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# New water-tolerant supported molten indium catalyst for the selective catalytic reduction of nitric oxide by ethanol

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The catalytic activity and selectivity of indium supported on controlled pore glass (In-CPG-SMMC) were investigated and compared with those of H-ZSM-5 and In/$\gamma$-$Al_2O_3$ for the selective catalytic reduction of nitric oxide by ethanol under net oxidizing conditions, in the absence and presence of water vapors. Even though the support of In-CPG-SMMC is almost pure silica (94–99% $SiO_2$, 1–6% $B_2O_3$, 0.05–0.3% $Na_2O$), the activity of this catalyst was found to be comparable with that of the conventional catalysts. The presence of steam in the feed enhanced catalyst activity over the entire temperature range studied. At temperatures above 500 °C the ability of H-ZSM-5 and In/$\gamma$-$Al_2O_3$ to reduce NO to $N_2$ was surpassed by the supported indium catalyst.

**Keywords:** NO SCR, molten indium catalyst, controlled pore glass, ethanol

## 1. Introduction

Research has been under way for many years to find viable catalysts that selectively reduce nitrogen oxides in an oxidizing environment. Since the first reports of the high selective catalytic reduction (SCR) activity of copper [1,2] and other cation ($Fe^{3+}$, $Co^{3+}$, $In^{3+}$, $Ga^{3+}$)-exchanged zeolites [3–6], a large variety of other catalysts have been tested as candidates for this reaction. Literally thousands of catalysts have been investigated, but despite this substantial work, a suitable catalyst has not yet been developed.

Unfortunately, over the zeolite-based catalysts, which have so far been found to be the most active ones, the non-selective combustion by oxygen begins to dominate over $NO_x$ reduction above 400 °C, and the nitrogen oxides conversion concomitantly begins to fall. Another major disincentive in using zeolitic catalysts commercially is their low hydrothermal stability. Water vapors also suppress their catalytic activity. The unavoidable presence of water vapor in high concentrations typical of combustion effluents, thus, make the practical application of zeolitic catalysts impossible. For example, Kikuchi and Yogo [3] showed that the catalytic activity of Ga-, Co-, In-, and H-ZSM-5 catalysts significantly decreases when 10% $H_2O$ is present in the feed. Another serious disadvantage of Cu- and zeolite-based catalysts is their vulnerability to deactivation by sulfur oxide poisoning as well as due to coking. Because of their strong acidity, the formation of coke is unavoidable especially when the reductant is a reactive olefin. Thus, d'Itri et al. [7] observed that the difference in effectiveness of a Cu–H-ZSM-5 is due to the different degrees of catalyst deactivation by carbonaceous deposits and that in the presence of propene more coke is formed than in the presence of propane.

Developing a non-zeolitic, commercially usable, catalyst for the NO abatement of emissions from both motor vehicles and stationary sources under net oxidizing conditions remains a challenge. Single metal oxides such as $Al_2O_3$ [8], $In_2O_3$ [9,10], and $Ga_2O_3$ [11] or Ag [12] supported on alumina have also been found to catalyze this reaction. It has recently been demonstrated [13] that $In_2O_3$ and $Ga_2O_3$ supported on a specially prepared "sol–gel" $\gamma$-$Al_2O_3$ outperforms all other materials, including existing commercial $\gamma$-$Al_2O_3$ supports. These authors found that $In_2O_3$ dispersed on $SiO_2$ is less active than the unsupported $In_2O_3$ [13]. Another $SiO_2$ support has been reported to eliminate the catalytic activity of $SnO_2$ for the SCR of NO by methanol [14]. The poor performance of $SiO_2$ supports for numerous transition-metal-based SCR catalysts has also been noted [10]. Since the acidic supports, such as $Al_2O_3$, $TiO_2$, and $Al_2O_3$–$SiO_2$, are active SCR catalysts themselves [10,14–17], while the largely inert $SiO_2$ is not [10,14,18], it is generally agreed that only acidic Brønsted or Lewis oxides are suitable supports for the SCR process.

In this study, we investigated the catalytic performance of indium supported as a molten metal on controlled pore glass (In-CPG-SMMC) for the selective catalytic reduction of nitric oxide by ethanol by comparing it with H-ZSM-5 and indium supported on gamma-alumina catalysts, under net oxidizing conditions in the absence and presence of water vapors. Even though the support in the In-CPG-SMMC is almost pure silica (94–99% $SiO_2$, 1–6% $B_2O_3$, 0.05–0.3% $Na_2O$) [19], its catalytic activity for the SCR of NO to $N_2$ was found to be comparable with that of the other two catalysts. The presence of steam in the feed

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enhanced the catalyst performance over the entire temperature range. Furthermore, at higher temperatures of practical interests, the ability of H-ZSM-5 and In/$\gamma$-$Al_2O_3$ to reduce NO to $N_2$ was surpassed by the In-CPG-SMM catalyst.

## 2. Experimental

### *2.1. Catalysts and materials*

Supported indium catalyst (In-CPG-SMMC) was prepared by the direct imbibition of the molten metal on support. The support (figure 1), a controlled pore glass (CPG75-120) (BET surface area = 171 $m^2$/g, and an average pore radius of 64 Å) from Sigma Chemical Inc. was heated in air at around 150 °C to remove adsorbed water. Then a known amount of metallic In (Aldrich 99.999% purity) was introduced into the test tube together with the support. The system was heated above the melting point of In (156.4 °C) while continuously and vigorously stirring the contents of the test tube, to facilitate the uptake and dispersion of the metal into the support pores. The resulting catalyst was a 28% In on porous glass (figure 2), with a final BET surface area of 138 $m^2$/g and an average pore diameter of 71 Å, containing nano- to micro-droplets of In (diameter $<1.43$ $\mu$m) in molten state at the catalytic reaction temperature. The loading in this study was arbitrarily chosen to correspond to a monolayer coverage of In, which is 28 wt%. Other metal loadings are being currently investigated. Details on the characterization of this catalyst are given elsewhere [20].

