (1)$$

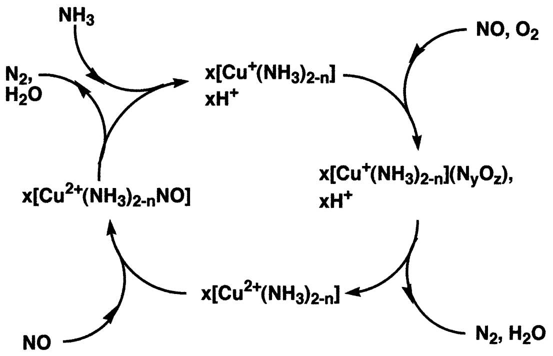

Fig. 6. Catalytic cycle of the SCR of NO to N_2 by NH_3 on Cu ions in Cu(*x*)-FAU.

This reaction is basically different from that claimed in the SCR on V_2O_5-TiO_2 catalysts [2]:

$$4NO + 4NH_3 + O_2 \rightarrow 4N_2 + 6H_2O.$$

The stoichiometry $NH_3/NO \approx 0.77$ of Eq. (1) was confirmed by SCR of ^{14}NO by $^{15}NH_3$ at 500 K [7]. The dinitrogen formed was composed of 84% $^{15}N^{14}N$ and 16% $^{14}N_2$.

From the present catalytic results two important points deserve a discussion: the SCR reactivity in presence of $NO + NO_2$ mixture and the N_2O formation.

It is worthy to note first that the SCR activity at 500 K on Cu(195)/FAU is only slightly increased from 60% for pure NO to 90–100% for $NO + NO_2$ mixtures. This can be compared to the report of Blanco et al. [11] on the SCR of NO_x over CuO/Al_2O_3: the NO_x conversion at 500 K increased from 75% for pure NO to 85–90% for $NO + NO_2$ (1 + 1). The authors claimed that the surface of copper oxide aggregates was mainly composed of Cu_2O which reacts with NO_2 to yield CuO and NO. The latter being in turn reduced to N_2 by NH_3.

In contrast, there are clear changes of SCR activity when $NO + NO_2$ mixtures are processed on Cu(*x*)-FAU ($x \leq 76$), with respect to pure NO. And this is particularly true at low Cu exchange, e.g. Cu(25)-FAU. This point could be discussed in parallel to the effect of the presence of O_2 on the NO reduction. It is now perfectly established that O_2 boosts the NO reduction whatever the catalyst used in the SCR process [2,14]. The main role of O_2 seems to react with NO to form an active intermediate species which could be NO^+ [10,17,18], NO_2 [1,18], a nitrate [19], or more generally N_xO_y adsorbed species [7,20]. Particularly unclear is the role of these oxidized adspecies of nitrogen oxide. On a H-MOR, which very likely contained significant amounts of iron, Kiovsky et al. [1] claimed that NO_2 is the only reducible species and that the role of O_2 is to oxidize NO into NO_2; there was no synergy between NO and NO_2. In contrast, on Cu-MOR Brandin et al. [10] reported a synergy based on the oxidation of NO to NO^+ by NO_2, which will be the actual active species. Centi and Perathoner [20] proposed that NO reacts with Cu^{2+}-NO_2 species to form a Cu^{2+}-N_2O_3 like complex intermediate. From our results the synergy between NO and NO_2 exists but depends on the reaction temperature domain, the catalyst, and the NO/NO_2 ratio. The behavior appears complex and the comments need to be mitigated.

