

SUPPLEMENTARY INFORMATION

New multicomponent catalysts for the selective aerobic oxidative condensation of benzylamines to N-benzylidenbenzylamine

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Catalysts characterization

Figures S1 and S2 show the high-angle XRD patterns of all synthesized UVM-7-based materials, after and before the gold incorporation and thermally processed at 200 and 500°C.

Figures S3 and S4 correspond to TEM images and N₂ adsorption-desorption isotherms of the gold-free support/catalysts. The nickel inclusion does not affect the mesostructure when compared with a TEM image of UVM-7 pure silica. The two pore systems (intraparticle surfactant generated mesopore system and the interparticle disordered large pores) result clearly appreciated. The Ce and Sn inclusion although does not affect in a qualitative way the original mesostructure, the incorporation of these heteroelements favors a certain interparticle condensation with the subsequent relatively loss of textural-type porosity.

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Catalysts characterization

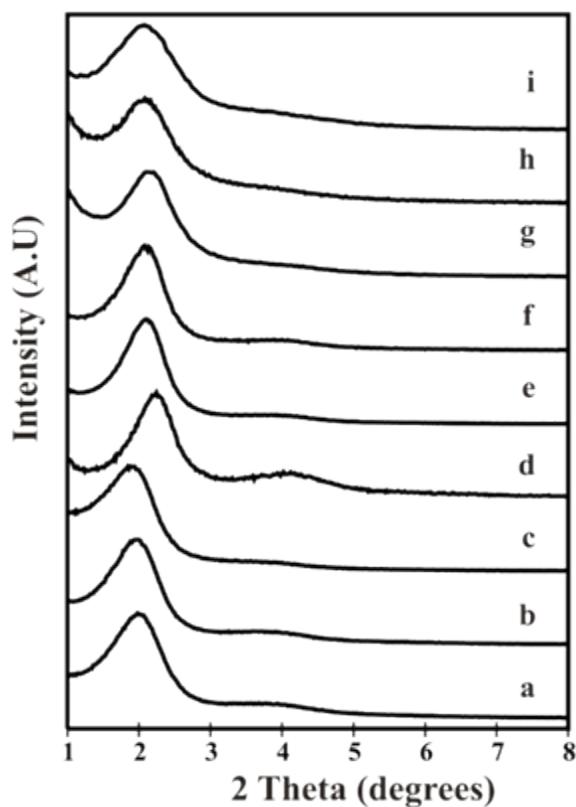


Fig. SI1. Low-angle XRD patterns of the gold-free supports. (a) Ni₆₀-UVM-7, (b) Ni₃₀-UVM-7, (c) Ni₁₀-UVM-7, (d) Ce₆₀-Ni₁₀-UVM-7, (e) Ce₃₀-Ni₁₀-UVM-7, (f) Ce₁₀-Ni₁₀-UVM-7, (g) Sn₆₀-Ni₁₀-UVM-7, (h) Sn₃₀-Ni₁₀-UVM-7 and (i) Sn₁₀-Ni₁₀-UVM-7.

Figures SI2 and SI3 show the high-angle XRD patterns of all synthesized UVM-7-based materials, after and before the gold incorporation and thermally processed at 200 and 500°C.

Figures SI4 and SI5 correspond to TEM images and N₂ adsorption-desorption isotherms of the gold-free support/catalysts. The nickel inclusion does not affect the mesostructure when compared with a TEM image of UVM-7 pure silica. The two pore systems (intraparticle surfactant generated mesopore system and the interparticle disordered large pores) result clearly appreciated. The Ce and Sn inclusion although does not affect in a qualitative way the original mesostructure, the incorporation of these heteroelements favors a certain interparticle condensation with the subsequent relatively loss of textural-type porosity.

Figs. SI5 and SI6 present N₂ adsorption desorption isotherms of catalysts : **SI5** (a) Au/Sn₁₀-Ni₁₀-UVM-7, (b) Au/Sn₃₀-Ni₁₀-UVM-7 and (c) Au/Sn₆₀-Ni₁₀-UVM-7, processed at (i) 200°C and (ii) 500°C. **SI6** (a) Au/Ce₁₀-Ni₁₀-UVM-7, (b) Au/Ce₃₀-Ni₁₀-UVM-7 and (c) Au/Ce₆₀-Ni₁₀-UVM-7, processed at (i) 200°C and (ii) 500°C

Figs. SI7 and SI8 present the BJH pore size distributions from the adsorption branches of the N₂ adsorption-desorption isotherms of catalysts: **SI7** (a) Au/Ce₁₀-Ni₁₀-UVM-7, (b) Au/Ce₃₀-Ni₁₀-UVM-7 and (c) Au/Ce₆₀-Ni₁₀-UVM-7, processed at (i) 200°C and (ii) 500°C. **SI8**. (a) Au/Sn₁₀-Ni₁₀-UVM-7, (b) Au/Sn₃₀-Ni₁₀-UVM-7 and (c) Au/Sn₆₀-Ni₁₀-UVM-7, processed at (i) 200°C and (ii) 500°C.

