Effect of Neodymium on the Physico-chemical Properties and N₂O Decomposition Activity of Co(Cu)–Al Mixed Oxides

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Cu–Al and Co–Al (M/Al = 0.8/0.2, molar ratio; M = Cu or Co) mixed metal oxides showed a moderate activity in N₂O decomposition (deN₂O) with full conversion at about 500–550 °C. Addition of Nd modified the physico-chemical and catalytic properties of both Cu–Al and Co–Al materials, though an optimal Nd loading was required. The addition of Nd in Cuₓ₀.₈ₐ₁₋ₓAl₂ and Coₓ₀.₈ₐ₁₋ₓAl₂ with Nd/Cu(Cu)/Al = 0.5/0.8/0.2 molar ratio, improved significantly their catalytic activity. In particular, the temperatures needed for full N₂O conversion were 100 °C lower than those of the bare materials. Furthermore, the most active Ndₓ₀.₅₅₈₈₉₀₂₈Al₂ catalyst exhibited a stable 60% N₂O conversion at 450 °C for 50 h in presence of NO (200 ppm), O₂ (20,000 ppm) and H₂O (2500 ppm). The enhancement in the activity of both Cu- and Co-containing materials was related to the formation of CuNdOₓ for Ndₓ₀.₅₅₈₈₉₀₂₈Al₂ and NdCoO₃ for Ndₓ₀.₅₅₈₈₉₀₂₈Al₂. The presence of CuNdO₃ or NdCoO₃ accounted for changes in redox properties, e.g., easier reduction or faster release of oxygen, and in turn an increased catalyst activity in deN₂O. The investigation of rare earth-based catalysts for deN₂O is limited in the scientific literature, while a comprehensive understanding of the involved active species may facilitate a knowledge-based catalyst optimization.

Introduction

Nitrous oxide (N₂O) significantly contributes to the global warming and the depletion of stratospheric ozone layer due to its long lifetime of approximately 150 years in the atmosphere, i.e., 265–298 times the Global Warming Potential (GWP) of CO₂ for a 100-year time-scale.[1,2] Depending on the natural or anthropogenic sources of N₂O, its abatement is realized by either limiting N₂O formation or using end-of-pipe remediation technologies. The after-treatment methods, including catalytic N₂O decomposition and/or its reduction are applied especially in adipic and nitric acid production plants, where N₂O is an inevitable by-product.[3] Compared to other routes for N₂O removal, i.e., utilization of N₂O in the selective catalytic oxidation or reduction with additional reactants, the direct N₂O decomposition constitutes the most appropriate solution to abate nitric oxide from the industrial sources due to its simplicity.[4] A great number of active catalytic systems have been reported for deN₂O, including zeolites,[5,6] hexaaluminates,[7,8] perovskites,[9,10] spinels,[11–14] hydrotalcite derived mixed metal oxides,[13,14] etc., and they are summarized in recent reviews.[2–4,17,18] Among others, high activity in N₂O decomposition were reported over Cu- and/or Co-containing hydrotalcite derived mixed metal oxides. For example, Kannan[19] investigated Cu–Al, Ni–Al and Co–Al (M/Al = 3/1, mol. ratio; M = Cu, Ni, or Co) hydrotalcite derived mixed metal oxides and found significantly higher activity in deN₂O over Ni- or Co-containing materials (N₂O conversion of 84–95% at 450 °C, 0.1 g of catalyst, 0.0985 vol.% N₂O/He, total flow rate of 100 cm³ min⁻¹) than Cu-containing one (N₂O conversion of 48% at 450 °C). The authors also claimed that catalytic activity of Co–Al significantly increased with molar ratio of Co/Al from 2.0 up to 3.0. Based on the results of Co–Al catalysts, Armor et al.[20] reported that a partial replacement of Al by La provided a more active catalyst at quite low temperature (79% conversion at 300 °C) after a standard thermal activation. Although, the presence of La suppressed the formation of hydrotalcite precursor. On the other hand, Chmielarz et al.[21] reported a significantly higher activity of Cu–Mg–Al–O, (Cu/Mg/Al = 10/61/29, mol. ratio) hydrotalcite derived mixed metal oxides in comparison to their Co–Mg–Al–O, analogues (full conversion versus 90% N₂O conversion at 650 °C, 0.1 g of catalyst, 0.5 vol.% N₂O/4.5 vol.% O₂/He, total flow rate 100 cm³ min⁻¹). Furthermore, modifications of mixed metal oxides with: (i) noble metals (e.g., Rh, Ir, Ag),[16,19,22,23] (ii) transition metals – in bimetallic systems (e.g., Cu–Co, Cu–Mn, Co–Fe),[15,21,24] (iii) alkali metals (e.g., Rb, Cs, Na),[25,26] or (iv) rare earth metals (e.g., Ce, Pr),[22,26] etc., were reported in order to improve the catalytic activity in deN₂O.

Recently, promotion by addition of rare earth metals is gaining more attention. For instance, Abu-Zied et al.[28] investigated the effect of Nd₃, Pr₃, Tb₃ and Y-doping of mesoporous
NIO, and found that the following activity trend in deN₂O: Pr-NiO ≈ Tb-NiO ≈ Y-NiO > Nd-NiO. The neodymium-containing catalyst (small amount of Nd, 4 mol%) showed the lowest activity (50% N₂O conversion at 360 °C, 0.5 g of catalyst, 0.05 vol.% N₂O/He, total flow rate 200 cm³ min⁻¹) than other studied rare earth-doped NIO (50% N₂O conversion below 350 °C). Despite Nd is grouped as a rare earth element as Co, Ni or Cu. Additionally, Nd has multiple oxidation states, e.g., +2, +3 and +4, which may enhance for redox properties of the Nd-doped materials - an important factor for N₂O decomposition. Inspired by the above studies, in the present work, we investigated the effect of neodymium on physicochemical properties and N₂O decomposition activity of Nd–Cu (Co)–Al mixed oxides. Nd amount (x = Nd/(M + Al), mol. ratio) varied from 0 to 1. The catalyst precursors were prepared by conventional coprecipitation using Na₂CO₃/NaOH as precipitating agents. Various characterization techniques, including XRD, BET, HR-TEM, XPS, H₂-TPR, O₂-TPD were employed to investigate the properties of the obtained materials. The catalytic properties of Nd–Cu (Co)–Al mixed oxides were evaluated in N₂O decomposition in either absence (N₂O/N₂) or presence of inhibitors (mixture of N₂O, O₂, NO, H₂O/N₂). The work about Nd-containing Cu–Al and Co–Al mixed metal oxides has not been explored to date, thus, the present contribution fills the literature gap.