H-ZSM-5 catalyst (Si/Al ratio = 25) was procured from Mobil Research and Development Corporation. Prior to use, it was calcined for 1 h in oxygen at 550 °C to remove water and residual organic templates remaining from its synthesis.

The In loading on the gamma-alumina support was 24 wt%. This sample was prepared by impregnating $\gamma$-$Al_2O_3$ obtained from United Catalysts Inc. (average particle diameter 2.5–5 mm, BET surface area = 202 $m^2$/g) with an aqueous solution of In(III) nitrate pentahydrate (Strem Chemicals). The catalyst was dried at 120 °C for 3 h, then calcined at 600°C for 12 h in air.

High-purity gases were used in all catalytic experiments. Hydrogen (99.99%) and helium (99.995%) from Air Products were further purified with Oxy-Purge $^{TM}$N purifiers from Matheson. Mixtures of 0.4% $C_3H_6$ in helium and 0.4% NO in helium were obtained from Matheson and were used as received. Ethanol (99.5%) was procured from Pharmaco Products.

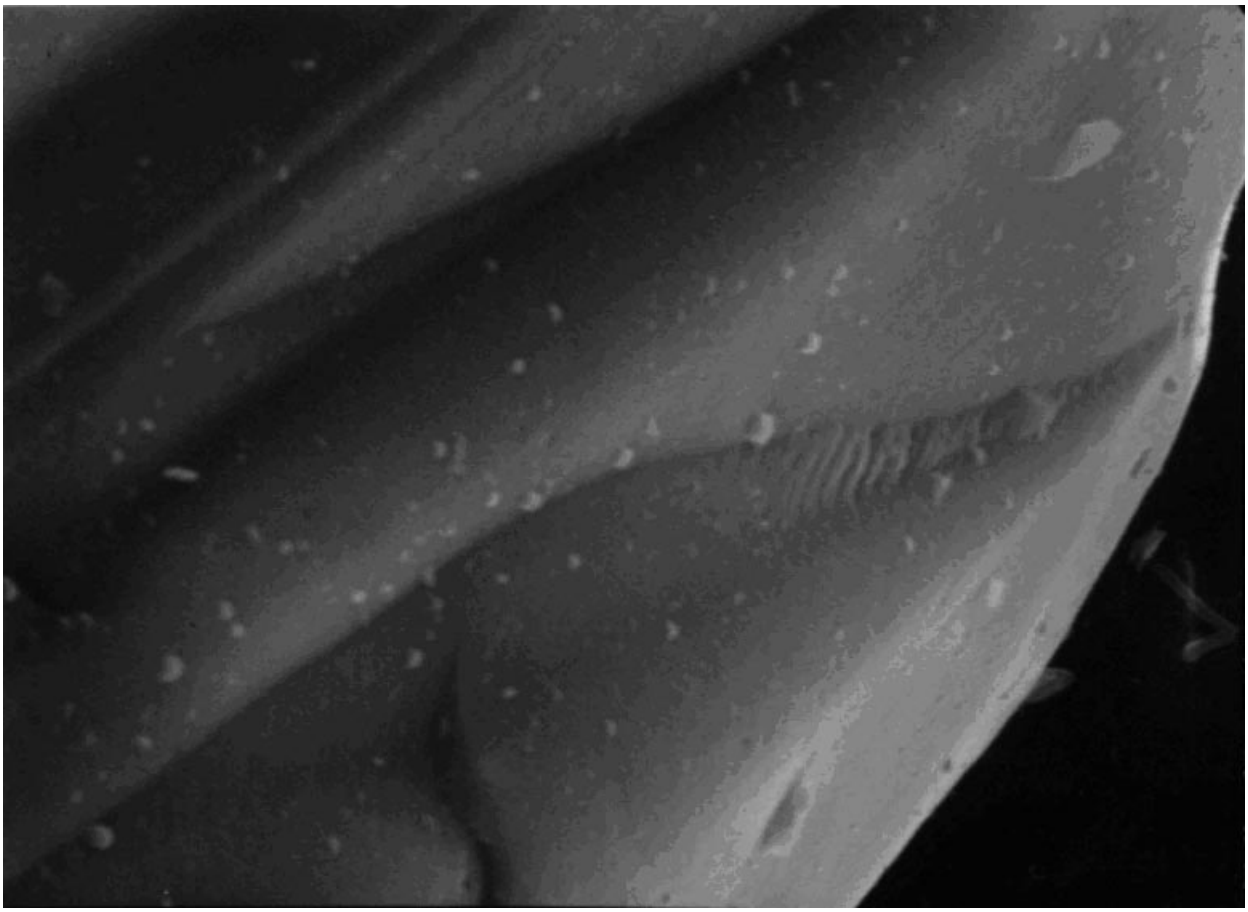

Figure 1. SEM picture of controlled porosity glass (CPG), $\times$7000 magnification, $V_{acc} = 5$ kV.

