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NO and C_3H_6 adsorption and coadsorption in oxygen excess—A comparative study of different type zeolites modified with gold

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ABSTRACT

Non-acidic NaY, and acidic H-Beta and H-ZSM-5 with various Si/Al ratios were used as supports for gold loaded by deposition-precipitation method. Acidity of the pristine supports and Au/zeolites was estimated from adsorption of pyridine followed by FTIR spectroscopy measurements. NO selective reduction with propene (SCR-HC) was studied by the use of FTIR technique. Adsorption and co-adsorption of the reagents of NO SCR-HC (NO, C_3H_6 and O_2) allowed a discussion of the reaction pathways. The catalytic behaviour of NaY referring to the intermediate formed was not significantly changed by gold modification and Au/NaY did not activate the partial oxidation of propene in the absence of NO. Au particles in Au/Na-Y play an important role in the total oxidation to CO_2. The role of Brønsted acidity in Au/H-Beta and Au/H-ZSM-5 is on the oxidation of NO towards various intermediates (NO_2^-/NO_3^-, N_2O_4, NO^+) and their further interaction with propene, whereas gold species enhance this process by inducing formation of additional active complex (Au^{n+}-NO). Moreover, gold species are responsible for the second reaction pathway via primary partial oxidation of propene to oxygenates and further reaction with NO and R-CN and R-NCO active intermediates as observed on Au/H-Beta and Au/H-ZSM-5.

1. Introduction

Catalytic removal of nitrogen oxide (NO_x) under lean-burn conditions is one of the major challenges in the environmental catalysis research [1]. Selective catalytic reduction (SCR) using hydrocarbons (HC) is a very promising NO_x control technology working in these conditions. In SCR-HC process, the zeolite-based catalysts have received much attention due to their high activity and relatively wide temperature window. It was found that the activity of zeolites is closely related to the type of zeolite and its structure and Cu-ZSM-5 and Co-ZSM-5 catalysts appear to be the most attractive [1].

Gold catalysts have been also studied in the selective reduction of NO_x [2–5]. Haruta and co-workers first reported the effectiveness of supported Au-oxides for this reaction [2]. It was shown that gold supported on ZnO, MgO, TiO_2, Al_2O_3, Fe_2O_3 [2,3] exhibits good activity in the reduction of NO_x with propene producing N_2 as the major product in the temperature range 473–773 K. Among the gold catalysts tested, Au/Al_2O_3 gives the highest conversion to N_2 at high temperature (673 K). The drawback of Au/Al_2O_3 is the fact that it is effective only at relatively high temperatures and is deactivated by SO_2 [2–7]. Gold-modified zeolites could be the alternative catalysts of SCR-HC.

A few possible mechanisms of SCR-HC on Au-catalysts (supported Al_2O_3, TiO_2, MCM-41) have been proposed in the literature [8–10]. According to Ueda et al. [2] the reduction of NO to N_2 on Au/oxides takes place through the formation of NO_2 (oxidation of NO with oxygen) in the first step and the subsequent reduction of NO_2 with C_3H_6. The detailed studies of the SCR-HC reaction mechanism using Fourier transform infrared spectroscopy (DRIFTS) were performed in the groups of Bamwenda [8] and Nguyen [9] over Au/Al_2O_3 and Au/TiO_2 catalysts, respectively. The results obtained confirmed that NO_2 intermediate benefits the NO SCR-HC over Au/Al_2O_3 as a reaction initiator or oxygen transfer agent. NO_2 or its adspecies (NO_x^-) react with activated C_3H_6 to form $C_nH_mN_xO_y$ species, such as –NCO or –CN. In contrast, it was documented that on Au/TiO_2 the first step of the SCR by C_3H_6 comprises the formation of adsorbed oxygenated hydrocarbons (acetate, formate, acetone, and acetaldehyde) and adsorbed nitrate (monodentate, bidentate, and bridging) on the catalyst surface [9]. The interaction of adsorbed oxygenated hydrocarbons with nitrate and/or NO_2 produces nitrogen-containing intermediate compounds such as (–CN) and (–NCO). The presence of nano-sized Au particles is necessary to form oxygenated hydrocarbons, especially acetate species, and it is crucial for production of (–NCO) intermediate compounds. Au species also contribute to the conversion of (–NCO) compounds to N_2.

Oxygen interaction with NO towards active NO_2 was also found as the first step of NO SCR with propene over AuMCM-41 catalyst [10]. It was shown by FTIR studies that NO_2 oxidises propene to

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carboxylate species reducing itself to N_2O at room temperature. At higher temperature (523 K) chemisorbed carboxylate species interact with gaseous NO towards carbonate and N_2O and moreover at 623 K to acetone, CO and CO_2.