### Table 1. Calculated and determined composition, specific surface area (a(BET)), total pore volume (Vₚ) and amounts of H₂ consumed during TPR measurements (H₂ uptake) of mixed oxides.

| Sample | Molar ratio | Nd/(Cu(Co) + Al) nominal | Nd/(Cu(Co) + Al) determined | Cu(Co)/Al determined | a(BET) [m² g⁻¹] | Vₚ [cm³ g⁻¹] | H₂ calculated [a] | H₂ uptake [b] |
|--------|-------------|--------------------------|-----------------------------|----------------------|-----------------|-------------|-----------------|---------------|
| Cu₆Al | 0           | 0                        | 3.80                        | 60                   | 0.36            | 10.7        | 10.3            |
| Nd₄Cu₆Al₂ | 0.10        | 0.12                     | 3.69                        | 61                   | 0.58            | 8.5         | 10.0            |
| Nd₂Cu₄Al₄ | 0.25        | 0.27                     | 3.61                        | 38                   | 0.45            | 6.5         | 8.0             |
| Nd₄Cu₄Al₄ | 0.50        | 0.55                     | 3.86                        | 32                   | 0.44            | 4.6         | 6.7             |
| Nd₄Cu₄Al₂ | 0.75        | 0.84                     | 4.05                        | 31                   | 0.36            | 3.7         | 5.5             |
| Nd₄Cu₄Al₂ | 1.00        | 1.10                     | 3.66                        | 29                   | 0.38            | 3.1         | 4.5             |
| Co₆Al | 0           | 0                        | 3.65                        | 52                   | 0.30            | 14.2        | 14.0            |
| Nd₄Co₄Al₂ | 0.10        | 0.11                     | 3.75                        | 75                   | 0.24            | 11.2        | 12.6            |
| Nd₄Co₄Al₂ | 0.25        | 0.29                     | 3.83                        | 71                   | 0.28            | 8.8         | 10.7            |
| Nd₄Co₄Al₂ | 0.50        | 0.59                     | 4.18                        | 37                   | 0.13            | 6.3         | 9.6             |
| Nd₄Co₄Al₂ | 0.75        | 0.86                     | 4.13                        | 29                   | 0.17            | 5.0         | 7.5             |
| Nd₄Co₄Al₂ | 1.00        | 1.08                     | 4.06                        | 13                   | 0.09            | 4.0         | 7.1             |
| NdO₂ | -           | -                        | -                           | -                    | 26              | 0.20        | 5.9             | 0.6          |

[a] Determined with ICP-MS analysis. Nominal molar ratio of M/Al = 4. [b] Calculated based on only calculated amount of Cu or Co in the mixed oxides. Calculated by equation: Y = 6.686-0.06X, R² = 0.9972, and X, Y referred to the area of each reduction peak and the H₂ consumption, respectively.

Results and Discussion

The chemical composition of the (Nd)–Co(Cu)–Al calcined catalysts confirmed the coprecipitation of Cu, Co and Al with molar ratios quite close to nominal ones as shown in Table 1. Only the Nd content in mixed oxides was slightly lower compared to the nominal values (especially for samples with high Nd loading). However, all deviation values were less than 10%.

Figure 1 presents the XRD patterns of mixed oxides after calcination of the precursors at 600 °C. The decomposition of the HT structure in Cu₆Al₂ led to formation of a CuAlO₂ mixed oxide, represented by the main reflections at angle 2θ 32, 46 and 57°. The formation of CuAlO₂ instead of Cu₄Al₂O₇ suggested that Nd³⁺ was more prone to form the Cu-containing spinel phase than Al³⁺. Otherwise, the formation of amorphous or small amounts of NdAl₂O₅ at 2θ ca. 11, 20 and 52°, and Nd₄O₈ at 2θ ca. 23, 31, 44 and 56° (Figure 1a). For additional amounts of Nd/(Cu + Al) > 0.5, a new phase of CuNd₃O₈ was formed as evidence of the reflections at 2θ ca. 32, 46 and 57°. The formation of CuNd₃O₈ instead of CuAlO₂ suggested that Nd³⁺ was more prone to form the Cu-containing spinel phase than Al³⁺. Otherwise, the formation of amorphous or small amounts of Nd₄Al₂O₇ at 2θ ca. 24 and 32°, and Nd₄O₈ at 2θ ca. 32 and 47° cannot be excluded. A further increase of Nd amount to x ≥ 0.75 provoked the formation of Nd₃(CO₂)₃(OH) at 2θ ca. 31 and 44°, and Nd₄O₈ at 2θ ca. 31 and 46° (Figure 1a). Therefore, it seems that neodymium is responsible for the stabilization of carbonates in the sample.

The pattern of the Co₆Al₂ calcined sample (Figure 1b) showed well-defined crystalline phases with Bragg reflections located at 2θ ca. 19, 32, 37, 39, 45, 56, 59, 65, 77°, related to Co₅O₄ CaAl₂O₄ and/or Co₅Al₂O₄ spinels. A decrease in the crystallinity of the spinel phases was observed after addition of low Nd amount (x ≤ 0.25). Increasing the Nd amount to x = 0.5 in Nd₅Co₅Al₅ resulted in even less intense reflections of spinel, and the formation of well-crystallized cubic NdO₃ at 2θ ca. 28, 32, 46 and 55° and perovskite phases (NdAl₂O₄ and/or Nd₂CoO₄) at 2θ ca. 24, 34, 42, 48, 60 and 71°. For the samples with x = 0.75 and 1, CoAlO₂ was still present, though with a very low intensity in comparison to Nd-containing phases. The excess of Nd in the high loaded samples (x ≥ 0.75) led to the formation of additional phases, such as Nd₂O₃ at 2θ ca. 11, 20 and 52°, Nd(OH)₂ at 2θ ca. 16, 26, and 27° and Nd (CO₃)₂(OH) at 2θ ca. 23, 31, 35, 44 and 47° (Figure 1b). However, the presence of oxycarbonate – Nd₀.₅CO₃ (2θ ca. 11, 23, 26, 31, 34, 46, 48 and 56°), already identified in the Cu-
The decrease in the specific surface area and pore volume when increasing Nd amount could be correlated with the appearance of relatively small specific surface area phases (e.g., Nd$_2$O$_3$: 26 m$^2$ g$^{-1}$ or NdAlO$_3$: 1.5 m$^2$ g$^{-1}$). In the case of Co–Al series, the addition of low Nd amount resulted in an increment of specific surface area from 52 m$^2$ g$^{-1}$ of Co$_{0.8}$Al$_{0.2}$ to around 71–75 m$^2$ g$^{-1}$ for Nd$_{0.8}$Co$_{0.2}$, $x = 0.1$, 0.25. However, again the decrease of specific surface area and pore volume for materials with a high Nd amount ($x \geq 0.5$) occurred together with the formation of Nd-containing phases, e.g., Nd$_2$O$_3$, NdCoO$_3$ and NdAlO$_3$ (as identified by XRD; Figure 1b). The sample with the highest Nd amount (Nd$_{0.8}$Co$_{0.2}$Al$_{0.2}$) showed the specific surface area of only 13 m$^2$ g$^{-1}$ and pore volume of 0.09 cm$^3$ g$^{-1}$.

