N$_2$O decomposition over structurally promoted Ir/Al$_2$O$_3$ catalysts

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Introduction

Nitrous oxide (N$_2$O) has been recently recognized as a powerful greenhouse gas as well as the dominant ozone-depleting contributor. Its catalytic decomposition to N$_2$ consists one of the most efficient technologies for N$_2$O abatement, while noble metal supported catalysts, in particular Rh, Pt and Pd, seem to exhibit the best activity [1]. In this work the N$_2$O decomposition efficiency of Al$_2$O$_3$ supported Ir catalysts is studied. Moreover, the impact of rare earth oxides (CeO$_2$ or Ce$_{0.8}$La$_{0.2}$O$_{1.9}$) as structural modifiers of Al$_2$O$_3$-based Ir catalysts is explored. Extensive characterization was undertaken to reveal the structure-activity correlation.

Materials and Methods

A series of Ir catalysts over alumina (Al$_2$O$_3$) and structurally promoted alumina (AlCe: consisting of 80 wt % Al$_2$O$_3$-20 wt % CeO$_2$ and AlCeLa: consisting of 80 wt % Al$_2$O$_3$-20 wt % Ce$_{0.8}$La$_{0.2}$O$_{1.9}$) supports were tested. All supports were synthesized via the co-precipitation method, while the Ir active phase (0.5 wt %) was impregnated using IrCl$_3$·H$_2$O as the precursor salt. All catalytic materials were characterized via: BET, XRD, HR-TEM and TPR-H$_2$ and tested for N$_2$O decomposition in a bench-scale fixed bed reactor unit.

Results and Discussion

The XRD patterns and TPR-H$_2$ profiles of all three Iridium catalysts are presented in **Figure 1**. Interestingly, the presence of IrO$_2$ is evidenced from the XRD patterns (peaks at 2θ=28°, 34.7° & 54°), in spite the very low metal loading (0.5 wt%). During TPR analysis, all catalysts (Ir/ Al$_2$O$_3$, Ir/ AlCe and Ir/ AlCeLa) exhibit a well-defined reduction peak at 220-226°C, which may be attributed to the reduction of this IrO$_2$ phase [2].

XRD detection of IrO$_2$ phase can be ascribed to the formation of large Ir particles, as confirmed by HR-TEM analysis (**Figure 2a**). In the case of Ir/Al$_2$O$_3$ catalyst, Ir seems to be additionally present at high dispersion, forming Ir clusters on alumina. On the contrary, Ir was solely detected as large crystalline IrO$_2$ particles, on both Ir/AlCe and Ir/AlCeLa catalysts.

Catalytic performance in N$_2$O decomposition in the absence and presence of oxygen is presented in **Figure 2b.** In the absence of O$_2$, all three Ir-based catalysts present similar catalytic behavior, achieving N$_2$O conversions as high as ~100% at 600°C, suggesting no significant effect of structural modifiers. On the contrary, in the presence of O$_2$ the deN$_2$O efficiency is significantly enhanced, when modifying the alumina support with Ce and La. The highest initial N$_2$O conversion (~66% at 600°C) of Ir/Al$_2$O$_3$ was enhanced to 90% and ~96% over the structurally-promoted Ir/AlCeLa and Ir/AlCe catalysts respectively. In addition, the N$_2$O conversion profiles were shifted to significantly (up to 100 °C) lower temperatures.
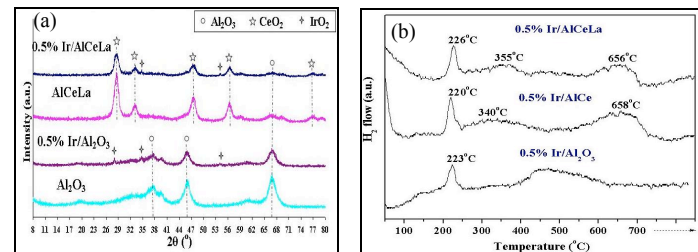

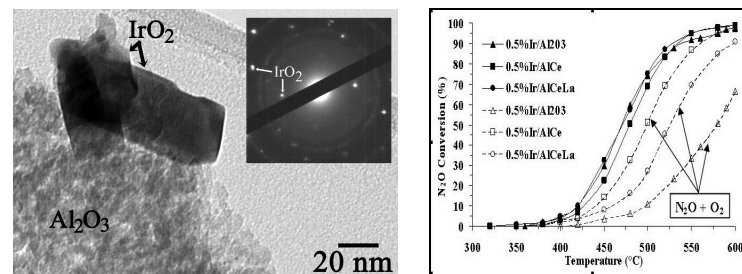**Figure 1**. Characterization results (a) XRD patterns and (b) TPR-H$_2$ profiles of Ir catalysts.

Figure 2. (a) TEM image and SAD pattern (inset) of Ir/ Al$_2$O$_3$ catalyst and (b) N$_2$O conversion in the absence and presence O$_2$ over all Ir catalysts (Feed: 1000 ppm N$_2$O, 0 or 2% O$_2$ in He).

Significance

So far, noble metal-based catalysts (in particular Rh, Pt and Pd) are presented as the most efficient for the de-N$_2$O process. However, their scarcity and consequent high price render their utilization unadvisable. On the contrary, Ir-based catalysts seem to be promising alternatives for N$_2$O decomposition.

References

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