H_2 AND/OR C_3H_6 ASSISTED SELECTIVE CATALYTIC REDUCTION OF NOx OVER Ir/ACZ CATALYSTS

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Selective Catalytic Reduction with C_3H_6

Hydrothermal Ir/ACZ

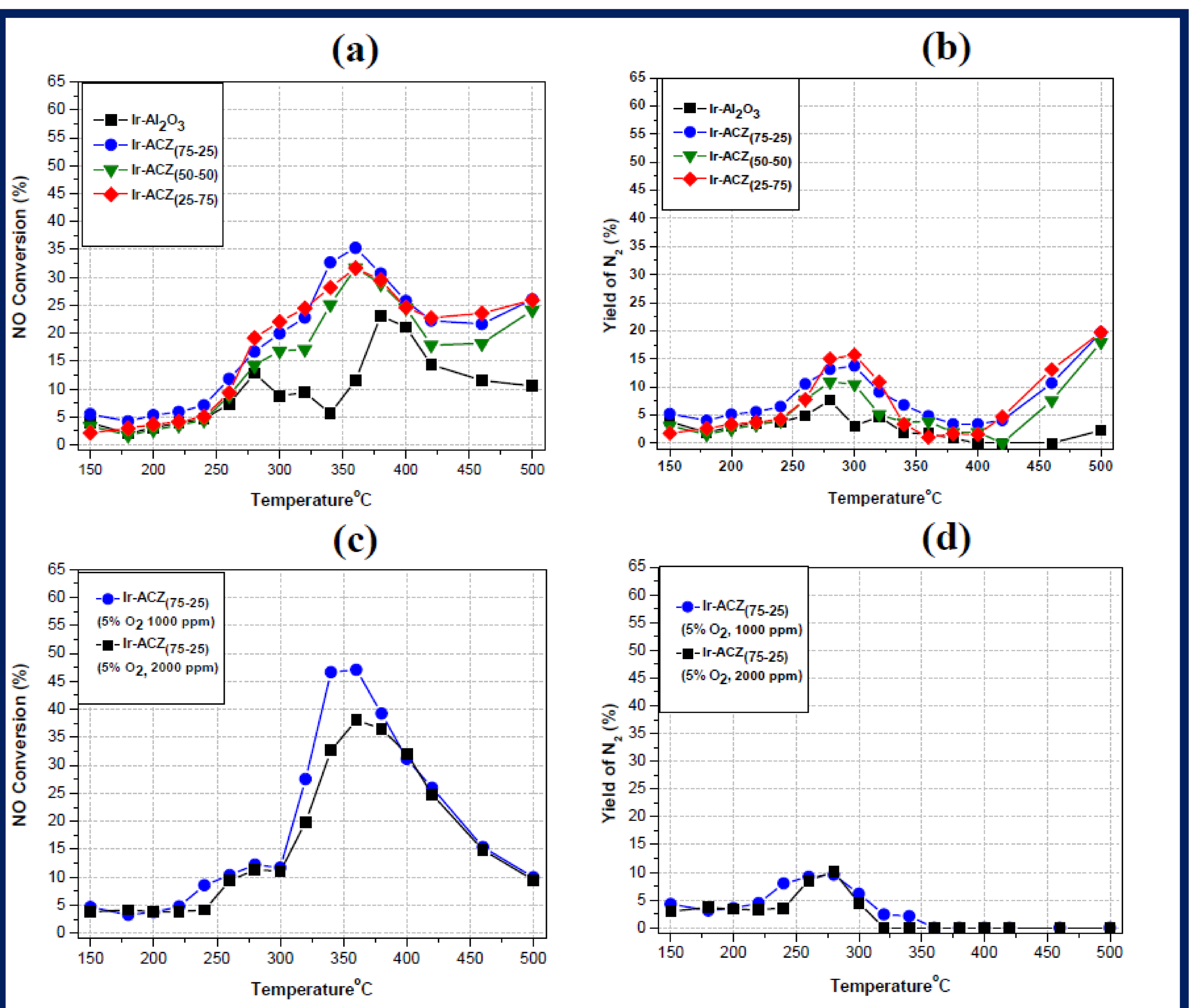

Figure 1: Hydrothermal catalysts: (a) NO conversion, and (b) Yield of N_2 (for both (a) and (b) reaction conditions: 1000 ppm NO, 1000ppm C_3H_6, 2% O_2). Ir-ACZ(75-25): (c) NO conversion, and (d) Yield of N_2 (for both (c) and (d) reaction conditions: 1000 ppm NO (1000ppm C_3H_6) vs 2000ppm NO (2000 ppm C_3H_6), 5% O_2)