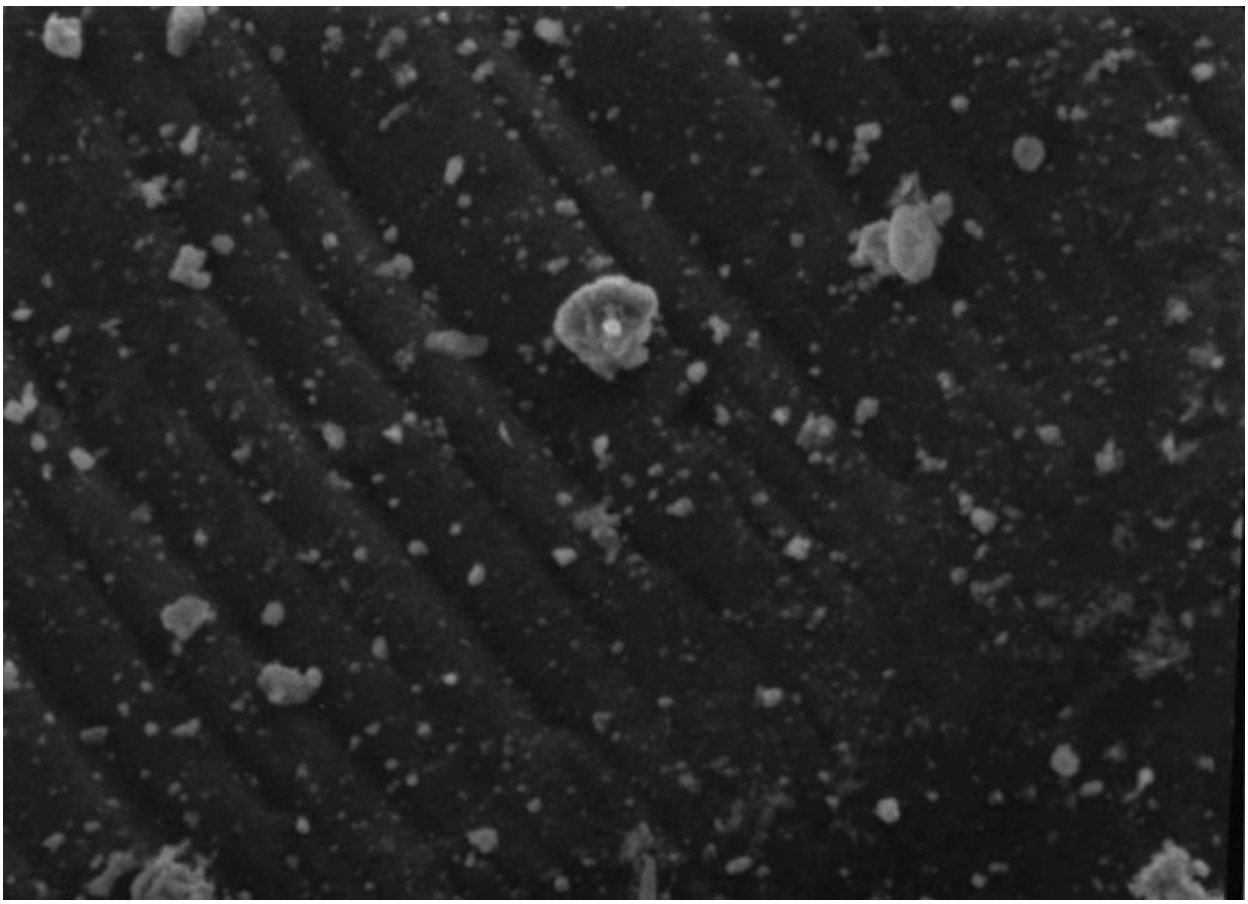

Figure 2. SEM picture of 28% In-CPG-SMMC, $\times$7000 magnification, $V_{acc} = 5$ kV. Indium is visible as microdroplets of diameter $<$1.43 $\mu$m.

### 2.2. Catalyst characterization

The specific surface area (BET) was determined by the standard nitrogen adsorption method at $-196\,^{\circ}$C after evacuation at $10^{-6}$ Torr for 1 h. Scanning electron microscope pictures were taken using a Hitachi S4000 SEM at an accelerating voltage of 5 kV. The samples were carbon coated to increase conductivity and to avoid cluttering of elemental lines to X-ray microanalysis spectra. The phase purity and composition of the In-CPG before and after reaction were analyzed by powder X-ray diffraction using a Siemens D5000 diffractometer equipped with an energy-sensitive detector (Cu $K_{\alpha 1,2}$ source). Peaks were matched with those in the corresponding computerized JCPDF file.

### 2.3. Catalytic experiments

The catalytic reactions were performed in a fixed-bed catalytic reactor consisting of a U-shaped fused quartz reactor. The catalyst samples (0.1 $cm^3$) were placed between two quartz wool plugs. Before the first catalytic measurement, the 28% In-CPG and the 24% In/$\gamma$-$Al_2O_3$ catalysts were evacuated at $10^{-6}$ Torr at 120 °C for 30 min, then heated at 550 °C for another 30 min. Finally these catalysts were treated in a flow of 100 $cm^3$/min $H_2$ for 60 min at 500 °C, then cooled to the reaction temperature in He flow. The H-ZSM-5 catalyst was evacuated at 250 °C, then calcined at 600 °C in 100 ml/min flow of He for 12 h, and finally cooled to the reaction temperature. Between successive catalytic measurements, the catalysts were kept under He at room temperature.