The present study focuses on the role of Au particles supported on different zeolite matrices, as well as on the effect of zeolite acidity in the SCR of NO with propene. For this purpose, non-acidic NaY zeolite and acidic H-Beta and H-ZSM-5 were applied. FTIR study of adsorption and co-adsorption of the reagents (NO, C_3H_6 and O_2) allows a discussion of the reaction pathways.

2. Experimental

Y (Si/Al = 2.7, Katalistiks), H-Beta (Si/Al = 12.5, Süd Chemie – Germany) and H-ZSM-5 (Si/Al = 44.9, Süd Chemie – Germany) – types of zeolites were used in this study. The gold catalysts were prepared according to [11] by deposition-precipitation method. The supported gold samples were obtained after filtration and washing, and then dried at 423 K.

The surface properties were characterised by pyridine adsorption followed by FTIR spectroscopy measurements. Infrared spectra were recorded with a Bruker Vector 22 FTIR spectrometer using an in situ cell. Samples of ca. 10 mg cm^{-2} were placed inside the cell and activated under vacuum at 623 K (3 h) and pyridine (PY) was then admitted at 423 K. After saturation with PY the samples were degassed at 423–623 K in vacuum for 30 min. The number of BAS and LAS was calculated after desorption at 473 K using extinction coefficient from Ref. [12].

NO and C_3H_6 adsorption and co-adsorption in the presence of oxygen were performed with the Vertex 70 (Bruker) spectrometer (resolution 4 cm^{-1}). The pressed wafers of the materials (~10 mg cm^{-1}) were placed in the vacuum cell and activated under vacuum at 623 K for 3 h. The experiments were carried out in various ways including different sequences of the reagents admission and heating treatment at 523 and 623 K.

All spectra were recorded at room temperature in the range from 4000 to 400 cm^{-1}. The spectrum without any sample ("background spectrum") was subtracted from all spectra recorded. The IR spectra of the activated samples (after evacuation) were subtracted from those recorded after the adsorption of probe molecules and various treatments. The spectra reported here are the results of this subtraction.

3. Results and discussion

The XRD patterns of gold-containing catalysts (not shown here), with the diffraction peaks characteristic of the Y, Beta and ZSM-5 structures, showed that the framework of parent zeolites is well preserved after deposition-precipitation of gold. The results of XPS and TEM measurements, in detail described in [13], clearly indicated that after calcination of all gold-modified zeolites only metallic gold was present on the surface. The average gold particles sizes estimated from TEM images of all Au–zeolites are similar (9–12 nm). Pyridine was adsorbed in order to evaluate the nature and amount of acidic centres on zeolites surfaces. Fig. 1 presents the ring stretching region of the FTIR spectra after adsorption of pyridine and desorption at 423–623 K over all parent and gold-zeolites studied. Pyridine interacts with Lewis acid sites (LAS) giving rise to characteristic IR bands at ~1450 and ~1620 cm^{-1} [12,14,15]. The bands at ~1550 and ~1640 cm^{-1} are assigned to pyridinium cations formed after pyridine adsorption on Brønsted acid sites (BAS) [12,14,15]. The intensities of the 1450 and 1550 cm^{-1} bands are related to the number of LAS and BAS, respectively. It is clear that the type of zeolite (sodium or hydrogen form and Si/Al ratio) influences the acidity of gold-modified materials. Among gold-containing samples Au/Na-Y, having the highest amount of aluminium, exhibits the highest concentrations of Brønsted acid sites. It indicates that the treatment of Na-Y with chloroauric acid generates BAS (bridged Al–OH–Si – the band at 1550 cm^{-1}), which are not present on the surface of parent NaY. The high number of LAS on NaY originates from Na cations and is deduced from the bands at 1441 and 1615 cm^{-1} typical of the pyridine coordinatively bounded to Na^+ cations [14]. Another band at 1595 cm^{-1} assigned to pyridine adsorbed on LAS (e.g. Al^{3+} cations) [16] or hydrogen-bounded pyridine (together with a band at 1444 cm^{-1}) [15] is also visible in the IR spectra of NaY and Au/NaY. The number of LAS decreases after Au loading. This behaviour supports the partial exchange of sodium cations by protons which generate BAS. In contrast, in hydrogen forms of Beta and ZSM-5 zeolites, modification with gold decreases the number of pristine BAS characterised by a band at ~1545 cm^{-1} from pyridinium cations. It is especially evident in Au/H-ZSM-5 zeolite. Moreover, Au/H-ZSM-5 posses the lowest number of LAS. The bands at 1445 cm^{-1} observed for Au/H-ZSM-5 originates from pyridine adsorbed on Lewis acid sites. On Au/H-Beta two IR bands at 1445 cm^{-1} and 1455 cm^{-1} are distinguished after pyridine adsorption. The first is accompanied by a band at 1596 cm^{-1} and disappears after evacuation at 473 K. Thus it can be assigned to hydrogen bonded pyridine (weakly acidic surface hydroxyl groups). The band at 1455 cm^{-1} originates from pyridine adsorbed on Lewis acid sites formed during dehydroxylation of hydrogen forms of zeolites (defected LAS and Al^{3+} in the framework). The strength of acidity can be deduced from the position of IR bands in the region 1610–1640 cm^{-1}. The higher wavenumber indicates the higher acidic strength. Examination of this region in the spectra of Au/zeolites indicates the following order of the acidic strength: Au/Na-Y (1615 and 1630 cm^{-1} for LAS and BAS respectively) < Au/H-Beta (1623 and 1637 cm^{-1}) ≅ Au/H-ZSM-5 (1625 and 1638 cm^{-1}). In fact, the positions of these bands in the spectra of Au/H-Beta and Au/H-ZSM-5 do not differ too much but the intensity of the bands related to the number of pyridine molecules adsorbed on acidic centres is much higher on Au/H-Beta and the bands are preserved even after evacuation at 623 K.