Co-precipitation Ir/ACZ

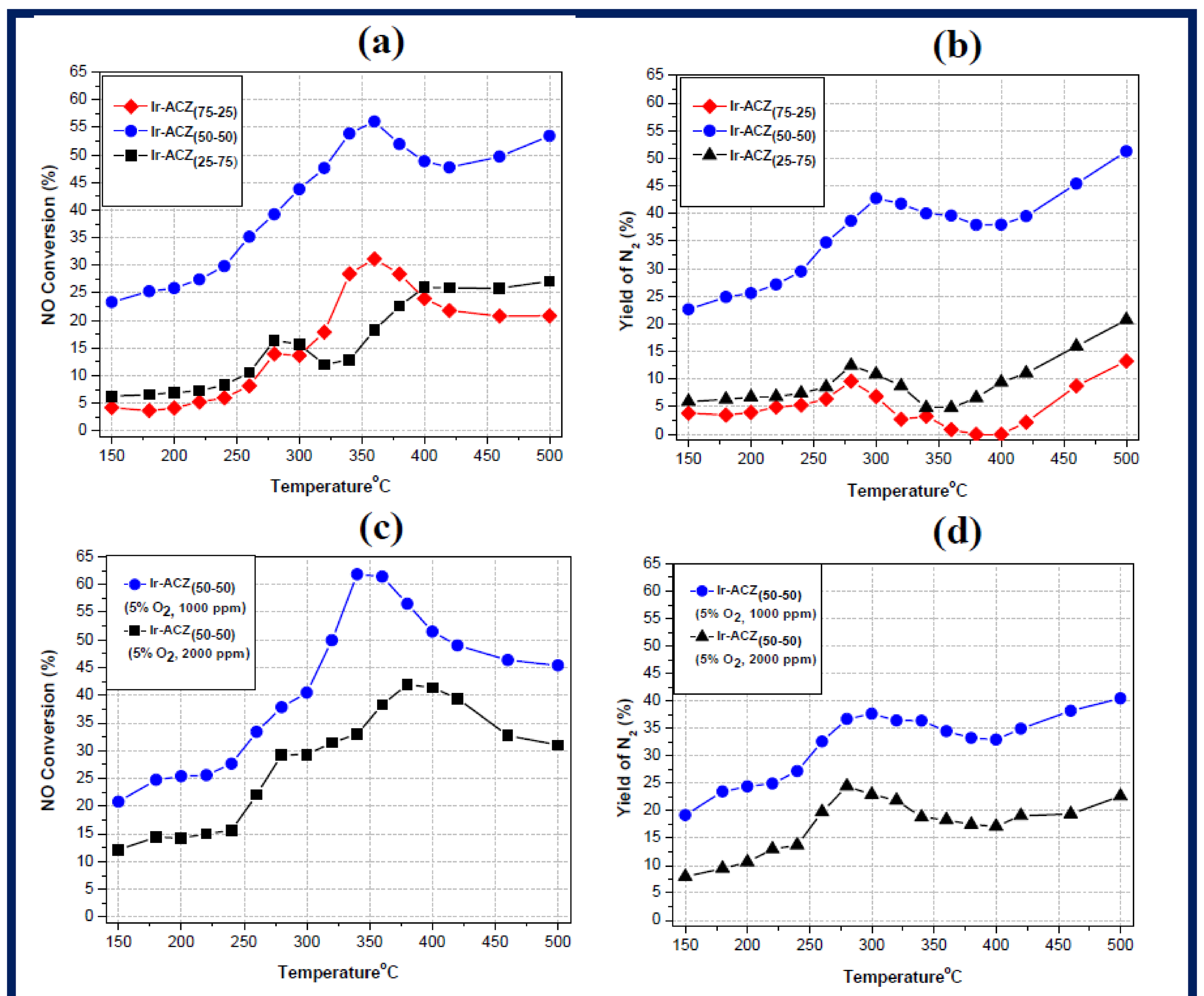

Figure 2: Co-precipitation catalysts: (a) NO conversion, and (b) Yield of N_2 (for both (a) and (b) reaction conditions: 1000 ppm NO, 1000 ppm C_3H_6, 2% O_2). Ir-ACZ(75-25): (c) NO conversion, and (d) Yield of N_2 (for both (c) and (d) reaction conditions: 1000 ppm NO (1000 ppm C_3H_6) vs 2000ppm NO (2000 ppm C_3H_6), 5% O_2)