Figs. SI9 and SI10 present conversion results in recycling experiments

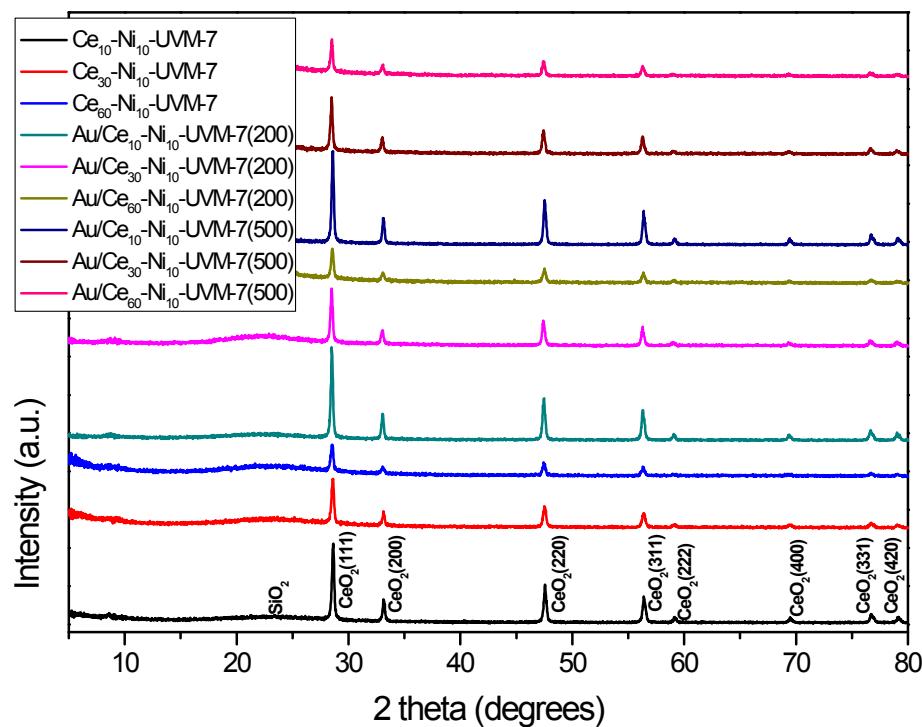


Fig. SI2. High-angle XRD patterns of the supports and final catalysts containing Ce.