The properties of zeolites studied in NO SCR-HC process were examined by the consecutive adsorption and co-adsorption of reagents (NO, O_2, C_3H_6). FTIR surface spectra of Au/Na-Y, Au/H-Beta and Au/H-ZSM-5 zeolites after NO + O_2 exposure at RT are shown in Fig. 2A–C. Different NO_x surface species were observed depending on the type of zeolites (Si/Al ratio) and their acidity (sodium or hydrogen forms). In the case of Au/H-Beta and Au/H-ZSM-5 samples (Fig. 2B and C), with high Si/Al ratios, bridging (~1630 cm^{-1}) and chelating nitrates (1580 cm^{-1}) [9,17,18] or nitrites (1420 cm^{-1}) [17] are formed on the surface. Moreover, the bands at 1745 cm^{-1} and ~2130 cm^{-1} appear. The first is assigned to surface N_2O_4 adsorbed on the zeolite acidic hydroxyls [17,19]. Most probably they are formed by dimerization of NO_2 [19]:

$$2NO_2 \leftrightarrow N_2O_4.$$

The disproportionation of N_2O_4 can be a source of $NO_3{}^-$ species:

$$N_2O_4 \rightarrow NO^+ + NO_3{}^-$$

The second band, at ~2130 cm^{-1}, is due to NO^+ species formed or in the above disproportionation reaction or on BAS of H-zeolites in the presence of oxygen according to the following equation [20]:

$$2NO + 1/2O_2 + 2H^+ \leftrightarrow 2NO^+ + H_2O$$

Note that for Au/H-Beta, the sample containing defected LAS, except the 2136 cm^{-1} band, the second band at 2167 cm^{-1} appears that could be attributed to coordinatively bonded NO^+ at cation