In the low temperature region (<550 K), the boosting effect of NO_2 on the conversion of NO_x generally occurs at $NO/NO_x \geq 1$, but a breaking in NO_x conversion happens when $NO/NO_2 < 1$, which shifted then to higher temperatures when the NO_2 content increases; this is due to the formation of 'NH_4NO_3-like' adsorbed species which inhibited the catalysts. In the high temperature range (>550 K) the boosting effect of NO_2 on NO_x conversion is of lower magnitude with Cu(76)-FAU but remains large for Cu(25)-FAU, since the light off temperature was decreased by ca. 130 K (Fig. 3). On that account, it appears clearly that the synergistic effect of NO_2 on the SCR of NO_x is all the larger as the Cu content decreases. Previous works on the SCR of NO on Cu(*x*)-zeolite [7,21] showed that the TOF, expressed per Cu site, increased with Cu content. From these observations it was proposed that the active site in Cu(*x*)-FAU would be composed of several next-nearest-neighbor (NNN) Cu ions located in supercages. This active site might be described as dimer, or trimer species, as proposed by Komatsu et al. [21] for the SCR of NO on Cu-MFI and Cu-MOR. From kinetic studies [12], we postulated also that the rate determining step would be the oxygen dissociation on this site with the subsequent formation of N_xO_y adsorbed species as intermediate. In the presence of NO_2 the oxygen dissociation step would be by-passed, as shown by the constancy of the SCR rate with pure NO_2 whatever the O_2 content in the feed (Fig. 2). From this point of view, the reactivity of NO + NO_2 mixtures on Cu(*x*)-FAU ($x \leq 76$) is in agreement with the proposed description of both the active site and the slow step in the SCR of NO. The much higher synergistic effect of NO_2 on the SCR on Cu(25)-FAU compared to Cu(76)-FAU fairly agrees with NNN Cu ions as the active site on which the dissociation of O_2 takes place to yield further the reactive N_xO_y intermediate.

There are two modes of N_2O formation in the SCR of NO_x on Cu-FAU. The first is centered at ca. 520 K and the second occurs for temperatures above 650 K. The low temperature formation of N_2O can be easily explained by the comparative behavior of Cu(25)-FAU and Cu(195)/FAU. The former sample, free of CuO aggregates, does not promote N_2O formation. In contrast, Cu(195)/FAU which mainly contains CuO aggregates promotes the N_2O formation whatever the feed composition. Centi et al. [22] proposed in the SCR of NO on CuO/Al_2O_3 that N_2O is generated from the decomposition of 'ammonium nitrate' intermediate formed at the surface of CuO aggregates.

The N_2O formation at high temperature in the SCR on Cu(*x*)-FAU is highly promoted by both the Cu amount and the NO_2 content in the feed. Fig. 7 presents the N_2O formation in the SCR of NO on Cu(*x*)-FAU ($x \leq 76$) as a function of Cu content ($T_R = 720$ K, full NO conversion). CuO aggregates cannot be charged for this N_2O formation, which exists on Cu(37)-FAU and Cu(56)-FAU, samples free of CuO [7]. NNN Cu ions, or $[CuOCu]^{2+}$ dimers, can be postulated as the active species for this N_2O formation. It is obvious that the population of such species should increase with increasing the Cu concentration, if Cu ions are located at random at the cation sites. At 700 K, there is no NH_3 or H_2O ligand permanently bonded to Cu ions, which could stabilized Cu at site III in the supercages [7]. On that account, the only cationic position of Cu in the supercage is site II, in

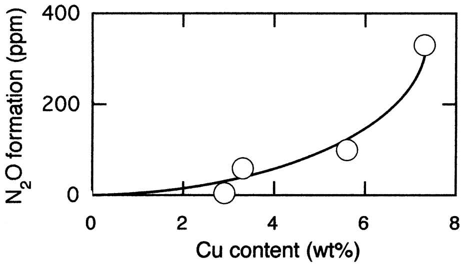

Fig. 7. N_2O formation at 720 K in the SCR of NO by NH_3 on Cu(*x*)-FAU ($x \leq 76$); full NO conversion.