Catalytic activities were measured in terms of $NO_x$ conversion at steady state, after 2–3 h on stream, at different feed temperature at a constant gas-hourly space velocity (GHSV) of 60,000 $h^{-1}$, in excess oxygen, in the absence and presence of water vapors. The inlet gas contained 1000 ppm NO, 5% $O_2$, 0.2% $C_3H_6$ or 0.65% ethanol, and 0 or 10% $H_2O$ vapors, with balance He, giving a total flow rate of 100 $cm^3$/min. The reaction temperature was increased stepwise (50 °C increment) from 350 to 550 °C using an Omega type temperature controller (CN76000).

The gaseous reactants were introduced into the reactor using MKS type 1179 mass flow controllers. The alcohol was fed to the reactor via a saturator set at 0 °C, while water was fed with a liquid pump (Gilson model 302, minimum flow rate 0.5 $\mu$l/min). Both water and alcohol were vaporized with a heater located at the reactor inlet. Water vapors and unreacted ethanol in the reactor outlet stream were trapped by a condenser and by Drierite (97% $CaSO_4 + 3\%$ $CoCl_2$) contained in a 100 ml glass column. The concentrations of $NO_x$ ($NO + NO_2$) and NO in the outlet gas streams were measured continuously by passing the outlet gas stream through a chemiluminescent NO/$NO_x$ analyzer

(Rosemount model NGA 2000), and collecting data every 20 s. The overall NO conversion was calculated as the difference between the inlet NO concentration (obtained before each experiment by running the feed through a by-pass line connected to the inlet of the $NO_x$ analyzer) and the averaged value of several hundreds (or even thousands) of measurements of the outlet NO concentration, divided by the inlet NO concentration and multiplying by 100. Since the formation of $N_2O$ was not monitored, the conversion of NO to $N_2$ (obtained as the difference of total conversion of NO minus conversion of NO to $NO_2$) also includes the conversion of NO to $N_2O$. Since $N_2O$ contains paired nitrogen, there is no danger of the reverse reaction back into NO during any further $N_2O$ decomposition when required.

The distribution of carbon-containing species was also analyzed using a gas chromatograph (SRI 8610C) with both FID and TCD detectors, equipped with a Porapak Q column (100–120 mesh, 10 ft. long, stainless steel, 100 °C). The effluent stream was routed to the GC by bypassing the water and unreacted ethanol trap. The carbon-containing species monitored on the GC were CO, $CO_2$, $CH_4$, $C_2H_4$, $CH_3CHO$, $CH_3CN$, and $C_2H_5OH$.

## 3. Results and discussion

Table 1 shows that the total NO conversion on 28% In-CPG-SMMC was very small when $C_3H_6$ was used as reductant. Independent studies conducted in our laboratory revealed that the indium-based catalyst is extremely active for the total oxidation of ethylene to $CO_2$ and $H_2O$, over a broad temperature range. It is, thus, possible that $C_3H_6$ was completely burned by oxygen, rather than participating in the SCR of NO. H-ZSM-5 was found to be most active for the SCR of NO at temperatures below 500 °C, however, its SCR activity fell drastically above 500 °C, when it became highly active for the total oxidation of the hydrocarbon. It is also noteworthy, as table 1 shows, that on H-ZSM-5 21% of NO was reduced to $N_2$ at 400 °C even when no reductant was present in the feed, while the In-CPG-SMMC showed a very low activity (1.7% conversion) for this. However,

Table 1
Catalytic activities for the SCR of NO in the presence of excess oxygen over H-ZSM-5, 24% In/$\gamma$-$Al_2O_3$ and 28% In-CPG-SMMC, in the absence and presence of reductants.[a]

| Catalyst | Temp. (°C) | Water (%) | Without reductant | | With reductant | | | |
|---|---|---|---|---|---|---|---|---|
| | | | Total conv. | Conv. to $N_2$ | $C_3H_6$ (0.2%) | | $C_2H_5OH$ (0.65%) | |
| | | | | | Total conv. | Conv. to $N_2$ | Total conv. | Conv. to $N_2$ |
| H-ZSM-5 (Si/Al 25) | 350 | 0 | 18 | 0 | 25 | 25 | 0 | 0 |
| | | 10 | nm[b] | nm | nm | nm | 7.5 | 7.5 |
| | 400 | 0 | 38 | 21 | 26.3 | 26.3 | 38 | 30 |
| | | 10 | | | | | 17.3 | 9 |
| | 450 | 0 | 29.3 | 9.7 | 54 | 54 | 71 | 65 |
| | | 10 | | | | | 33.3 | 33.3 |
| | 500 | 0 | 28 | 3 | 36.2 | 36.2 | 52 | 20 |
| | | 10 | | | | | 21 | 15.3 |
| | 550 | 0 | 20 | 5 | 28 | 28 | 18.8 | 10 |
| | | 10 | | | | | 8 | 8 |
| 28% In-CPG-SMMC | 350 | 0 | 4 | 1 | 0 | 0 | 0 | 0 |
| | | 10 | nm | nm | nm | nm | 9.6 | 3.6 |
| | 400 | 0 | 2 | 1.7 | 1.5 | 0 | 0 | 0 |
| | | 10 | | | | | 10 | 5.5 |
| | 450 | 0 | 2 | 0.5 | 2 | 0 | 12.8 | 10.3 |
| | | 10 | | | | | 18.3 | 12.5 |
| | 500 | 0 | 1.3 | 0.5 | 8.9 | 0 | 25 | 16.5 |
| | | 10 | | | | | 39 | 23 |
| | 550 | 0 | 0 | 0 | 0 | 0 | 36 | 22.5 |
| | | 10 | | | | | 71 | 45 |
| 24% In/$\gamma$-$Al_2O_3$ | 350 | 0 | nm | nm | nm | nm | 13.5 | 9.7 |
| | | 10 | | | | | 10.7 | 8.7 |
| | 400 | 0 | nm | nm | nm | nm | 25.6 | 20 |
| | | 10 | | | | | 19.5 | 17.2 |
| | 450 | 0 | nm | nm | nm | nm | 30 | 28 |
| | | 10 | | | | | 25.8 | 23.5 |
| | 500 | 0 | nm | nm | nm | nm | 21.7 | 16.6 |
| | | 10 | | | | | 16.8 | 13.6 |
| | 550 | 0 | nm | nm | nm | nm | 11.4 | 7.25 |
| | | 10 | | | | | 6.2 | 4.5 |