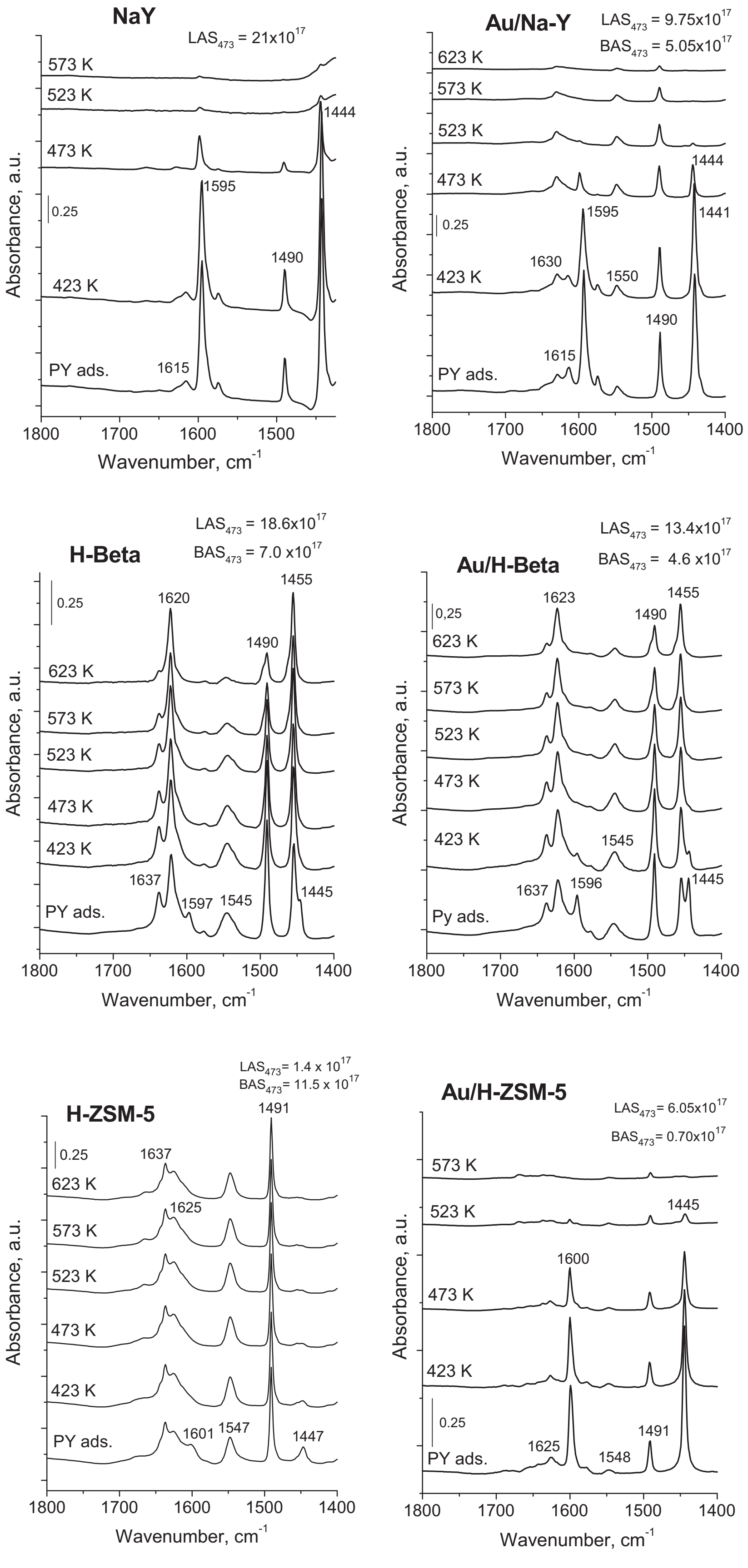

Fig. 1. FTIR spectra of zeolites (recalculated to 10 mg of the material) after adsorption of pyridine at 423 K and desorption at 423–623 K.