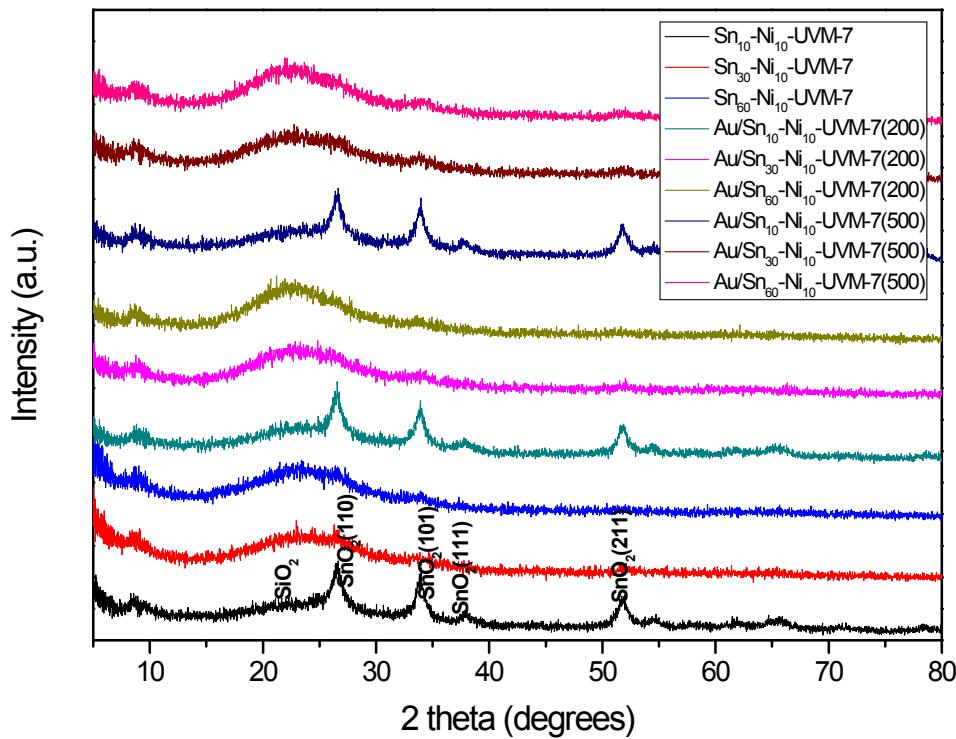


Fig. SI3. High-angle XRD patterns of the supports and final catalysts containing Sn.

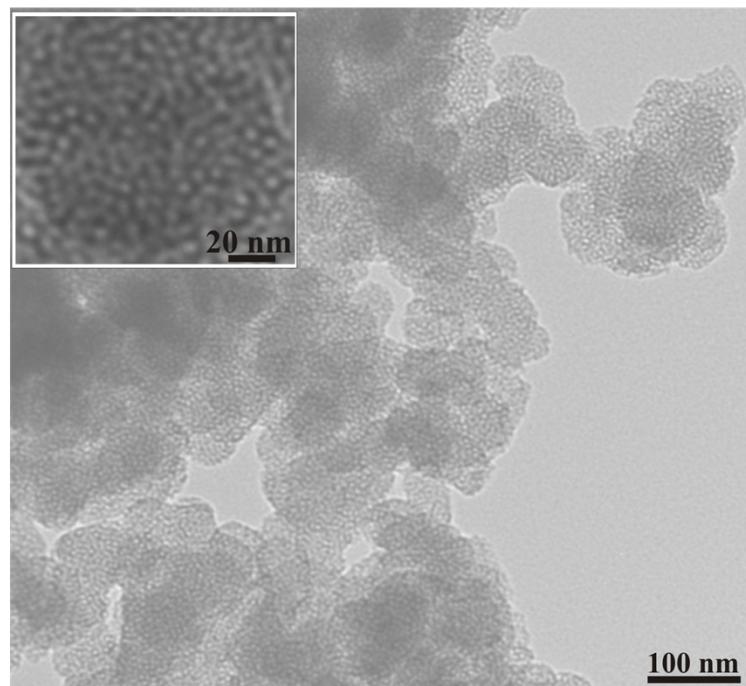


Fig. SI4. Representative TEM image of $\text{Ni}_x\text{-UVM-7}$ samples ($x=10$).