[a] Feed: 1000 ppm NO, 5% $O_2$, 0.2% $C_3H_6$ or 0.65% $C_2H_5OH$, + 0 or 10% $H_2O$, balance He. GHSV = 60,000 $h^{-1}$, $P = 101.3$ kPa.
[b] nm = not measured.

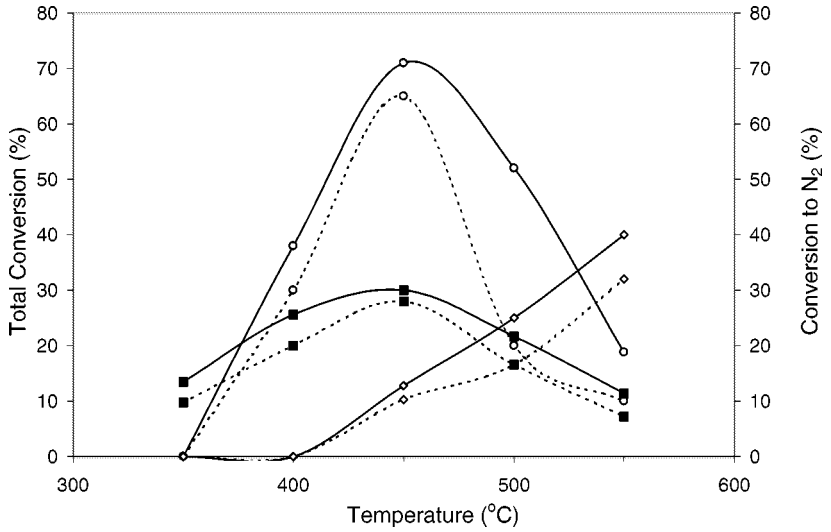


Figure 3. Total NO conversion (solid lines) and conversion to $N_2$ (dashed lines) over H-ZSM-5 (○), 24% In/$\gamma$-$Al_2O_3$ (■), and 28% In-CPG-SMMC (◊) under dry conditions. Feed: 1000 ppm NO, 5% $O_2$, 0.65% $C_2H_5OH$, balance He, GHSV = 60,000 $h^{-1}$.

the indium catalysts are very active with ethanol as the reductant.

Figure 3 compares the catalytic activities of the H-ZSM-5 and both the indium-based catalysts for the SCR of NO in the presence of excess oxygen with ethanol as the reductant, in the absence of water vapors. Both H-ZSM-5 and the 24% In/$\gamma$-$Al_2O_3$ catalysts had a higher catalytic activity at temperatures below 500 °C than the In-CPG-SMMC. However, this latter catalyst surpassed the activity of the other two at temperatures above 500 °C. The rate of the NO-SCR reaction going through a maximum with rising temperature is a common feature for zeolites and alumina-supported catalysts. The narrow range of temperatures in which these conventional catalysts are active constitutes a major impediment for their commercial use. Interestingly, the activity of In-CPG appears to increase monotonically with increasing temperature in the range investigated, making it potentially attractive for higher temperatures applications. SCR with In-CPG-SMMC at even higher temperatures are now in progress. Controlled pore glasses maintain their structure up to temperatures as high as 650 °C [19]. However, a further temperature increase brings about a decrease of the mean pore diameter and porosity.

It has been suggested before, that the SCR reaction pathway might change depending on the catalyst used [21]. Comparative experiments with different feed mixtures of (EtOH + NO), (EtOH + $O_2$), (NO + $O_2$) and (EtOH + $O_2$ + $H_2O$) indicate that the bimolecular reactions of EtOH with $O_2$ and/or NO are likely initial steps in the SCR of NO by ethanol over In-CPG-SMMC [22]. Experiments [22] indicate that the direct decomposition and direct oxidation of ethanol by oxygen are faster reactions than the oxidation of NO to $NO_2$ by $O_2$ over this catalyst. Consequently, the formation of $NO_2$ is probably not an essential step in the SCR of NO over In-CPG-SMMC, but rather either the partial oxidation of ethanol or the direct reaction of ethanol with NO possibly initiates the SCR process. Such a mechanism has been proposed before for the HC SCR of NO over Cu–H-ZSM-5 and Pt/$Al_2O_3$ catalysts [21].