Selective Catalytic Reduction with H_2

Hydrothermal Ir/ACZ

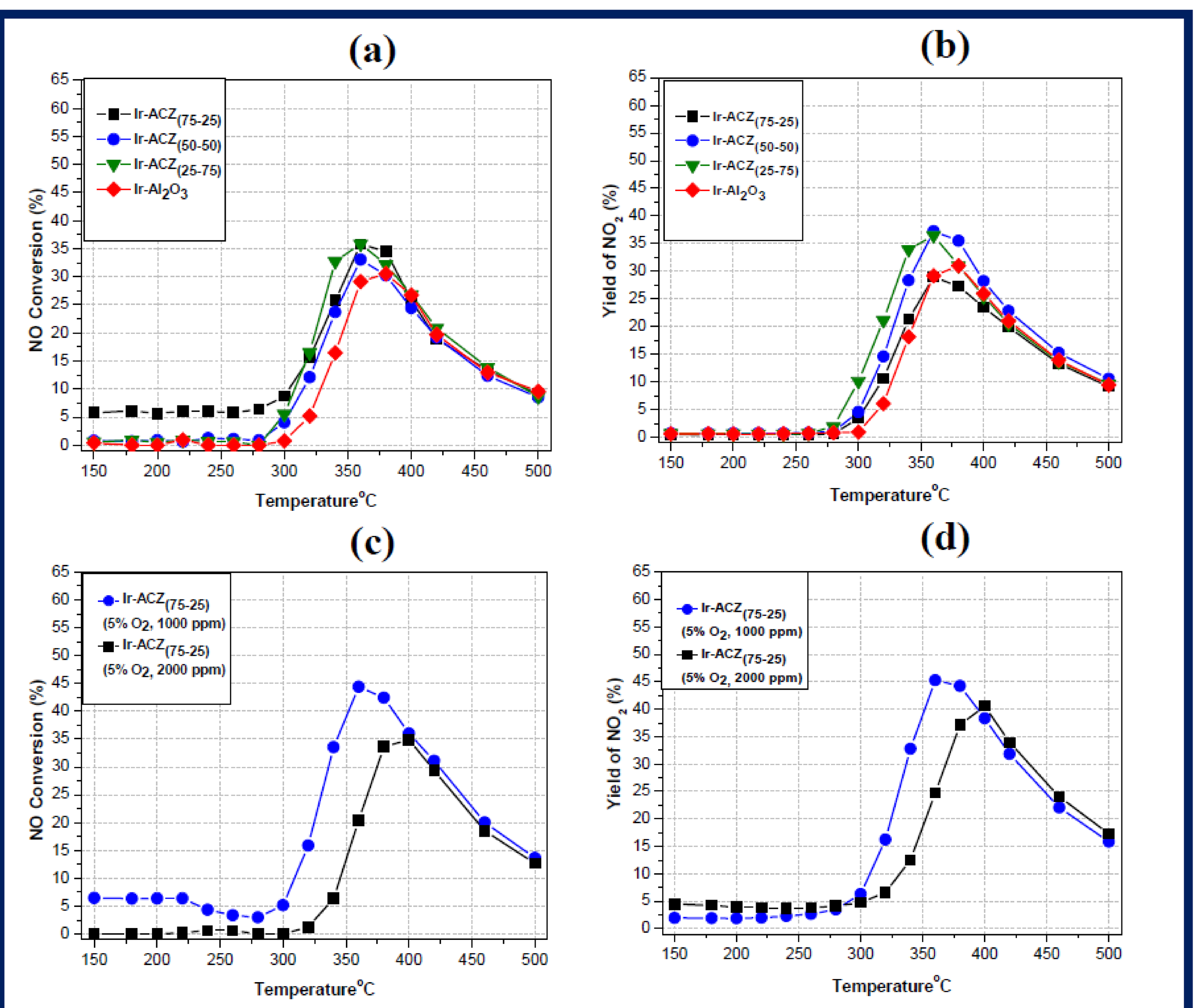

Figure 3: Hydrothermal catalysts: (a) NO conversion, and (b) Yield of NO_2 (for both (a) and (b) reaction conditions: 1000 ppm NO, 1000 ppm H_2, 2% O_2). Ir-ACZ(75-25): (c) NO conversion, and (d) Yield of NO_2 (for both (c) and (d) reaction conditions: 1000 ppm NO (1000 ppm H_2) vs 2000 ppm NO (2000 ppm H_2), 5% O_2)