The Nd addition did not have any remarkable effect on the morphology of the Cu–Al catalysts. Broadly speaking, ill-defined nanoparticles with sizes around 10–30 nm were identified as shown in Figure 2a and 2b. The presence of particles with different compositions was supported by STEM/EDS. For instance, three typical compositions were quantified: (i) Cu-rich (no Al) (point 1 image a2 or point 4 image b2); (ii) Nd-rich (point 2 image a2, point 1 image b2); (iii) close to nominal one (point 4 image a2). The differences in composition should be related
to the crystalline phases identified by XRD. However, SAED measurements did not help to support this statement (Figure 2a1, 2b1).

In contrast, addition of Nd significantly influenced on the morphology of the Co–Al samples (Figure 2c and 2d). In Nd$_{0.8}$Cu$_{0.2}$Al$_{2}$, 5–15 nm sized Co-rich particles (ill-defined shape) (Figure 2c1), and 15–30 nm spherical Nd-rich particles (Figure 2c3) were identified. HAADF-STEM images (Figure 2c2 and 2c4) and EDS measurements (Table on the right of Figure 2c2, 2c4) provided more information about the differences in composition. Further, increasing the amount of Nd in Nd$_{1-x}$Co$_{0.8}$Al$_{2}$ resulted in the appearance of flakes (Figure 2d). Compositions identified by EDS measurements from HAADF-STEM images (Figure 2d1) revealed the presence of particles with various molar ratio, including the one close to nominal value (point 3), Nd-rich (point 1) as well as Nd-less (point 4). Remarkably, in some locations, particles mainly made by Nd (no Al signal) (point 2) were identified, suggesting that it could be assigned to NdO$_{2}$.

The reducibility of mixed oxides was studied by H$_{2}$-TPR in the range of 50–1000°C (Figure 3). The H$_{2}$ consumption obtained from the H$_{2}$-TPR profiles was compared to the calculated H$_{2}$ uptakes (Table 1). Influence of Nd addition on the reducibility of the materials can be visualized by the change of the shape of H$_{2}$-TPR profiles and H$_{2}$ consumption. Cu$_{0.8}$Al$_{2}$ showed a broad peak in range from 200–400°C, associated to the reduction of copper oxide to metallic copper.$^{[44]}$ In fact, its hydrogen consumption was 10.3 mmol g$^{-1}$, almost the same as expected amount (Table 1). Addition of Nd shifted the reduction temperatures but the trend is not clear. Despite the increment of reduction temperature of Nd$_{0.8}$Cu$_{0.2}$Al$_{2}$ and Nd$_{0.5}$Cu$_{0.8}$Al$_{2}$, other samples, in particular Nd$_{0.1}$Cu$_{0.8}$Al$_{2}$, showed decrements of reduction temperature. On the other hand, in Nd-containing catalysts the experimental H$_{2}$ consumption values overcome those calculated based on the total reduction of Cu$^{2+}$ in the catalysts. Moreover, the deviation was linearly proportional to Nd amount. Therefore, the contribution of the Nd$^{3+}$ reducibility in H$_{2}$ consumption had to be taken into account. A temperature peak at around 610°C was observed in the samples containing high Nd amount. Indeed, Nd$_{0.8}$O$_{2}$ showed a main reduction peak around 600°C and H$_{2}$ consumption of about 0.6 mmol g$^{-1}$. This H$_{2}$ consumption was about 10% of the calculated value, suggesting that Nd$_{0.8}$O$_{2}$ was hardly reducible in the applied range of 50–1000°C. At this stage, the single redox behavior of other Nd-containing phases (i.e., NdAlO$_{4}$, CuNd$_{0.5}$O$_{4}$, Nd(CO$_{3}$)(OH) and Nd$_{2}$O$_{5}$CO$_{3}$) were not explored in detail in the H$_{2}$-TPR studies. However, they could contribute to the H$_{2}$ consumption. Our catalytic investigations were conducted up to 600°C, i.e., temperatures significantly lower than reduction of neodymium-containing oxide species.

Co$_{0.8}$Al$_{2}$ reduction profile showed two broad peaks centered at 466 and 807°C and total hydrogen consumption of about 14.0 mmol g$^{-1}$ (98.6% reduction). The low temperature peak (below 515°C) could be related to the two-reduction steps of CoO$_{2}$ to CoO and then to metallic Co. While the reduction temperature peak at higher temperatures was ascribed to the reduction of cobalt species in Co$_{3}$AlO$_{4}$ and/or CoAlO$_{4}$ to metallic cobalt, respectively.$^{[11,24,44-46]}$ Samples with low Nd loading (Nd$_{0.8}$Cu$_{0.2}$Al$_{2}$ and Nd$_{0.5}$Cu$_{0.8}$Al$_{2}$) showed similar redox properties as Co$_{0.8}$Al$_{2}$. On the other hand, for Nd$_{0.8}$Cu$_{0.2}$Al$_{2}$ the first peak shifted to higher temperatures (from 466 to 506°C) and the second peak to lower temperatures (from 807 to 786°C) in comparison to those of Co$_{0.8}$Al$_{2}$. Otherwise, the reduction of NdCoO$_{3}$ (presence revealed by XRD, Figure 1b) in the same temperature range cannot be excluded.$^{[45]}$ A main H$_{2}$ consumption at around 468°C and a broad peak between 510–
770 °C were observed for Nd$_{0.75}$Co$_{0.8}$Al$_{0.2}$ and an extra small peak at around 805 °C for Nd$_{1.0}$Co$_{0.8}$Al$_{0.2}$. The change in the redox properties of such materials agreed with XRD results (decrease of reflections corresponding to CoAlO$_y$ and increase in those of Nd-containing phases). Indeed, the reduction peaks of the perovskite phase[47] and Nd$_2$O$_3$ were clearly observed in the TPR profiles. Again, the total amount of H$_2$ consumed in the presence of Nd was higher than that calculated by considering only the reduction of Co present in the samples to Co$^0$. Thus, the reduction of Nd-containing phases should be taken also into account. Not only the reducibility but also the ability to release oxygen influences the activity of the catalyst in N$_2$O decomposition.[11,12]