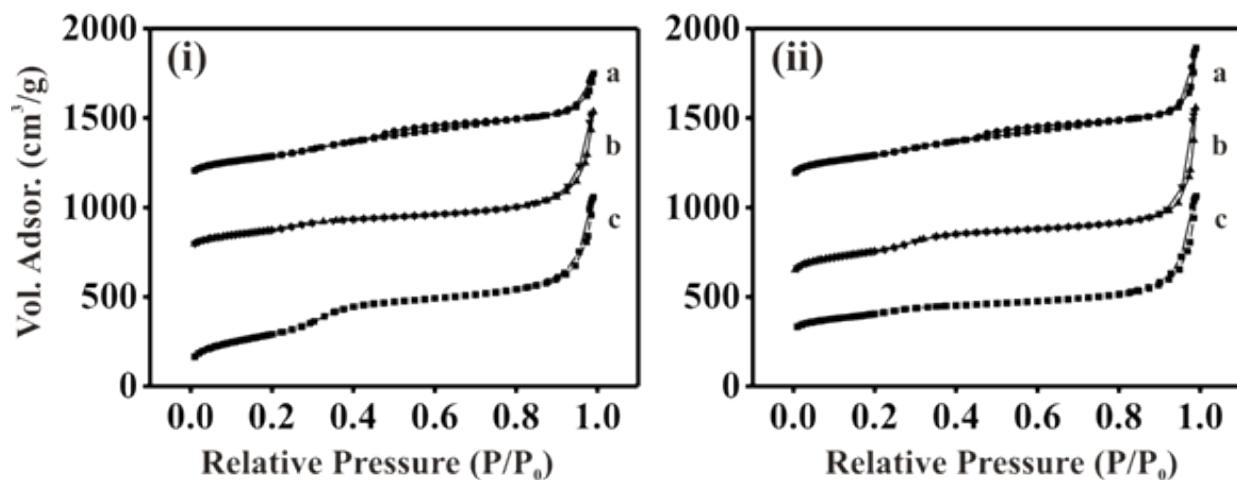


Fig. SI5. N_2 adsorption desorption isotherms of catalysts (a) $\text{Au/Sn}_{10}\text{-Ni}_{10}\text{-UVM-7}$, (b) $\text{Au/Sn}_{30}\text{-Ni}_{10}\text{-UVM-7}$ and (c) $\text{Au/Sn}_{60}\text{-Ni}_{10}\text{-UVM-7}$, processed at (i) 200°C and (ii) 500°C.

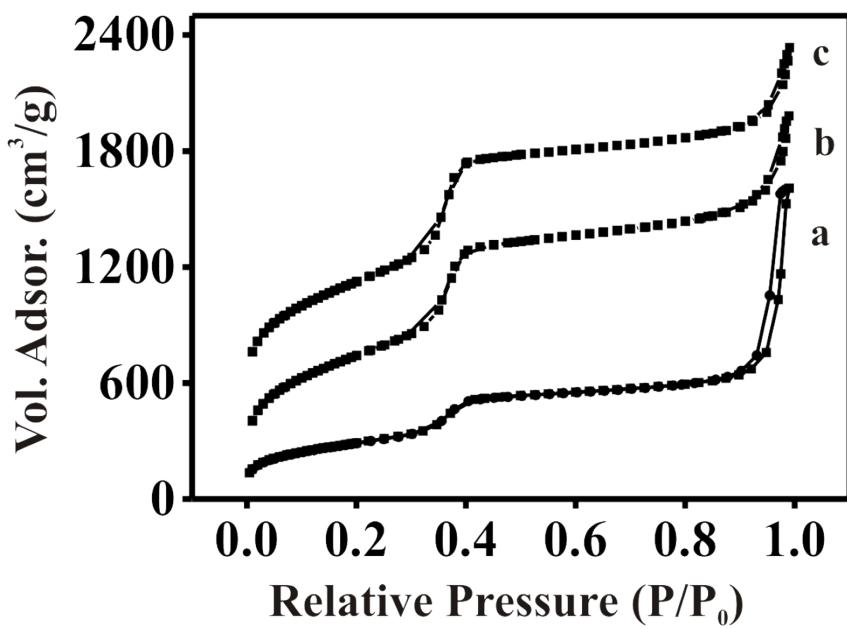


Fig. SI6. Representative N₂ adsorption-desorption isotherms of the gold-free supports. (a) Ni₆₀-UVM-7, (b) Ce₆₀-Ni₁₀-UVM-7 and (c) Sn₆₀-Ni₁₀-UVM-7.

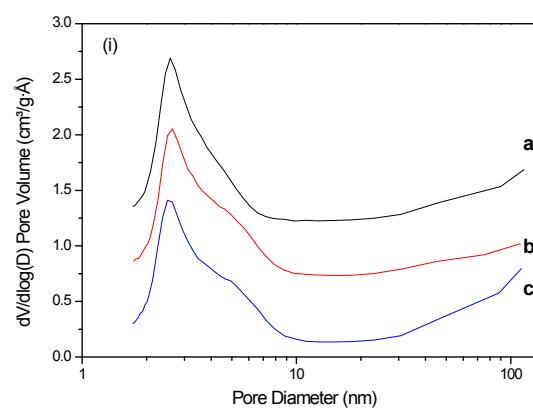
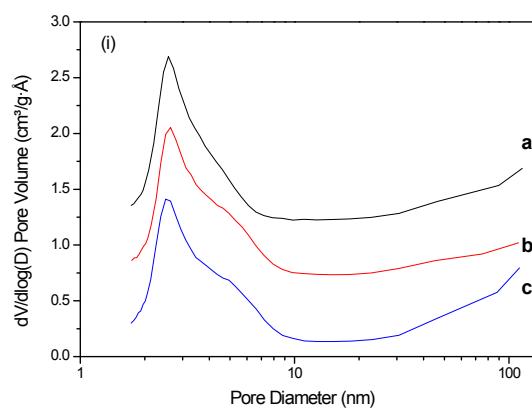


Fig. SI7. BJH pore size distributions from the adsorption branches of the N₂ adsorption-desorption isotherms of catalysts (a) Au/Ce₁₀-Ni₁₀-UVM-7, (b) Au/Ce₃₀-Ni₁₀-UVM-7 and (c) Au/Ce₆₀-Ni₁₀-UVM-7, processed at (i) 200°C and (ii) 500°C.