Co-precipitation Ir/ACZ

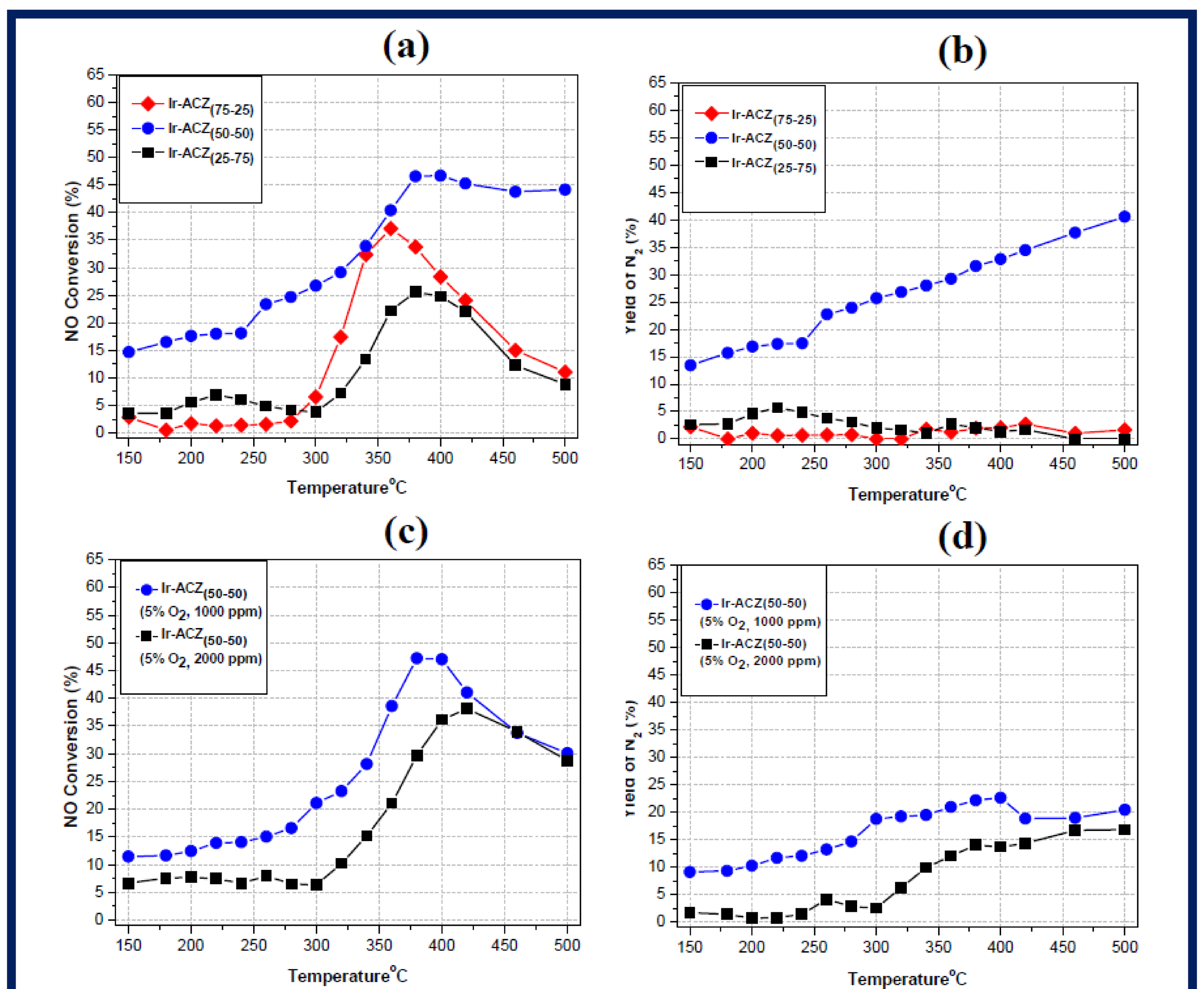

Figure 4: Co-precipitation catalysts: (a) NO conversion, and (b) Yield of N_2 (for both (a) and (b) reaction conditions: 1000 ppm NO, 1000 ppm H_2, 2% O_2). Ir-ACZ(75-25): (c) NO conversion, and (d) Yield of N_2 (for both (c) and (d) reaction conditions: 1000 ppm NO (1000 ppm H_2) vs 2000 ppm NO (2000ppm H_2), 5% O_2)