To evaluate the influence of Nd on this property, temperature programmed desorption of oxygen (O$_2$-TPD) coupled with mass spectrometry (MS) was carried out, as shown in Figure 4. The tracking of $m/z = 32$ revealed negligible release of oxygen from Cu$_{0.8}$Al$_{0.2}$ at temperatures lower than 400 °C, a gradual increase in the range of 400–600 °C and a significant increase with temperature higher than 750 °C. The species desorbed...
below 400 °C can be attributed to the surface oxygen species (O\textsubscript{α}) weakly bonded on the surface, while the species evolved above 400 °C were associated to lattice oxygen (O\textsubscript{β}) and bulk oxides (O\textsubscript{γ}). Similarly, O\textsubscript{2} released gradually from 400 to 850 °C and significantly at temperatures higher than 850 °C on Co\textsubscript{0.8}Al\textsubscript{0.2}. The presence of Nd in the catalysts resulted in different abilities to release oxygen for each series in the temperature range of 50–600 °C (where the catalytic reaction was later performed). Up to 600 °C, the Nd-containing materials did not show significant differences in oxygen release ability compared to Co\textsubscript{0.8}Al\textsubscript{0.2}. However, at the same temperature range, Nd\textsubscript{0.5}Co\textsubscript{0.8}Al\textsubscript{0.2} (x = 0.5, 1.0) showed larger amounts of oxygen release compared to that of Co\textsubscript{0.8}Al\textsubscript{0.2}. It should be noted that an analogous trend between TCD and m/z = 44 signal indicated for a significant contribution of simultaneous CO\textsubscript{2} release during the TPD run. Thus, in case of Nd-containing materials, CO\textsubscript{3}\textsuperscript{2−} anions were partially preserved (mainly in the form of Nd(CO\textsubscript{3})OH and Nd\textsubscript{2}O\textsubscript{2}CO\textsubscript{3} phases as proved by XRD analysis) after calcination at 600 °C and were subsequently released above this temperature during the O\textsubscript{2}-TPD.

Figures 5, 6 and S1, S2 (Supporting information) show Cu 2p, Co 2p Nd 3d, and O 1s XPS spectra and Table 2 summarizes positions of binding energy (BE), full-width at half-maximum (FWHM), peak area and molar ratio of O\textsubscript{α}/(O\textsubscript{β} + O\textsubscript{γ}) of selected mixed oxides. As shown in Figure 5a, the spectra of Cu\textsubscript{0.8}Al\textsubscript{0.2} and Nd\textsubscript{0.5}Cu\textsubscript{0.8}Al\textsubscript{0.2} were characterized by two main peaks, Cu

![Figure 5](https://example.com/fig5.png)

**Figure 5.** XPS spectra of Nd\textsubscript{x}Cu\textsubscript{0.8}Al\textsubscript{0.2} (Cu 2p, a,b) and Nd\textsubscript{x}Co\textsubscript{0.8}Al\textsubscript{0.2} (Co 2p, c,d), x = 0, 0.5.

| Sample             | Position/FWHM [eV] | Molar ratio\textsuperscript{[a]} O\textsubscript{α}/(O\textsubscript{β} + O\textsubscript{γ}) |
|--------------------|--------------------|---------------------------------|
| Cu\textsubscript{0.8}Al\textsubscript{0.2} | 528.25/2.08 | 1.15 |
|                    | 529.82/2.06 |     |
|                    | 531.31/1.77 |     |
|                    | 532.60/2.25 |     |
| Nd\textsubscript{0.5}Cu\textsubscript{0.8}Al\textsubscript{0.2} | 528.04/2.39 | 0.77 |
|                    | 529.63/2.23 |     |
|                    | 531.15/1.92 |     |
|                    | 532.42/2.07 |     |
| Nd\textsubscript{0.5}Co\textsubscript{0.8}Al\textsubscript{0.2} | 528.08/2.02 | 0.56 |
|                    | 529.69/2.00 |     |
|                    | 531.17/1.72 |     |
|                    | 532.41/2.16 |     |
| Co\textsubscript{0.8}Al\textsubscript{0.2} | 528.46/1.85 | 0.22 |
|                    | 530.11/1.86 |     |
|                    | 531.66/2.03 |     |
|                    | 533.12/2.22 |     |
| Nd\textsubscript{0.5}Co\textsubscript{0.8}Al\textsubscript{0.2} | 528.26/2.30 | 0.84 |
|                    | 529.77/1.98 |     |
|                    | 531.18/1.77 |     |
|                    | 532.43/1.95 |     |
| Nd\textsubscript{1.0}Co\textsubscript{0.8}Al\textsubscript{0.2} | 528.28/1.95 | 0.81 |
|                    | 530.09/2.12 |     |
|                    | 531.42/1.77 |     |
|                    | 532.52/2.13 |     |