front of the hexagonal window. The separation of sites II in supercages is larger than 0.7 nm and the involvement of two (or more) Cu ions in the same active site can be hardly conceived. In contrast, the formation in sodalite cavities of Cu ions pair, bridge-bonded by O^{2-} has been proposed [23–26]. We may thus postulate that N_2O is likely formed on Cu ions pair, may be $[CuOCu]^{2+}$ dimer species, located in the sodalite cavities. Furthermore, in SCR experiments at 720 K with $^{14}NH_3$/$^{15}NH_3$ switches, the only occurrence of $^{14}N^{15}NO$, with traces of $^{15}N_2O$, provides evidence that N_2O is selectively formed from reaction of NO with NH_3 to yield adsorbed ‘ammonium nitrate-like’ species. The formation of these species is accelerated when NO_2 is present in the feed. Studies to identify the exact nature of these intermediates are in progress.

We proposed above that N_2O would be formed inside the sodalite cavities. It is known that some cations have a great affinity for sitting within the sodalite cavities and hexagonal prisms, and this is true for Ba^{2+} and Ca^{2+} [27]. On that account, a pre-exchange with such a cation will push Cu to be located preferentially inside the supercages, preventing thus the N_2O formation. For that purpose a Cu(35)-Ba(58)-FAU catalyst was prepared. TPR experiments provided evidence of the preferential location of Ba inside the sodalite cavities (Fig. 5 and Table 1). The surface area ratio of the 1st and 2nd peaks in the low temperature region of the TPR profile of Cu(25)-FAU is ca. 0.36, which means that 26% of Cu^{2+} is in the supercages. The TPR profile of Cu(35)-Ba(58)-FAU looks very different with a broad peak showing a maximum at 580 K and a shoulder at ca. 680 K. The deconvolution of this broad feature was undertaken graphically by assuming symmetrical profiles (Fig. 5). The surface area ratio of the 1st and 2nd contributions at 580 and 680 K is 2.90. We interpret this behavior by a much higher proportion of Cu^{2+} in supercages, and by a lower reducibility of Cu^{2+} to Cu^{+}, with respect to Cu(25)-FAU.

The SCR of NO_x on Cu(35)-Ba(58)-FAU is shown on Fig. 8. For pure NO the activity at low temperature is much higher as compared to that found on Cu(25)-FAU and becomes comparable to that of Cu(76)-FAU. This is explained easily since the active Cu species in the range 400–550 K are those located in the supercages, which are the main species in Cu(35)-Ba(58)-FAU. Moreover, when compared to a sample of similar high activity, e.g. Cu(76)-FAU, the N_2O formation is clearly lowered, which was one of the aim of this work. This novel generation of Cu-M-FAU catalysts with dedicated co-cations M has been thus patented [6].

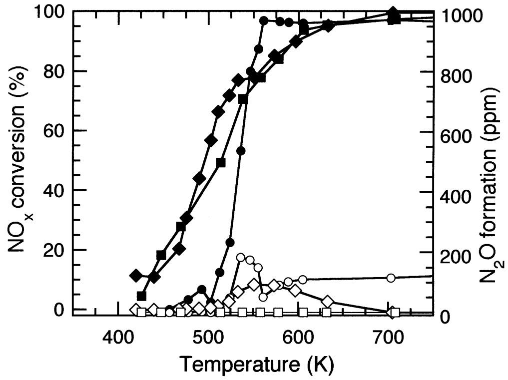

Fig. 8. SCR of NO_x by NH_3 on Cu(35)-Ba(58)-FAU at various NO_x compositions; same symbols as in Fig. 1.

5. Conclusions

Cu-FAU are very active in the SCR of NO_x by NH_3 but two N_2O formations may occur in presence of CuO aggregates and of NNN Cu ions located within the sodalites cages. Moreover, this latter N_2O formation increases with the amount of NO_2 in the feed and results from the interaction of NO_2 with NH_3. The formation of N_2O from 350 to 750 K can be lowered very significantly by dedicated preparation of Cu-FAU based catalysts, and in particular by the occupation of sites in sodalite cages with dedicated co-cations, e.g. Ba.

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