Selective Catalytic Reduction with C_3H_6 + H_2

Hydrothermal Ir/ACZ

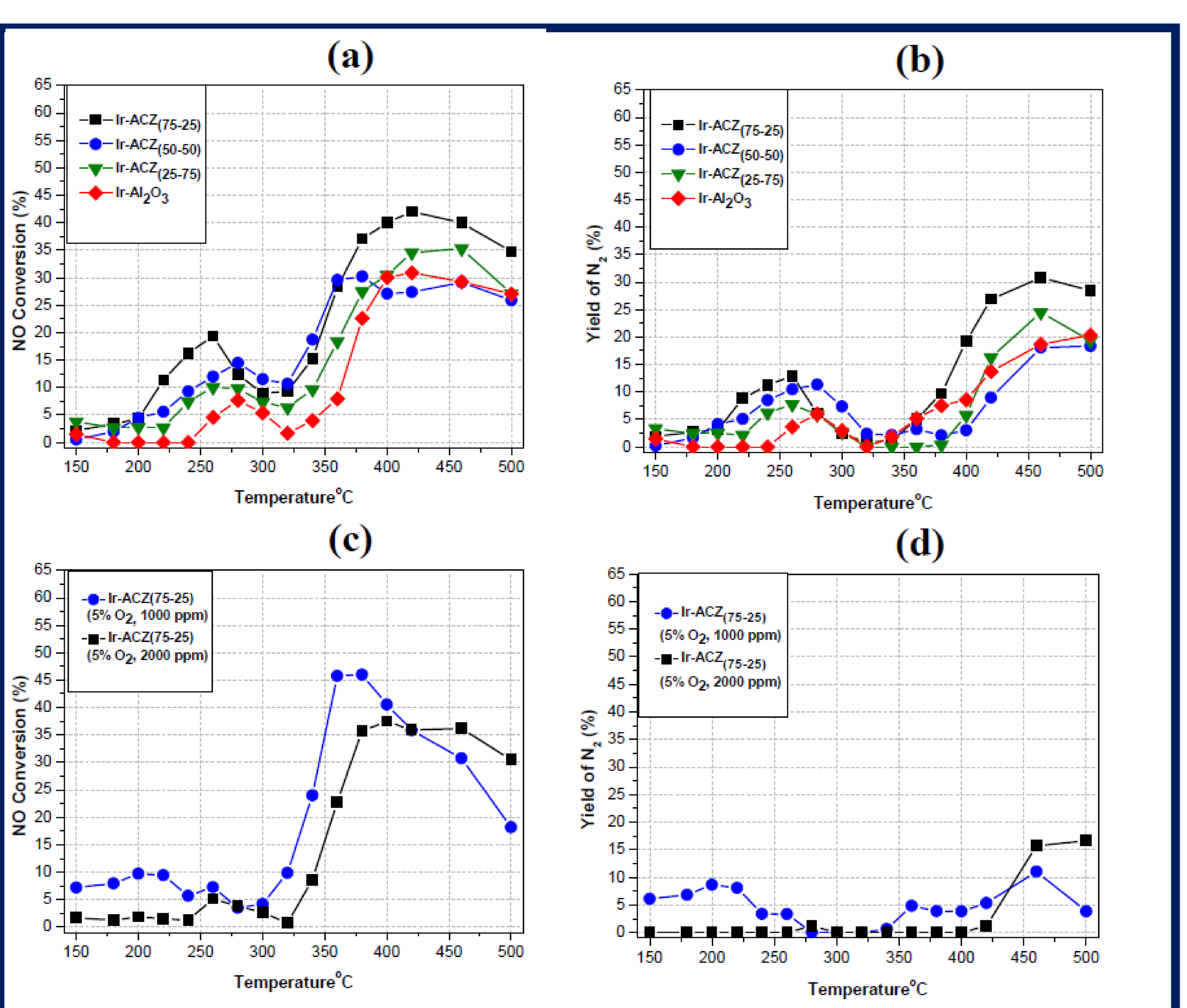

Figure 5: Hydrothermal catalysts: (a) NO conversion, and (b) Yield of N_2 (for both (a) and (b) reaction conditions: 1000 ppm NO, 1000 ppm C_3H_6, 1000 ppm H_2, 2% O_2). Ir-ACZ(75-25): (c) NO conversion, and (d) Yield of N_2 (for both (c) and (d) reaction conditions: 1000 ppm NO (1000 ppm C_3H_6, 1000 ppm H_2) vs 2000 ppm NO (2000 ppm C_3H_6, 2000 ppm H_2, 5% O_2)