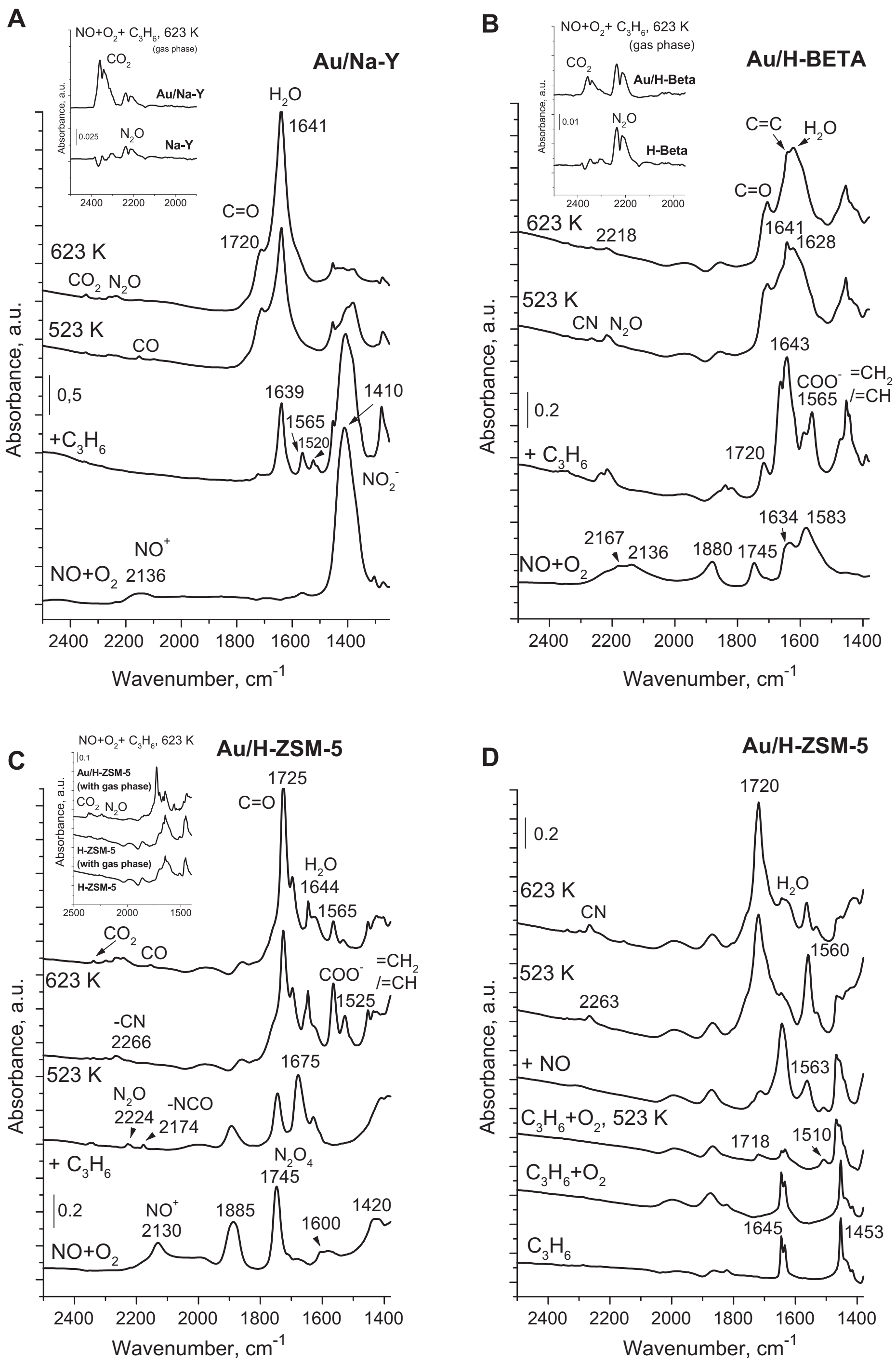

Fig. 2. FTIR spectra of Au–zeolites (recalculated to 10 mg of the material) after the admission of reagents in the following order: NO, O_2, C_3H_6 and heating at 523 and 623 K (A–C); and C_3H_6, O_2, NO and heating at 523 and 623 K (D).

vacancy sites or from [NO^+][N_2O_4] adducts [18]. The same adsorbed species (nitrates/nitrites, N_2O_4, NO^+ species) were formed on pristine supports, H-ZSM-5 and H-Beta, (spectra not shown here). This suggests that Au particles do not contribute to the formation of these adsorbed species. However, it is noteworthy that an additional band assigned to monomeric Au–nitrosyl complex at ~1880 cm^{-1} is clearly seen in the spectra of Au/H-Beta and Au/H-ZSM-5 zeolites. According to Ref. [21] this band can be assigned to NO adsorbed as Au^{n+}-NO ($n \approx 0$) complex. This behaviour indicates that in the excess of oxygen Au^0 is partially oxidised to Au^{n+}.

In contrast to the above, the NO + O_2 admission to Au/Na-Y gives rise to the very intense band at ca. 1410 cm^{-1} (Fig. 2A). It indicates the formation of Na-nitrite species (NO_2^-) [18] resulting from the NO oxidation to NO_2 and electron transfer from the solid to NO_2 molecule. It can be concluded that Na cations can play a role of the storage component for NO_2. It is important to note that also in the spectra of Au/Na-Y zeolite a small intense band at ~2130 cm^{-1} assigned to NO^+ appears in the IR spectra. NO^+ species are formed by the interaction with acidic OH groups generated during gold modification and identified by pyridine adsorption.