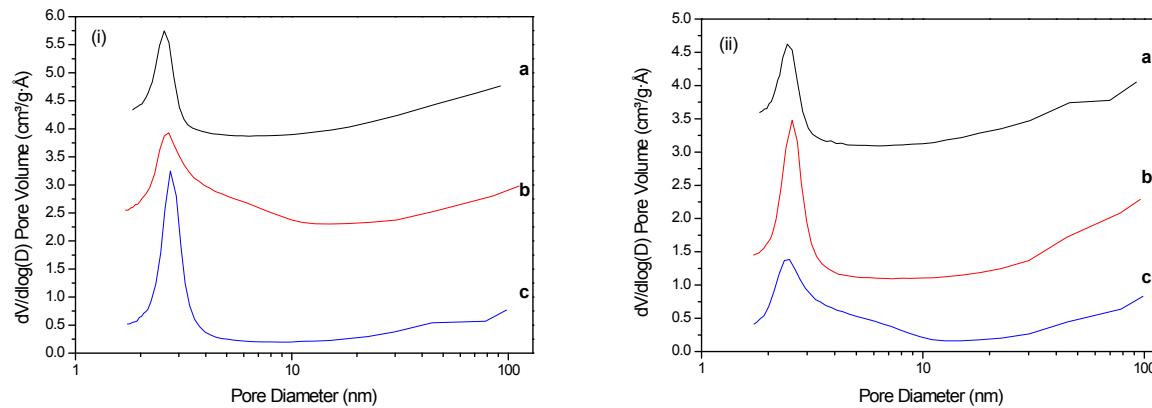
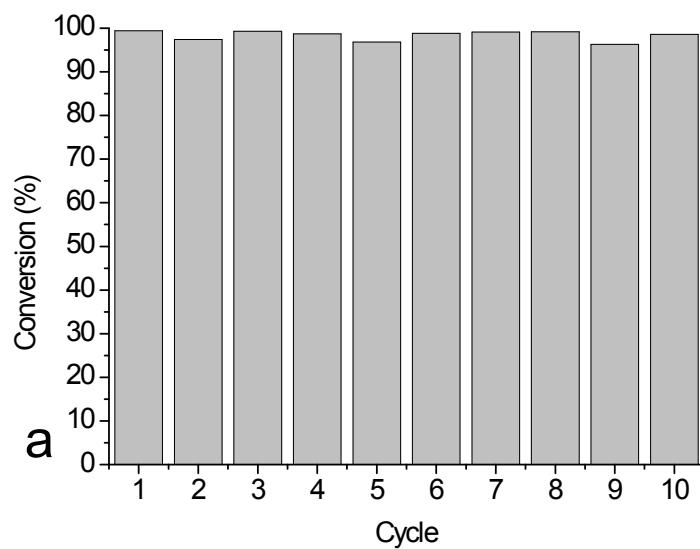


Fig. SI8. BJH pore size distributions from the adsorption branches of the N₂ adsorption desorption isotherms of catalysts (a) Au/Sn₁₀-Ni₁₀-UVM-7, (b) Au/Sn₃₀-Ni₁₀-UVM-7 and (c) Au/Sn₆₀-Ni₁₀-UVM-7, processed at (i) 200°C and (ii) 500°C.