Figures 4 and 5 show the carbon species distribution on 24% In/$\gamma$-$Al_2O_3$ and 28% In-CPG, respectively, when no water is present in the feed. Thus, nitrogen and $NO_2$ were produced along with $CO_2$, CO, $CH_3CHO$, $CH_3CN$, $CH_4$ and traces of $C_2H_4$. However, we were not able to detect HCN, which is formed in relatively high concentrations on a Ag/$Al_2O_3$ catalyst [23]. The formation of $CH_3CHO$ increases with temperature and goes through a maximum at around 400 °C over In/$\gamma$-$Al_2O_3$. Separate experiments indicate that ethanol readily reacts over 28% In-CPG-SMMC even in the absence of other reactants at very high space velocities, and the major product is acetaldehyde. At 400 °C the effect of $O_2$ and NO on the reactivity of ethanol was studied [22]. Addition of small amounts of NO has an even more pronounced effect on the total conversion of ethanol than addition of 4% $O_2$, when the conversion of ethanol was 12% as compared to 20%. The two oxidants added together result in 25% EtOH conversion, which is higher than the conversion obtained when either oxidants are fed individually.

At temperatures above 500 °C, ethanol is completely burned to $CO_2$ and $H_2O$ (few ppm of $CH_3CN$ and $CH_4$) over the In/$\gamma$-$Al_2O_3$ catalyst (figure 4), while 370 ppm acetaldehyde are still formed over In-CPG-SMMC at 550 °C (figure 5).

Figure 6 compares the activities of the three catalysts when 10% water was fed to the reactor, under conditions otherwise identical to those in figure 3. A comparison to figure 3 shows that, while water vapors clearly quenched the activity of H-ZSM-5 over the entire temperature range, they curiously had a dramatic enhancing effect on the activity of In-CPG-SMMC. H-ZSM-5 and other cation-exchanged zeolites have been found to be very sensitive to water vapor. It has been suggested [3,24] that water competes with NO for adsorption sites, thus lowering the NO surface cov-

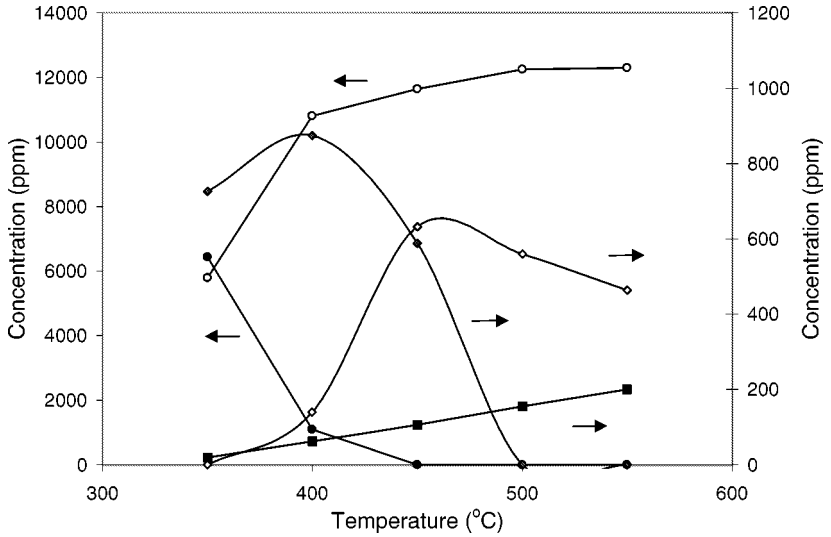


Figure 4. Distribution of carbonaceous species in the products of the reduction of NO by ethanol over 24% In/$\gamma$-$Al_2O_3$ in the absence of water in the feed. Feed: 1000 ppm NO, 5% $O_2$, 0.65% $C_2H_5OH$, balance He, GHSV = 60,000 $h^{-1}$. (●) 2 × $C_2H_5OH$, (♦) 2 × $CH_3CHO$, (■) $CH_4$, (◇) 2 × $CH_3CN$, and (○) $CO_x$.

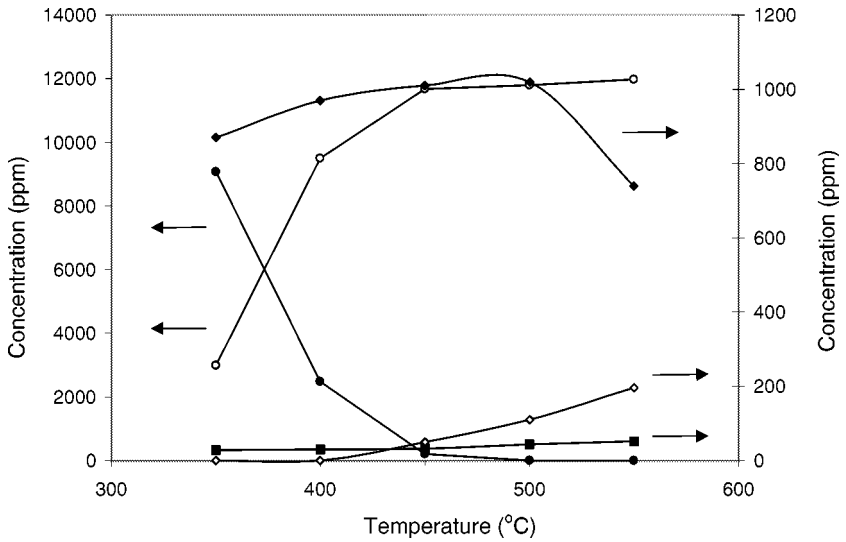


Figure 5. Distribution of carbonaceous species in the products of the reduction of NO by ethanol over 28% In-CPG-SMMC in the absence of water in the feed. Feed: 1000 ppm NO, 5% $O_2$, 0.65% $C_2H_5OH$, balance He, GHSV = 60,000 $h^{-1}$. (●) 2 × $C_2H_5OH$, (♦) 2 × $CH_3CHO$, (■) $CH_4$, (◇) 2 × $CH_3CN$, and (○) $CO_x$.