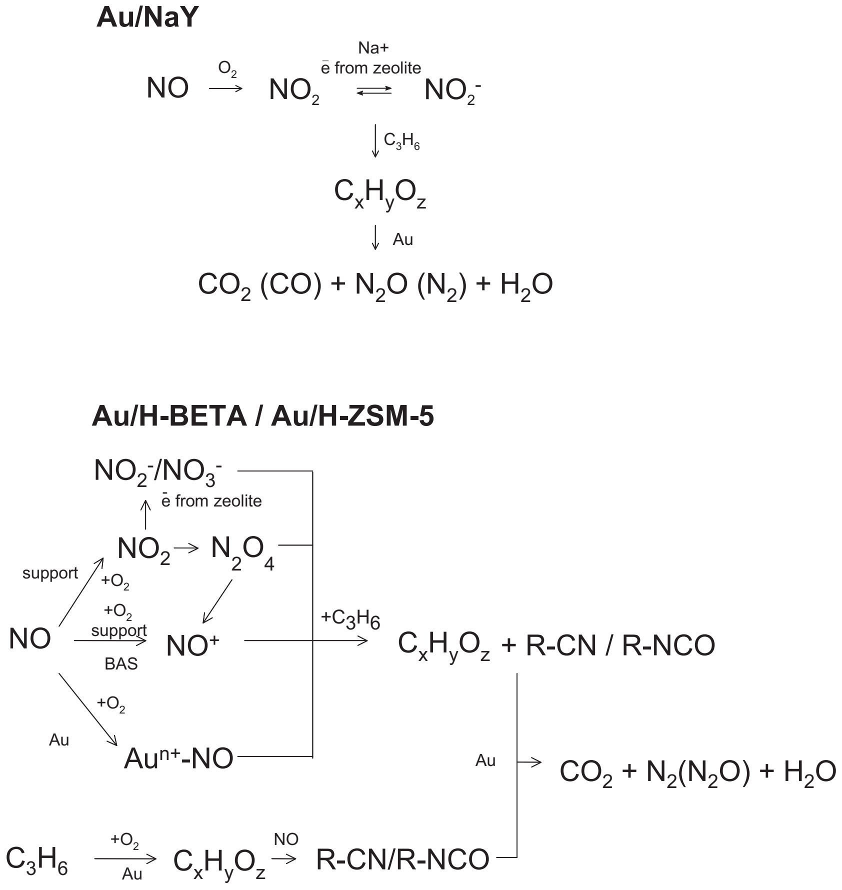

Fig. 3. Schematic diagrams of the SCR reaction mechanism over Au–zeolites.

The introduction of propene after the adsorption of NO followed by O_2 admission, results in the appearance of new IR bands in the spectra of gold-zeolites. These bands originate from oxygenates being products of propene partially oxidation. For Au/Na-Y sample the bands related to acetates groups (1565 cm^{-1}) and carbonates (1520 cm^{-1}) appear simultaneously with a significant decrease in the intensity of the band assigned to Na-nitrites (1410 cm^{-1}). With increasing temperature (523–623 K) the carbonyl groups are formed (1720 cm^{-1}) [9]. The spectra of the gas phase show the presence of CO_2 and N_2O after heating at high temperature (623 K), whereas in the spectra of the zeolite surface the intense band from H_2O (1641 cm^{-1}) is visible. This indicates that total oxidation of propene dominates at this temperature. CO_2 is not formed on parent NaY suggesting that gold species take part in total oxidation to CO_2 (Fig. 2A – gas phase spectra). Sodium cations and nitrate species play important role in the generation of oxygenates which are formed on both samples (without and with gold).

The propene partial oxidation to oxygenates is more effective on Au/H-Beta and Au/H-ZSM-5 catalysts than on Au/NaY. It is because the increase in the strength of acidity enhances the zeolite activity in the formation of intermediates. The new band at 1565 cm^{-1} generated after propene admission is assigned to acetates [9,10,22], whereas that at ~1725 and 1675 cm^{-1} to the ν C=O vibrations of carboxylic groups in acetone (the band at 1725 cm^{-1} from physisorbed acetone and at 1675 cm^{-1} from acetone hydrogen bonded to hydroxyl groups [9,23]). The bands characteristic of NO^+ at ~2130 cm^{-1} and Au^{n+}-NO at 1880 cm^{-1} disappear because of a fast reaction of NO^+ and NO adsorbed on Au^{n+} with C_3H_6. At higher temperatures, 523–623 K, the intensity of the band at 1725 cm^{-1} from acetone strongly increases. It is especially visible for Au/H-ZSM-5 zeolite. Simultaneously, the bands at ~1640 cm^{-1} from adsorbed H_2O and at ~2300 cm^{-1} from adsorbed CO_2 are observed. The appearance of these bands indicates that the total oxidation of C_3H_6 by O_2 takes place. Moreover, after heating at 523 K the band at ~2260 cm^{-1} from the (CN) vibration in acrylonitrile [9,24] appears in the spectra of Au/H-ZSM-5 and Au/H-Beta zeolites. The acid catalysed reaction of NO^+ with C_3H_6 to acrylonitrile was proposed by Gerlach et al. [25] over acidic mordenite. Importantly,–NCO species (the band at 2174 cm^{-1}) are also formed on Au/H-ZSM-5. The cyanide and isocyanate species have been considered as important intermediates in the SCR of NO by hydrocarbons, which are further converted to N_2 [9]. The role of Au particles in the reduction of NO with propene may be deduced from the difference in the spectra of H-ZSM-5 and Au/H-ZSM-5 shown in Fig. 2C (in the insert). After NO and C_3H_6 admission in the presence of oxygen at 623 K only weak band of acetone at 1725 cm^{-1} is observed (together with the band at 1642 cm^{-1} (ν C=C) from unreacted propene) in the spectra of the H-ZSM-5 support comparing to the much intense bands of acetone (1725 cm^{-1}) and acetate (1565 cm^{-1}) in Au/H-ZSM-5′ spectra. Thus, Au particles are essential for the partial oxidation of C_3H_6 towards oxygenated hydrocarbons, similarly as it was found for Au/TiO_2 [9]. Moreover, gold species take part in total oxidation to CO_2 (Fig. 2 – gas phase spectra) and H_2O (Fig. 2 – surface spectra).