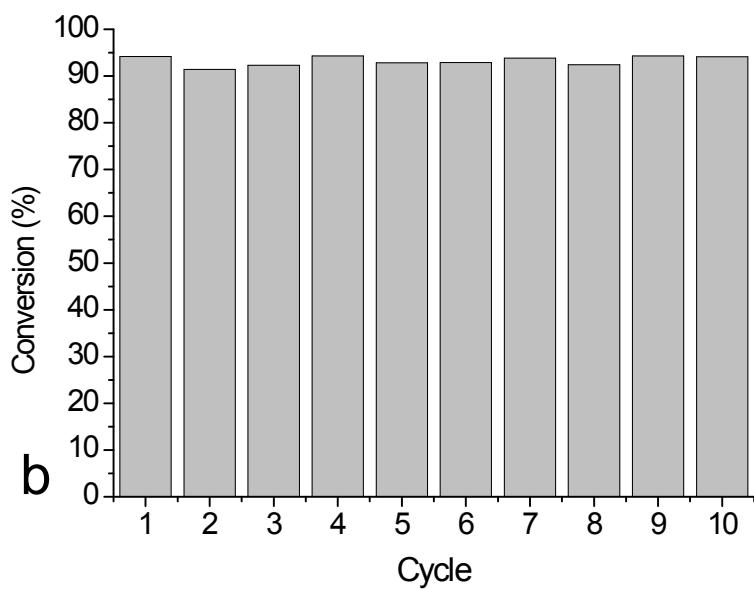


Fig. SI9. Conversion of benzylamine on $\text{Au}/\text{Ce}_{60}\text{-Ni}_{10}\text{-UVM-7(200)}$ (a) and $\text{Au}/\text{Ce}_{30}\text{-Ni}_{10}\text{-UVM-7(200)}$ (b) catalysts over 10 successive cycles at 5 bar O_2 (115°C , 4h, selectivity 100%).

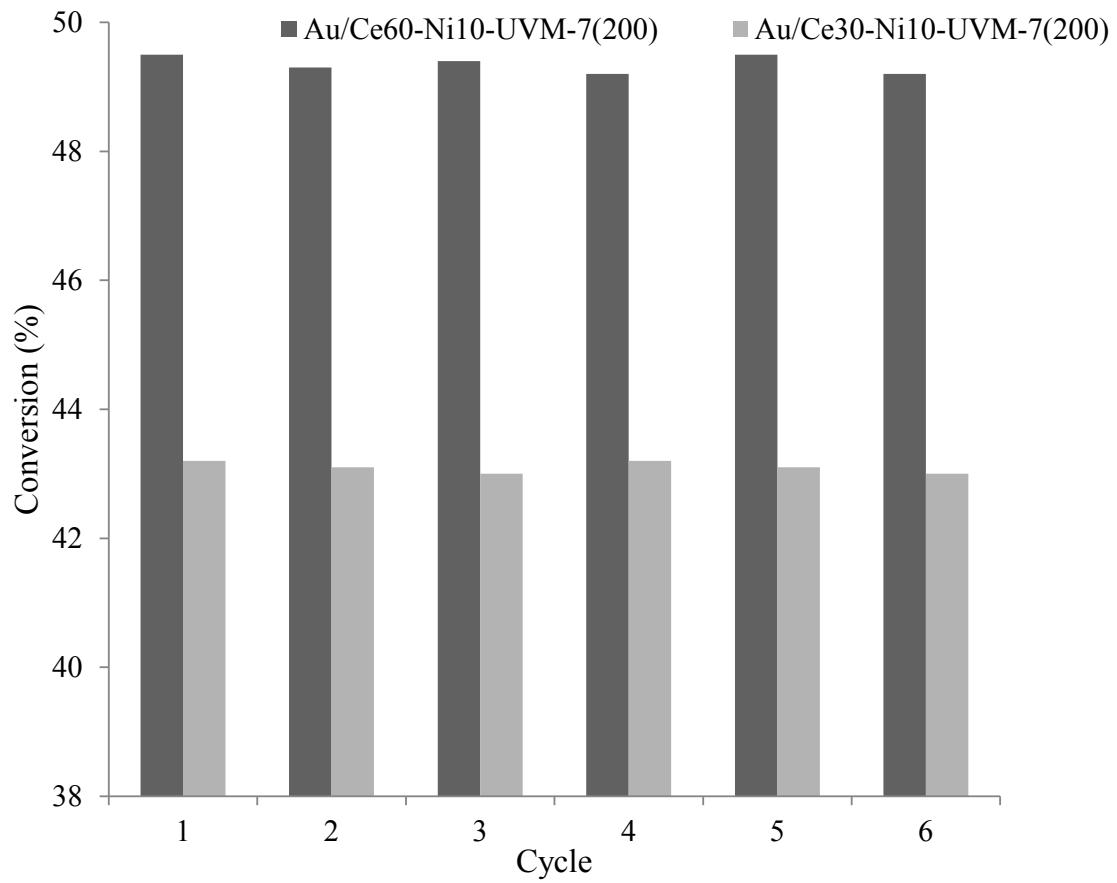


Fig. SI10. Conversion of benzylamine on Au/Ce₆₀-Ni₁₀-UVM-7(200) and Au/Ce₃₀-Ni₁₀-UVM- 7(200) catalysts over 6 successive cycles at 2 bar O₂ (115 °C, 4h, selectivity 100%).