\[\textsuperscript{[a]} \text{Estimated from the integrated areas of the respective XPS peaks and normalized by metal loading.}\]
2p_{1/2} (around 953 eV) and Cu 2p_{3/2} (933 eV), with shake-up satellites. The presence of shake-up peaks as well as the Cu 2p_{3/2} peak at around 933 eV suggested that Cu^{2+} is the prime oxidation state in these samples.\(^{[50]}\) An analogous character was observed for Nd_{1-x}Cu_{x}Al_{2} (Figure S1a). In Figure 6a, the Co 2p XPS spectra of Co_{0.8}Al_{2} exhibited two main peaks characterized by BE of around 781 and 797 eV corresponding to the Co 2p_{3/2} and 2p_{1/2} spin-orbit peaks, respectively.\(^{[51,52]}\) The binding energy of Co^{2+} and Co^{3+} is in the range of 780.9–780.3 and 779.7–779.4 eV, respectively.\(^{[14,52,53]}\) The slightly higher binding energy of the Co^{3+} explains the asymmetric shape of the Co 2p peak.\(^{[52,53]}\) Some of us reported similar BE values in our previous work concerning Co_{0.7}Al_{2.5} hydrotalcite derived mixed metal oxide.\(^{[16,14]}\) The spin-orbit values for Co_{0.8}Al_{2} varied in the range of 15.3–15.4 eV, indicating mainly the presence of CoO_{4} (spin-orbit value of 15.2 eV) with the minor part of CoAlO_{4} (spin-orbit value of 15.9 eV).\(^{[51,54,55]}\) Thus, the surface of the catalyst was enriched in CoO_{4}. Addition of Nd, shifted the range to 15.5–15.6 eV for Nd_{0.8}Co_{0.2} (x = 0.5, 1.0), demonstrating a higher amount of CoAlO_{4} or NdCoO_{3} (as confirmed with XRD analysis) on the surface (Figure 6b, 6b). As shown in Figure 6, the Nd 3d core level showed a characteristic of multiplet splitting with the presence of the low binding energy satellites due to the 3d–4f coupling.\(^{[56]}\) Nd 3d_{5/2} at BE 981 eV and Al 2p at 74 eV (results not shown) were rather close to those reported for NdAlO_{3} from literature\(^{[58,57]}\) and thus they could be related to Nd^{3+} in NdAlO_{3} for the Nd_{x}CuCo_{0.8}Al_{2} (x = 0.5, 1.0) samples. While higher BEs of Nd 3d_{5/2} in the range of 982–983 eV were similar to those reported for Nd^{3+} in Nd_{3}O_{4} (983.1 eV)\(^{[58]}\) or NdCoO_{2} (982.0 eV).\(^{[47]}\) Nd(CO_{3})(OH), Nd_{2}O_{3}CO_{3} or CuNd_{2}O_{4} (in case of Cu-containing materials). Since our materials were calcined at 600 °C, the presence of trace Nd(OH)_{3} was possibly included due to its relative thermal stability. In fact, it was probably confirmed by a closeness of the BE of 983.2 eV to value of 983.4 eV reported for Nd(OH)\(_{3}\).\(^{[56,59]}\) This might be similar to the case of La(OH)\(_{3}\) which was stable up to 600 °C as reported from the literature.\(^{[61]}\)

In Figure S2, the O 1s spectra were deconvoluted to lattice oxygen (O\(_{\alpha}\), BE 529 eV), surface-adsorbed oxygen, OH groups or oxygen vacancies (O\(_{\beta}\), BE 531 eV) and adsorbed molecular water (O\(_{\gamma}\), BE 532 eV), respectively.\(^{[62]}\) An additional peak at about BE 528 eV appeared possibly due to the differential charging of the materials.\(^{[63]}\) The ratio of O\(_{\alpha}/(O_{\beta}+O_{\gamma})\) was 1.15 and 0.22 for Cu_{0.8}Al_{2} and Co_{0.8}Al_{2}, respectively. However, rising the amount of Nd addition resulted in a different behavior for Cu–Al and Co–Al series. For Nd-containing materials, the ratio decreased to 0.77 for Nd_{0.8}Cu_{0.2}Al_{2} and further to 0.56 for Nd_{1.0}Cu_{0.2}Al_{2}. While for Nd_{0.8}Co_{0.2}Al_{2} and Nd_{1.0}Co_{0.2}Al_{2}, it remained in the range of 0.81–0.84. The aforementioned differences on O\(_{\alpha}/(O_{\beta}+O_{\gamma})\) are expected to notably affect the reaction on the basis of the redox mechanism followed in denN\(_{2}O\). However, in our case we did not observe clear link between the strength of lattice...

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Figure 6. XPS spectra of Nd 3d of Nd_{x}Cu_{0.8}Al_{2} (a, b), Nd_{x}Co_{0.8}Al_{2} (c, d), x = 0, 0.5, 1.0.
oxygen-metal bonds and the catalysts activity. Thus, further work is required in order to precisely determine the individual role of catalysts components in deN$_2$O.

Lastly, it was noted that in both Nd$_{0.5}$Cu$_{0.8}$Al$_{0.2}$ and Nd$_{1.0}$Cu$_{0.8}$Al$_{0.2}$ samples, the appearance of C 1s non-adventitious carbon species (~289.9 eV) was identified on the surface (not shown), but they was not observed for Co$_{0.8}$Al$_{0.2}$, Cu$_{0.8}$Al$_{0.2}$ and Nd$_{0.5}$Co$_{0.8}$Al$_{0.2}$. The coupling of these high BEs of C 1s (289.9 eV) and O 1s (531 eV) was due to the formation of oxo-carbonaceous species on the surface,\textsuperscript{[64]} which agreed with XRD data.

Figure 7 shows the results of catalytic tests (a–d) and reaction rate (e, f) of mixed oxides: (a, e) Nd$_{x}$Cu$_{0.8}$Al$_{0.2}$ and (b, f) Nd$_{x}$Co$_{0.8}$Al$_{0.2}$; $x$ = 0, 0.1, 0.25, 0.5, 0.75, 1.0; (c, d) Nd$_{0.5}$Co$_{0.8}$Al$_{0.2}$. Experimental conditions: mass of sample = 300 mg, reactant concentrations at the reactor inlet: 1000 ppm N$_2$O in N$_2$ as balance (a–d), 1000 ppm N$_2$O, 200 ppm NO, 20,000 ppm O$_2$, 2500 ppm H$_2$O in N$_2$ as balance (e, f), total flow rate = 80 ml min$^{-1}$. Stability test (d) was carried out at 450 °C for 50 h.
conversion at about 500 and 550 °C for Co$_3$O$_4$Al$_2$O$_4$ and Cu$_3$Al$_2$O$_4$ (respectively). A higher activity was also obtained for Nd-containing Co–Al materials compared to Cux-Al, while not clear correlation existed between Nd-loading materials and their activity in deN$_2$O. Remarkably, before doing the investigation of current materials with high loading of Nd, the Nd$_3$Cu$_2$Al$_2$ catalysts doped with small amount of Nd (x = 0.025, 0.05, 0.075) were tested and their catalytic activity was only slightly improved than that of Cu$_3$Al$_2$O$_4$ (Figure S3). The activity for Nd$_3$Co$_2$Al$_2$O$_4$ was relatively low with about 94% conversion at 500 °C. Among tested materials, Nd$_3$Co$_2$Al$_2$O$_4$ revealed the highest activity with T$_{50}$ at 385 °C and full conversion at 450 °C. Thus, this catalyst was further examined under harsh conditions of 1000 ppm N$_2$O, 200 ppm NO, 20 000 ppm O$_2$, and 2500 ppm H$_2$O/N$_2$. T$_{50}$ was shifted to around 435 °C (~ 120 °C higher than for test carried out in 1000 ppm N$_2$O/N$_2$ (Figure 7c). In the stability test performed under the same feed at 450 °C, the catalyst exhibited a stable conversion of N$_2$O around 60% for 50 h of time-on-stream. At the end of the test only 3% loss in the activity was observed (Figure 7d).