The different sequences (C_3H_6, followed by O_2 and NO) of reagents admission onto Au–zeolites leads to the different scenarios. After the admission of C_3H_6, the spectra of zeolites show the typical bands in 3000–2800 cm^{-1} range assigned to adsorbed propene and at ~1640 cm^{-1} (ν C=C; a band of physisorbed propene), ~1450 cm^{-1} (ν =CH_2 and =CH physisorbed and weakly chemisorbed on different centres) and ~1380 cm^{-1} (δs –CH_3) [26]. The admission of O_2 does not change the FT-IR spectra indicating that the oxidation of C_3H_6 does not occur under these conditions. However, in the case of Au/H-Beta and Au/H-ZSM-5 (Fig. 2D) the admission of $C_3H_6 + O_2$ and heating at 523 K leads to partial oxidation of propene to formates (small band at 1510 cm^{-1} assigned to carboxylate in formate ion [22]) and acetone ($\sim$1720 cm^{-1} – only on Au/H-ZSM-5). These oxygenates interact with NO towards nitrogen-containing intermediate compounds (–CN, –NCO). It is worth noting that after NO admission onto gold-zeolites, the bands from NO^+ and $Au^{\delta+}$-NO are not observed indicating that the pre-adsorbed propene occupies the sites active for NO adsorption. This is why the number of NO chemisorbed decreases and the interaction between NO and propene becomes weaker.

In summary, reaction pathways on Au/Na-Y, Au/H-Beta and Au/H-ZSM-5 can be proposed (Fig. 3). NO is oxidised with O_2 to NO_2 on Au/Na-Y and stored in the form of nitrites on Na cations. Nitrite species are active in the interaction with propene towards total oxidation products (CO_2, H_2O) *via* generation of acetates and carbonates. In contrast, on Au/H-Beta and Au/H-ZSM-5 zeolites NO was adsorbed on Au^{n+} and NO^+, N_2O_4, NO_3^- species formed after NO adsorption in the presence of oxygen interacted with propene towards oxygenates adsorbed (acetone, acetate, carbonates) and oxidised further to CO_2 and H_2O. Au particles play an important role in the partial and total oxidation of C_3H_6. The interaction between propene and/or oxygenates and different forms of adsorbed NO species (NO, NO^+, N_2O_4 and/or nitrate/nitrite) produces (–NC) and (–NCO) compounds which are then converted to N_2. It is important to note that on Au/H-Beta and Au/H-ZSM-5 the parallel reaction path could be the C_3H_6 oxidation to oxygenates (acetone, formate), which further interact with NO. Taking into account the selectivity to N_2, deduced from (–NC) and (–NCO) formation, Au/H-Beta and Au/H-ZSM-5 zeolites with high Si/Al ratio and high strength of acid sites, are the most attractive catalysts for SCR of NO with propene. It indicates that the strength of Brønsted acid sites (BAS) on the surface (depending on Si/Al ratio) is an important parameter in the SCR process. Among these two catalysts, Au/ZSM-5 could be even better in NO SCR-HC because not only R-CN but also R-NCO species are formed, which are commonly agreed as active intermediates.