erage. Thus, TPD data [24] indicates that the amounts of NO adsorbed on partially dried Co-ZSM-5 are significantly smaller than on the fully dried sample. It is conceivable that water moderates the acidity of H-ZSM-5 and $\gamma$-$Al_2O_3$, thus reducing NO adsorption. In contrast, water enhanced the catalytic activity of In-CPG-SMMC over the entire temperature range. To the best of our knowledge, In-CPG-SMMC is the most active catalyst for the SCR of NO among the siliceous catalysts, its activity being comparable, and its hydrothermal stability superior to that of the more conventional catalysts. This behavior is most probably associated with the surface chemistry of the CPG that appears to be more complex than that of the silica gel surface. The In-CPG-SMMC seems to have no aging problems since the catalytic performance remains intact when catalytic tests were carried out at different reaction conditions for extended periods of time.

The infrared spectroscopy shows the presence of surface Si–OH groups together with surface B–OH groups [19], as well as free boron atoms of strong electron acceptor properties resulting from the residue of $B_2O_3$ (1–6%) which remains in the siliceous structure of CPG after its preparation procedure. Due to the neighboring boron atoms, hydroxyl groups on CPG surface represent strong Brønsted acidic sites, their presence increasing considerably the adsorption properties of CPG compared to silica gel. Investigations of the thermal treatment of CPG by means of infrared spectroscopy proved [19] the enrichment of its surface in boron atoms migrating from the interior of the siliceous CPG skeleton and suggested the formation of borate clusters on

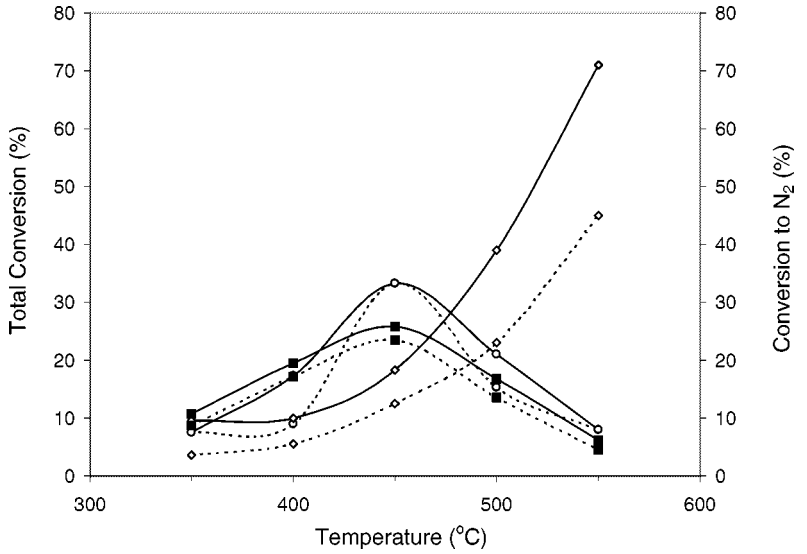


Figure 6. Total NO conversion (solid lines) and conversion to $N_2$ (dashed lines) over H-ZSM-5 (○), 24% In/$\gamma$-$Al_2O_3$ (■), and 28% In-CPG-SMMC (◊) in the presence of water. Feed: 1000 ppm NO, 5% $O_2$, 0.65% $C_2H_5OH$, 10% $H_2O$, balance He, GHSV = 60,000 $h^{-1}$.

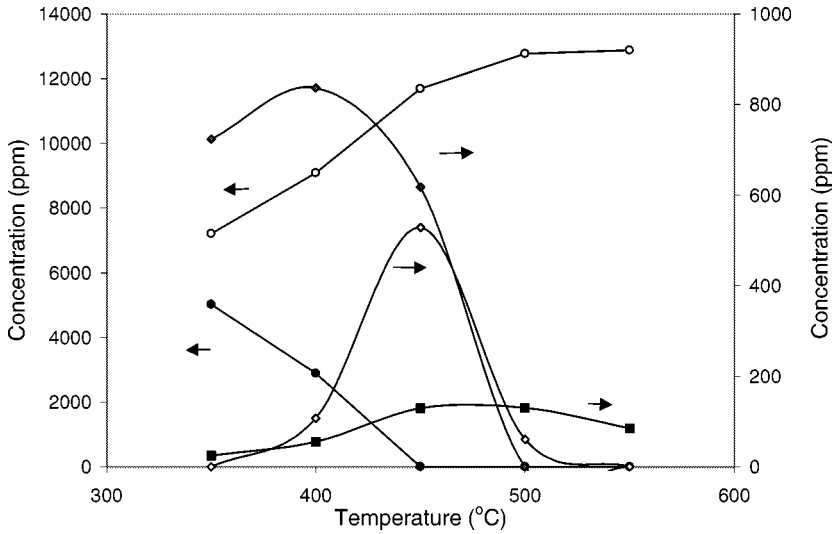


Figure 7. Distribution of carbonaceous species in the products of the reduction of NO by ethanol over 24% In/$\gamma$-$Al_2O_3$ in the presence of steam. Feed: 1000 ppm NO, 5% $O_2$, 0.65% $C_2H_5OH$, 10% $H_2O$, balance He, GHSV = 60,000 $h^{-1}$. (●) 2 × $C_2H_5OH$, (♦) 2 × $CH_3CHO$, (■) $CH_4$, (◊) 2 × $CH_3CN$, and (○) $CO_x$.

its surface. The surface boron concentration increases with time and temperature. So at higher temperatures the acidity of CPG also increases. There must also be a synergism between the metallic indium and the porous glass support, but further studies need to be conducted in order to elucidate this aspect.