Co-precipitation Ir/ACZ

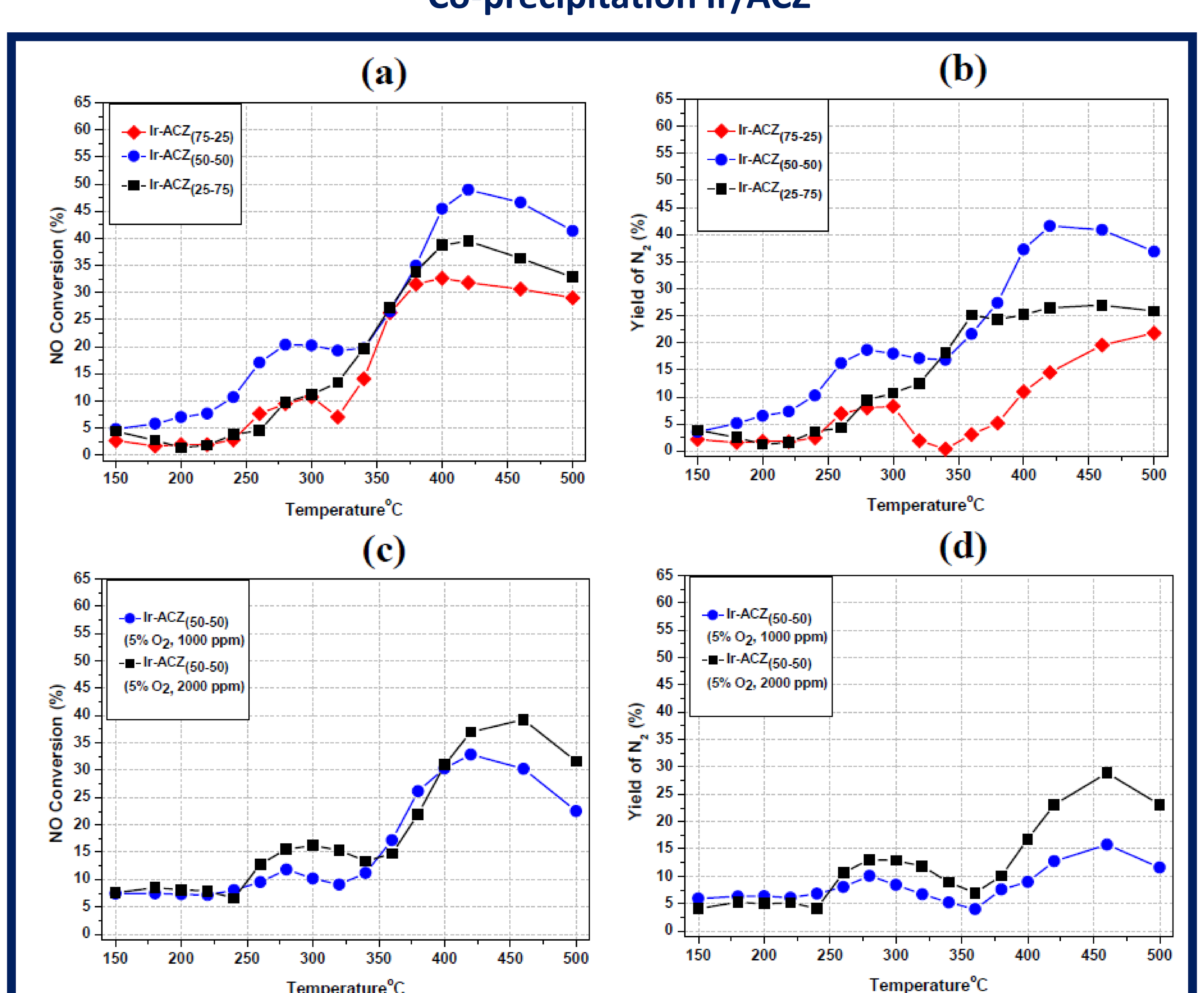

Figure 6: Co-precipitation catalysts: (a) NO conversion, and (b) Yield of N_2 (for both (a) and (b) reaction conditions: 1000ppm NO, 1000 ppm C_3H_6, 1000ppm H_2, 2% O_2). Ir-ACZ(75-25): (c) NO conversion, and (d) Yield of N_2 (for both (c) and (d) reaction conditions: 1000 ppm NO (1000 ppm C_3H_6, 1000 ppm H_2) vs 2000ppm NO (2000 ppm C_3H_6, 2000ppm H_2, 5% O_2)

Summary

Selective Catalytic Reduction (SCR) is widely considered as one of the most effective aftertreatment technologies for the control of the emissions of nitrogen oxides (NOx) in the effluents of lean combustion. In the present work, the SCR of NO was investigated at the temperature range of 150-500 °C over low loading (1 wt.%) Ir catalysts based on Al_2O_3-CeO_2-ZrO_2 (ACZ) supports with different Al-Ce-Zr molar compositions. The study examined three reaction schemes (NO + C_3H_6 + O_2 (R#1), NO + H_2 + O_2 (R#2) and NO + C_3H_6 + H_2 + O_2 (R#3)) over two groups of catalysts, one with ACZ supports synthesized by a hydrothermal method and another with the ACZ supports synthesized by co-precipitation. The results showed that the maximum NO conversion was attained over the Ir-ACZ (C:Z=75:25) hydrothermal catalyst and the Ir-ACZ (C:Z=50:50) co-precipitated catalyst regardless of the reducing agent in the reaction mixture. An increase of the O_2 concentration from 2% to 5% increased the conversion of NO, but led to a decline in the yield towards N_2, for both catalysts.