4. Conclusions

The results presented allowed us to estimate the chemisorbed species after the admission of NO HC-SCR reagents and to observe their interaction on gold catalysts supported on zeolite matrices. It is concluded from the FTIR study combined with NO, O_2 and C_3H_6 co-adsorption that the type of zeolite support and the strength of BAS influence the intermediates formed and their further transformations. There is no significant difference between the intermediates (NO_2^- and $C_xH_yO_z$) formed on NaY and Au/NaY suggesting that sodium cations play essential role in their generation. Au particles in Au/Na-Y play an important role in the total oxidation to CO_2. Acidic H-ZSM-5 and H-Beta are active in the formation of NO_2^-/NO_3^-, N_2O_4, NO^+, R-CN, $C_xH_yO_z$ proposed as intermediates in NO SCR-HC. Modification of these zeolites with gold decreases the number of BAS, increasing the acidity strength. This behaviour and the presence of gold induce the formation of R-NCO on Au/ZSM-5 in the reaction of Au^{n+}-NO with $C_xH_yO_z$ and enhance the activity in SCR-HC. Without the presence of gold the reaction pathway through partial oxidation of C_3H_6 does not occur. Thus, the presence of gold induces the oxidation of C_3H_6 towards $C_xH_yO_z$ reacting further with NO. Two parallel reaction pathways, by oxidation of NO or partial oxidation of propene in the first step, are proposed on gold/H-Beta and H-ZSM-5.

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References

[1] Z. Liu, S.I. Woo, Catal. Rev. 48 (2006) 43–89.
[2] A. Ueda, T. Oshima, M. Haruta, Appl. Catal. B 12 (1997) 81–93.
[3] A. Ueda, M. Haruta, Appl. Gold Bull. 32 (1999) 3–11.
[4] E. Seker, E. Gulari, Appl. Catal. A 232 (2002) 203–217.
[5] D. Niakolas, C. Andronikou, C. Papadopoulou, H. Matralis, Catal. Today 112 (2006) 184–187.
[6] N. Jagtap, S.B. Umbarkar, P. Miquel, P. Granger, M.K. Dongare, Appl. Catal. B 90 (2009) 416–425.
[7] J. Li, Y. Zhu, R. Ke, J. Hao, Appl. Catal. B 80 (2008) 202–213.
[8] G.R. Bamwenda, A. Obuchi, S. Kushiyama, K. Mizuno, Stud. Surf. Sci. Catal. 130 (2000) 1271–1276.
[9] L.Q. Nguyen, Ch. Salim, H. Hinode, Appl. Catal. B 96 (2010) 299–306.
[10] I. Sobczak, A. Kusior, M. Ziolek, Catal. Today 137 (2008) 203–208.
[11] J.-H. Chen, J.-N. Lin, Y.-M. Kang, W.-Y. Yu, Ch.-N. Kuo, B.-Z. Wan, Appl. Catal. A: Gen. 291 (2005) 162–169.
[12] S. Khabtou, T. Chevreau, J.C. Lavalley, Micropor. Mater. 3 (1994) 133–148.
[13] I. Sobczak, H. Pawlowski, J. Chmielewski, M. Ziolek, J. Hazard. Mater. 179 (2010) 444–452.
[14] M.R. Basila, T.R. Kantner, K.H. Rhee, J. Phys. Chem. 68 (1964) 3197–3207.
[15] G. Busca, Catal. Today 41 (1998) 191–206.
[16] G. Busca, in: S.D. Jackson, J.S.J. Hargreaves (Eds.), Metal Oxide Catalysis, Wiley-VCH, 2008.
[17] K. Hadjiivanov, Catal. Rev. Sci. Eng. 42 (2000) 71–144.
[18] Ch. Sedlmair, B. Gil, K. Seshan, A. Jentys, J.A. Lercher, Phys. Chem. Chem. Phys. 5 (2003) 1897–1905.
[19] J. Szanyi, M.T. Paffett, J. Catal. 164 (1996) 232–245.
[20] K. Hadjiivanov, J. Saussey, J.L. Freysz, J.C. Lavalley, Catal. Lett. 52 (1998) 103–108.
[21] T.M. Salama, R. Ohnishi, T. Shido, M. Ichikawa, J. Catal. 162 (1996) 169–178.
[22] T.E. Hoost, K.A. Laframboise, K. Otto, Appl. Catal. B: Environ. 7 (1995) 79–93.
[23] J. Datka, P. Kozyra, E. Kukulska-Zajac, W. Kobyzewa, Catal. Today 101 (2005) 117–122.
[24] F. Poignant, J.L. Freysz, M. Daturi, J. Saussey, Catal. Today 70 (2001) 197–211.
[25] T. Gerlach, F.-W. Schutze, M. Baerns, J. Catal. 185 (1999) 131–137.
[26] J. Datka, E. Kukulska-Zajac, J. Phys. Chem. B 108 (2004) 17760–17774.