The reaction rates were calculated at temperatures ≤ 350 °C, where the conversion was low enough (<20%) to minimize the influences of gradient of concentration as well temperature due to exothermic reaction. Nd$_3$Cu$_2$O$_4$Al$_2$ exhibited the highest reaction rate of about $1.1 \times 10^{-9}$ mol m$^{-2}$ s$^{-1}$ at 350 °C (Figure 7e). Remarkably, Nd$_3$Co$_2$O$_4$Al$_2$ showed the same maximum value of reaction rate ($1.1 \times 10^{-9}$ mol m$^{-2}$ s$^{-1}$) as for Nd$_3$Cu$_2$O$_4$Al$_2$ but at significantly lower temperature, i.e., 250 °C (Figure 7f).

The activity of both Cu- and Co-containing catalysts depended on the new phase formation of the corresponding mixed oxides (mainly CuNd$_2$O$_4$ for Nd$_3$Cu$_2$Al$_2$O$_4$ and NdCoO$_4$ for Nd$_3$Co$_2$Al$_2$O$_4$). Although, the XRD analysis did not reveal CuNd$_2$O$_4$ for materials with small amount of Nd (Nd$_{12}$Cu$_8$Al$_2$O$_4$ and Nd$_{022}$Cu$_0$Al$_0$O$_4$), its presence in the amorphous phase cannot be excluded. A promising tool for further investigations will be certainly in situ EXAFS/XRD analysis. However, such studies need to be investigated separately.

In short summary, when Nd$^{3+}$ is added to Cu$_3$O$_4$Al$_2$O$_4$ or Co$_3$O$_4$Al$_2$O$_4$ materials, it modifies their physico-chemical properties, and in turn the catalytic activity. The amount of Nd and the nature of the active species in the original catalyst (Cu or Co cations) determine the catalyst modifications. With Nd/(M + Al) ≤ 0.25 (where M = Cu or Co), Nd-containing phases were well dispersed and the structure of the mixed oxides for Cu$_3$O$_4$Al$_2$O$_4$ and mixed oxide/spinel for Co$_3$O$_4$Al$_2$O$_4$ is almost preserved. For Nd/(M + Al) ≥ 0.5, in Cu-catalysts, Nd$^{3+}$ interacts with Cu$^{2+}$ to form crystalline CuNd$_2$O$_4$ spinel or with Al$^{3+}$ to form a NdAl$_2$O$_4$ perovskite or further segregates to separate Nd$_2$O$_3$ and Nd$_2$O$_3$CO$_3$. Whereas Nd$^{4+}$ reacts with Co$^{3+}$ or Al$^{3+}$ to form NdCoO$_3$ or NdAl$_2$O$_4$, respectively, as well as further creates separate oxide and oxocarbonate of Nd. Hence, the addition of Nd provokes the formation of new crystalline phases containing active Cu and Co species, which coexist with the mixed oxide or spinel phases, and in turn modifies the redox properties or oxygen release. The other Nd-containing phases, dependent on the Nd amount, greatly determine specific surface area and reduction temperature, playing also a role in the availability of the active sites and therefore in the deN$_2$O catalytic activity. Obviously, further work is required to reveal the local surface chemistry of active sites of Nd-containing catalysts under reaction conditions by means of operando measurements.

According to the literature, Ln$_2$CuO$_4$ (Ln = La, Pr, Nd, Sm, and Gd) are active in the N$_2$O decomposition,[46] while their activity appeared due to the presence of redox pair Cu$^{2+}$/Cu$^{4+}$ and oxygen vacancy.[65] Daquinou et al.[66] investigated LaCoO$_3$ and pointed out that the restoration of anionic vacancies Co$^{3+}$/V$_{\text{O}}$ at low temperature responding for a high activity of the catalyst. Interestingly, in our case pure Nd$_2$O$_3$ revealed poor activity. Otherwise, Nd$_2$O$_3$ was reported as a promoter in deN$_2$O.[28,67] For instance, Abu-Zied et al.[68] reported that Nd-NiO (prepared by precursors containing 5 mol.% of Nd) reduced the T$_{50}$ value of about 50 °C (0.5 g catalyst, 200 ml min$^{-1}$ of 500 ppm N$_2$O/He). Xue et al.[69] investigated that the best catalyst in a series of Fe–Cu–Nd mixed oxides with different Nd ratio (Fe/Cu = 0.5 and Nd ratio varied from 0.5 to 2.5) was Fe$_{0.3}$Cu$_{0.7}$Ni$_{0.4}$ with N$_2$O conversion of 40% and 90% at 450 and 550 °C, respectively (5 ml catalyst, 200 ml min$^{-1}$ of 2000 ppm N$_2$O/He).

The apparent activation energy (E$_a$) of N$_2$O decomposition was derived assuming the pseudo first order reaction (Figure 8). Nd-containing catalysts showed significant decrease in apparent activation energy for both types of active phases. Nd$_3$Cu$_2$O$_4$Al$_2$O$_4$ and Nd$_{12}$Cu$_8$Al$_2$O$_4$ exhibited E$_a$ = 56 and 63 kJ mol$^{-1}$, respectively, which were almost a half of that on Cu$_3$O$_4$Al$_2$O$_4$. Otherwise, Co$_3$O$_4$Al$_2$O$_4$ showed a relatively low E$_a$ = 45 kJ mol$^{-1}$ and surprisingly, Nd$_3$Co$_2$O$_4$Al$_2$O$_4$ showed only E$_a$ of 19 kJ mol$^{-1}$. On the other hand, with high Nd loading, Nd$_{12}$Co$_8$Al$_2$O$_4$ exhibited slightly higher value of E$_a$ = 64 kJ mol$^{-1}$. Tendency in change of apparent activation energy was in accordance with reaction rate (Figure 7e, 7f) as well as overall their catalytic activity (Figure 7a, 7b). Furthermore, Stelmachowski et al.[70] reported that changes in the activation energy strongly correlated to the location of Co in the spinel lattice, being 63–71 kJ for octahedral Co$^{3+}$ ions (prime active sites) and 113–117 kJ for tetrahedral Co$^{2+}$ ions (much less active). A
Conclusions
Nd0.5Co0.5Al0.2 mixed oxides \((x = 0.1, 0.15, 0.25, 0.5, 0.5, 0.75, 1.0)\) were prepared by coprecipitation and subsequent calcination. Nd addition to Cu0.8Al0.2 or Co0.8Al0.2 significantly varied their activity in \(N_2O\) decomposition. The highest activity among tested materials revealed Nd2.5Cu0.5Al0.2 (in Cu-containing series) and Nd2.5Co0.5Al0.2 (in Co-containing series) with the light-off temperature at 385 °C and 315 °C in 1000 ppm \(N_2O/N_2\) respectively. Remarkably, Nd2.5Cu0.5Al0.2 catalyst exhibited about 60% \(N_2O\) conversion with 50 h of time-on-stream in the presence of \(N_2O, O_2, H_2O\). We correlated the high activity of the Nd-containing catalysts with the formation of new phases: CuNdO2 for Nd2.5Cu0.5Al0.2 and NdCoO2 for Nd2.5Co0.5Al0.2, respectively. While higher activity of the cobalt-containing materials related to their larger amount of surface oxygen and redox properties. The higher additional amount of Nd decreased the catalytic activity, possibly due to the formation of unfavorable phases (including NdAlO3 or Nd2O3).

