

Innovative sol-gel routes to mesoporous bifunctional catalysts for the upgrading of bioethanol to butadiene

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Bioethanol production has been promoted by strong political and societal incentives. In parallel, butadiene is one of the compounds that is expected to suffer future shortage due to the shift towards shale gas from the traditional steam cracking of naphta. Therefore, intensive research is currently conducted towards the direct catalytic conversion of ethanol to butadiene. The reaction is long known, and consists of a complex network of dehydration and dehydrogenation reactions catalysed respectively by acid and redox active sites that must operate in a controlled and balanced fashion to maximize the butadiene yield.¹

The aerosol-assisted sol-gel (AASG) is a promising method to obtain nanomaterials with controlled properties (texture, surface chemistry, homogeneity), which showed great promises for the development of high-performance heterogeneous catalysts.²⁻⁴

Here, we present the first example of the preparation of bifunctional Ta-Ag-SiO₂ and Ta-Cu-SiO₂ mesoporous catalysts synthesized by AASG. After calcination, the catalysts display advantageous texture (SSA $\approx$ 450 m² g⁻¹; V_p $\approx$ 0.5 mL g⁻¹; D_p $\approx$ 4.5 nm) with good incorporation and dispersion of Ta atoms compared to impregnated counterparts as shown by XPS, IR and STEM-EDS. The best catalyst tested in the ethanol-to-butadiene reaction (2 wt.% Ta and 5 wt.% Cu) reached a 20 % butadiene yield at 325 °C.

- 1 Dochain, D. D. *et al.* Ag-and Cu-promoted mesoporous Ta-SiO₂ catalysts prepared by non-hydrolytic sol-gel for the conversion of ethanol to butadiene. *Catalysts* **9**, 920 (2019).
- 2 Debecker, D. P. *et al.* One-Pot Aerosol Route to MoO₃-SiO₂-Al₂O₃ Catalysts with Ordered Super Microporosity and High Olefin Metathesis Activity. *Angewandte Chemie International Edition* **51**, 2129-2131 (2012).
- 3 Smeets, V. *et al.* Aerosol route to TiO₂-SiO₂ catalysts with tailored pore architecture and high epoxidation activity. *Chemistry of Materials* **31**, 1610-1619 (2019).
- 4 Debecker, D. P. Innovative Sol-Gel Routes for the Bottom-Up Preparation of Heterogeneous Catalysts. *The Chemical Record* **18**, 662-675 (2018).