Introduction

The combustion of fossil fuels in both transportation and industrial applications is known to produce significant emissions of nitrogen oxides, i.e., NOx (NO and NO_2) [1-4]. Selective Catalytic Reduction (SCR) is widely considered as one of the most effective aftertreatment technologies for the control of nitrogen oxides in the combustion effluents [5-8]. The method intends the heterogeneous catalytic reduction of NOx in the presence of a homogeneous reducing agent and is particularly suitable for oxygen-rich effluents coming from lean combustion applications, as it is the case in residential and industrial burners or in vehicular diesel engines [9-12]. The present experimental work investigates the SCR of NOx over Ir/Al_2O_3-CeO_2-ZrO_2(ACZ) catalysts prepared by two different methods (hydrothermal and co-precipitation) and in the presence of three alternative reducing agents: C_3H_6 (R#1), H_2 (R#2) and C_3H_6 + H_2 (R#3).

Catalytic Activity Tests

Ir/ACZ (75-25), Ir/ACZ (50-50), Ir/ACZ (25-75) and Ir/Al_2O_3 catalysts were tested at the same experimental conditions for all the reducing agents. The initial gas mixture composition was 1000 ppm or 2000 ppm NO, 1000 ppm or 2000 ppm C_3H_6 (when appropriate), 1000 ppm or 2000 ppm H_2 (when appropriate), 2 or 5% O_2 and inert Ar as balance at 1 bar. For each experiment 0.18 g of catalyst was used and was positioned in a fixed bed quartz tubular flow reactor. Then experiments were carried out keeping the gas hourly space velocity equal to 100,000 h^{-1}.

Conclusions

In the work presented herein, Ir catalysts with a low loading (1 wt.%), supported on Al_2O_3-CeO_2-ZrO_2 (ACZ), were tested for the selective catalytic reduction of NO. The supports were prepared using two different synthesis methods, (a) hydrothermal and b) co-precipitation; for each method three different Ce:Zr molar compositions were utilized (75:25, 50:50, and 25:75). The as prepared catalysts were tested in the temperature range of 150-500 °C under three reaction schemes: NO + C_3H_6 + O_2 (R#1), NO + H_2 + O_2 (R#2) and NO + C_3H_6 + H_2 + O_2 (R#3). The results showed that the maximum NO conversion was attained over the Ir-ACZ (C:Z=75:25) hydrothermal catalyst and the Ir-ACZ (C:Z=50:50) co-precipitated catalyst regardless of the reducing agent in the reaction mixture. An increase of the O_2 concentration from 2% to 5% increased the conversion of NO but led to a decline in the yield towards N_2, for both catalysts.

References

1. M.A. Goula, et al. J. Env. Chem. Eng. 2016, 4, 1629–1641.
2. Z. Gholami et al. Catal. Rev. 2020, 68-119.
3. I.V. Yentekakis, at al. Appl. Catal. B Environ. 2005, 56, 229–239.
4. M. Konsolakis, et al. J. Catal. 2001, 198, 142–150.
5. E. Papista, et al. Top Catal. 2016, 59, 1020–1027.
6. H. Hamada, et al. Appl. Catal. A Gen. 2012, 421– 422.
7. A. Väliheikki, et al. Appl. Catal. B Environ. 2014, 156–157, 72–83.
8. C. Yin, et al. Catal Lett. 2015, 145, 1491–1499.
9. M. Ogura, et al. Chem. Lett. 2000, 29, 2, 146-147.
10. T. Yoshinari, et al. Appl. Catal. B Environ. 2003, 41, 157–169.
11. A. Wang, et al. Appl. Catal. B Environ. 2003, 40, 319–329.
12. M. Konsolakis, et al. Appl. Catal. B Environ. 2001, 33, 293–302.

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