Experimental Section

Catalyst Preparation
A series of Nd\(_x\)Cu(Co)\(_{1-x}\)Al\(_0.2\) (where \(x = 0, 0.1, 0.15, 0.25, 0.5, 0.5, 0.75, 1.0\)) mixed hydroxide precursors was prepared by coprecipitation of the corresponding 1.0 M aqueous nitrate solutions of Co(NO\(_3\))(3)6\(\cdot\)6\(\cdot\)H\(_2\)O (Roth), Cu(NO\(_3\))(3)6\(\cdot\)3\(\cdot\)H\(_2\)O (Sigma), Nd(NO\(_3\))(3)6\(\cdot\)9\(\cdot\)H\(_2\)O (Sigma) and Al(NO\(_3\))(3)6\(\cdot\)9\(\cdot\)H\(_2\)O (Sigma). The solution of metal salts was dropped to a vigorously stirred aqueous solution containing 1.0 M aqueous nitrate solutions of Co(NO\(_3\))(3)6\(\cdot\)6\(\cdot\)H\(_2\)O (Roth), Cu(NO\(_3\))(3)6\(\cdot\)3\(\cdot\)H\(_2\)O (Sigma), Nd(NO\(_3\))(3)6\(\cdot\)9\(\cdot\)H\(_2\)O (Sigma) and Al(NO\(_3\))(3)6\(\cdot\)9\(\cdot\)H\(_2\)O (Sigma). The pH was maintained constant at 10 ± 0.2 using 1.0 M NaOH (Chemsolute) solution. The resulting precipitate was aged for 0.5 h at 60 °C, filtered and washed thoroughly with distilled water until neutral pH around 7.0. The final product was dried at room temperature for 24 h and subsequently calcined at 600 °C for 6 h in static air. Mixed metal oxides were kept in a desiccator in order to avoid the reconstitution of the precursor structure. For catalytic tests, the catalysts were crushed and a fraction of materials particle size in range of 0.25–0.50 mm was collected to use.

Nd0.5O used as a reference material was prepared by precipitation of Nd(NO\(_3\))(3)6\(\cdot\)9\(\cdot\)H\(_2\)O (Sigma) using 28–30 wt.% NH\(_4\)OH solution. 35 ml of 1.0 M Nd(NO\(_3\))(3)6\(\cdot\)9\(\cdot\)H\(_2\)O aqueous solution was dropwise added into a 100 ml NH\(_4\)OH solution at pH 10 ± 0.2. The pH was controlled at this value by concentrated NH\(_4\)OH solution. The resulted precipitate was dried at room temperature and calcined at 600 °C for 6 h.

Catalysts Characterization
X-ray diffraction (XRD) measurements of the calcined powders were performed using a Siemens D5000 XRD diffractometer with Cu–K\(_\alpha\) radiation (\(\lambda = 1.54056 \, \text{Å}, 45 \, \text{kV}, 40 \, \text{mA}\)) in 20 range of 5–80° with a step size of 0.05° and scan step time of 15.25 s.

The chemical composition of mixed metal oxides was determined by ICP-MS using an Agilent Technologies 8800 Triple Quad spectrometer. Prior to measurement, sample (∼50 mg) was dissolved in 6 ml mixture of concentrated acids (HCHNO\(_3\), 1:1), and afterwards the resulted mixture was diluted with 64 ml deionized water before warming up to 40 °C for 24 h.
Specific surface area of the catalysts was determined by N₂ adsorption/desorption at −196 °C using a Micromeritics ASAP 2020 instrument. Samples (0.03 g) were degassed under vacuum (<0.1 Pa) at 250 °C for 30 min before measurement. The specific surface area (S₈₅₆) was calculated using the Brunauer-Emmett-Teller (BET) multiple-point method in the relative pressure range p/p₀ from 0.05 to 0.3, while total pore volume Vₚ was calculated at p/p₀ = 0.99.

High resolution transmission electron microscopy (HRTEM) characterization was carried out by a TEM/STEM FEI Tecnai F20 microscope, equipped with an EDS analyzer. Fine powder of catalysts was suspended in ethanol under ultrasonication for 20 min. The suspension was subsequently deposited on a Cu grid with lacy quasi-foil carbon film and dried at 100 °C before doing the measurement.

The X-ray photoelectron spectra (XPS) of selected mixed metal oxides were measured using a VSW spectrometer equipped with a hemispherical analyzer. The photoelectron spectra were measured using a magnesium MgKα source (E = 1253.6 eV). The base pressure in the analysis chamber during the measurements was 3 × 10⁻⁹ Pa and the spectra were calibrated on a main carbon C 1s peak at 284.6 eV. The composition and chemical surrounding of the sample surface were investigated based on the areas and binding energies of Cu 2p, Co 2p, Al 2p, Nd 3d, C 1s and O 1s photoelectron peaks.

Mathematical analyses of the XPS spectra were carried out using the XPSpeak 4.1 computer software (RWM Kwok, The Chinese University of Hong Kong).