It may be noted, by comparing with figures 4 and 5, respectively, that the presence of water had a negligible effect on the distribution of carbonaceous species (figures 7 and 8) for both the In/$\gamma$-$Al_2O_3$ and In-CPG-SMMC catalysts. It is noteworthy, however, that the formation of $CH_3CN$ over In-CPG-SMMC (figure 8) was completely inhibited in the presence of water vapors, while over 24% In/$\gamma$-$Al_2O_3$ (figure 7), 270 ppm is still formed at 450 °C. A similar effect was observed by Sumiya et al. [25] when the addition of water considerably decreased the formation of CN species over a $Ag/Al_2O_3$/cordierite catalyst.

XRD patterns of the 28% In-CPG-SMMC before and after reaction are compared in figure 9 (feed composition: 0.1% NO + 5% $O_2$ + 0.65% EtOH in balancing He, 60 h of continuous reaction at a total gas flow rate of 100 $cm^3$/min, reaction temperatures were varied from 350 to 550 °C). The fresh catalyst (figure 9(a)) contained only metallic indium. Indium oxide or any other crystalline impurities were not observed. The broad peak at around 22° $2\theta$ angle is due to the amorphous $SiO_2$. The spectra taken after reaction (figure 9(b)) suggest that most of the indium has been oxidized to $In_2O_3$. Thus, the equilibrated catalyst was mostly $In_2O_3$ after having been exposed to reactants for an extended period of time.

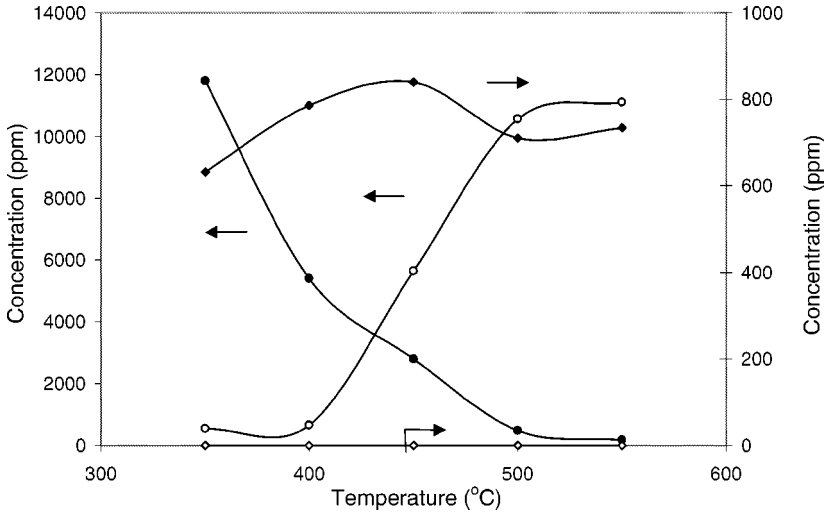


Figure 8. Distribution of carbonaceous species in the products of the reduction of NO by ethanol over 28% In-CPG-SMMC in the presence of steam. Feed: 1000 ppm NO, 5% $O_2$, 0.65% $C_2H_5OH$, 10% $H_2O$, balance He, GHSV = 60,000 $h^{-1}$. (●) 2 × $C_2H_5OH$, (♦) 2 × $CH_3CHO$, (■) $CH_4$, (◊) 2 × $CH_3CN$, and (○) $CO_x$.

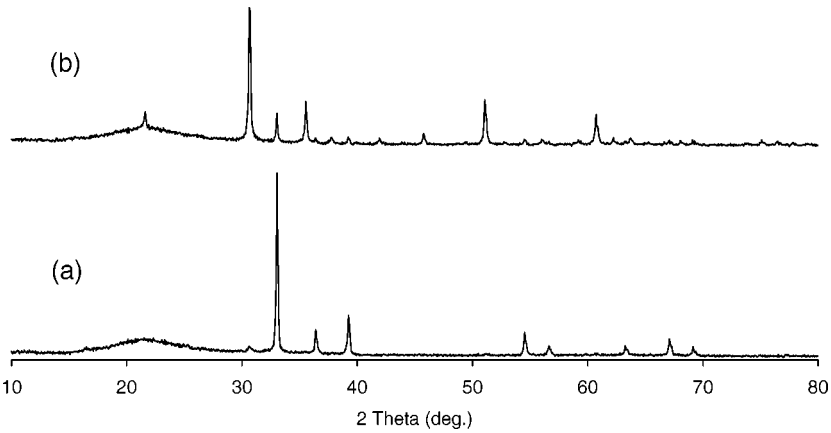


Figure 9. XRD patterns of the 28% In-CPG-SMMC before (a) and after reaction (b).

## 4. Conclusions

In-CPG-SMMC was found to be a highly active catalyst in the SCR of NO by ethanol under net oxidizing conditions in the presence of water vapors at high GHSV. As opposed to H-ZSM-5 and In/$\gamma$-$Al_2O_3$ catalysts, the catalytic activity of In-CPG-SMMC was enhanced by the presence of 10% water vapor in the feed. The formation of harmful $CH_3CN$ over In-CPG-SMMC was completely inhibited in the presence of water.

The activity of this new catalyst appears to increase monotonically with temperature. This feature makes In-CPG-SMMC attractive for practical applications at temperatures above 500 °C for the SCR of NO in exhaust streams containing water and excess oxygen.

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