The H₂ temperature programmed reduction (H₂-TPR) of mixed metal oxides were measured using a Quantachrome ChemBET Pulsar TPR/TPD. Prior to measurement, samples (30 mg) were degassed at 600 °C for 1 h in pure Ar (20 ml min⁻¹), and subse-

### Table 4. Comparison of the catalytic activity with Cu- and Co-based catalysts in the recent literature.

| Sample name | Preparation method (Calcination temperature [°C]) | Reaction conditions | N₂O conversion [%] (temperature [°C]) | Ref. |
|-------------|---------------------------------------------------|---------------------|----------------------------------------|------|
| CuO         | Precipitation method (400)                        | 5000 ppm N₂O, 20 000 ppm O₂/He, W/F = 0.3 g ml⁻¹ | 50 (365) | [70] |
| Co₃O₄      | Thermal decomposition (500)                       | 1000 ppm N₂O/N₂, or *wet conditions 1000 ppm N₂O, 20 000 ppm O₂, 200 ppm NO, 5000 ppm H₂O/N₂, W/F = 0.2 g ml⁻¹ (GHSV = 54,000 h⁻¹) | 50 (382) | [71] |
| Co₃O₄/Co₂O₃ | Coprecipitation (500)                             | 10 000 ppm N₂O/He, or *wet conditions 10 000 ppm N₂O, 50 000 ppm O₂, 20 000 ppm H₂O, 800 ppm NO, 60 ml min⁻¹, GHSV 60,000 h⁻¹, 0.1 g catalyst | 50 (370) | [72] |
| CuO/ZrO₂    | Coprecipitation (500)                             | 4000 ppm N₂O/He, GHSV 45,000 h⁻¹, 0.25 g catalyst, 50 ml min⁻¹ | 50 (440) | [73] |
| CuO/CeO₂    | Coprecipitation (500)                             | 500 ppm N₂O/N₂, or *wet conditions 500 ppm N₂O, 30 000 ppm O₃, 30 000 ppm H₂O/N₂, GHSV 45,000 h⁻¹ | 50 (340) | [74] |
| CuO–CeO₂    | Hydrothermal method (500)                         | 2000 ppm N₂O, 20 000 ppm O₂, GHSV 70,000 h⁻¹, 0.2 g catalyst | 50 (426) | [75] |
| CuO/CeO₂    | Citrate acid method (450)                         | 2600 ppm N₂O/He, GHSV 19,000 h⁻¹, p = 0.3 MPa | 50 (370) | [76] |
| CuO/CeO₂    | Hard template (KIT-6) repl. (400)                 | 2500 ppm N₂O, 15 000 ppm NO or 15 000 H₂O/Ar, GHSV 45,000 h⁻¹, 50 mg catalyst | 50 (465, with NO) | [77] |
| CuO/CoOₓ(10 wt.% Cu) | Glycol thermal approach (400) | 2500 ppm N₂O, 15 000 ppm NO or 1 5 000 ppm H₂O/Ar, GHSV 45,000 h⁻¹, 50 mg catalyst, 50 ml min⁻¹ | 50 (467) | [78] |
| LaCoO₃      | Solution combustion synthesis (600)               | 1000 ppm N₂O, 30 000 ppm O₃, 1000 ppm NO, 5000 ppm H₂O/He, 0.7 g catalyst, 250 ml min⁻¹ | 50 (531) | [79] |
| CuO/BEA(6.5 wt.% Cu) | Coprecipitation (600) | 5000 ppm N₂O/He or 5000 ppm N₂O + 5 % O₂/He, 50 mg catalyst + 200 mg silica, 200 ml min⁻¹ | 50 (445) | [80] |
| CoBEA(2.5 wt.% Co) | Ions-exchanged (550) | 1000 ppm N₂O/He, or *wet conditions 1000 ppm N₂O, 40 000 ppm O₂, 200 ppm NO, 30 000 ppm H₂O, 0.1 g catalyst, 50 ml min⁻¹ | 50 (380) | [81] |
| Ndₓ/Coₓ/Alₓ/Al₂O₃ | Coprecipitation (600) | 1000 ppm N₂O/N₂, or *wet conditions 1000 ppm N₂O, 20 000 ppm O₂, 200 ppm NO, 2500 ppm H₂O/N₂, 0.30 g catalyst, 80 ml min⁻¹ | 50 (385) | [82] |
| K/Cu        | Impregnation (550)                                | 4000 ppm N₂O/He, GHSV 45,000 h⁻¹, 0.25 g catalyst, 50 ml min⁻¹ | 50 (440) | [83] |
quently cooled down to room temperature. H₂-TPR runs were carried out starting from room temperature to 1000 °C, with a linear heating rate of 10 °K·min⁻¹ and in a flow (25 ml·min⁻¹) of 5.0 vol% H₂/Ar. Water vapor was removed from effluent gas by means of a cold trap placed in an ice-water bath.

The temperature programmed desorption of oxygen (O₂-TPD) of selected mixed metal oxides were measured using an AutoChem II (Chemisorption analyzer, Micromeritics) coupled with an online Mass spectrometer (Cirrus 2, MKS Instrument). Prior to measurement, samples (150 mg) were degassed at 400 °C for 1 h in pure Ar (20 ml·min⁻¹) and subsequently cooled down to 120 °C. Afterwards, 10 vol% O₂/He (20 ml·min⁻¹) was flushed through the catalysts at 120 °C for 1 h. Finally, the system was cooled down to 40 °C before starting O₂-TPD runs by heating up from 40 °C to 900 °C in pure He (30 ml·min⁻¹). The signals were recorded by both thermal conductive detector (TCD) and mass spectrometer (m/z = 32 and 44) to discriminate the contribution of CO₂ for interpretation of oxygen evolution.

**Catalytic Tests**

The experiments were carried out under atmospheric pressure in a fixed-bed flow microreactor (ID, 8 mm; L, 320 mm). Prior to the reaction, the catalysts (300 mg, particle size 0.25–0.5 mm) were outgassed at 600 °C for 0.5 h in a flow of pure N₂ (80 ml·min⁻¹). The catalysts were then cooled down to 50 °C under the same N₂ flow before switching to the reactant. The total flow rate of the feed stream was 80 ml·min⁻¹, controlled by mass flow controllers. The temperature was raised in steps of 50 °C starting from 50 up to 600 °C. Each temperature was set constant for 0.5 h. The stability test was carried out at a constant temperature of 450 °C for 50 h with the presence of the inhibitors. The conversion of N₂O (X(N₂O)) was estimated according to X(N₂O) = ([c(N₂O)in]–[c(N₂O)out])/[c(N₂O)in]×100 %, where c(N₂O)in and c(N₂O)out – concentration of N₂O in the inlet gas, and concentration of N₂O in the outlet gas.

The reaction rate was calculated by r = n×X×S/m²g where: n-molar flow rate of N₂O/mol s⁻¹, X-N₂O conversion under N₂O/N₂ conditions at 350 °C for Nd₂Cu₁.₆Al₂₃ and 250 °C for Nd₂CuAl₂ catalysts, m-weight g, S/m²g-specific surface area/m²·g⁻¹. It is noted that the reaction rate represents a true value only under differential reactor conditions (conversion lower than 20%), otherwise being an average reaction rate. Here we report the reaction rate at 250 °C for Nd₂CuAl₂₃ and 350 °C for Nd₂CuAl₂ catalysts to ensure that the conversions were low enough for reliable values of the reaction rate.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Keywords:** neodymium · mixed metal oxides · coprecipitation · N₂O decomposition · transition metal-based catalysts

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