

## Isotopic exchange studies in heterogeneous catalysis

For nearly 80 years, the isotopic exchange of oxygen was used to investigate the reactivity and mobility of lattice oxygen in oxide catalysts. Correlations have been proposed between activities in oxidation and oxygen exchange reactions (Fig. 1).

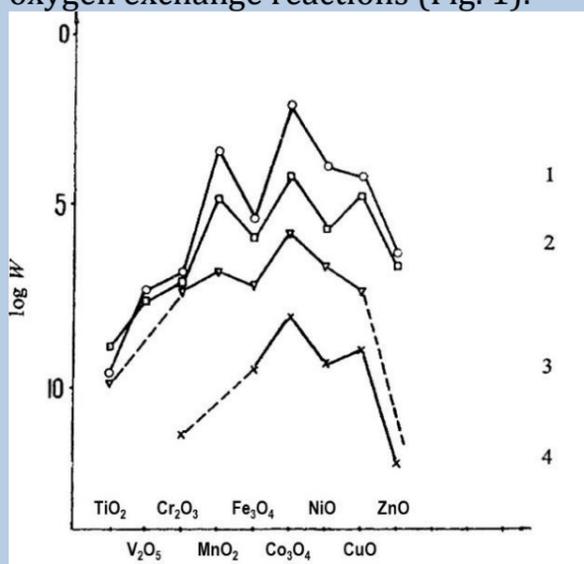
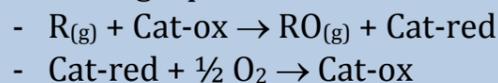


Fig. 1: Activity determined at 300°C over group IV transition metal oxide catalysts in (1) oxygen isotopic homomolecular exchange, (2) hydrogen oxidation, (3) methane oxidation and (4) N<sub>2</sub>O decomposition (Reproduced from G.K. Boreskov, *Discuss. Faraday Soc.*, 1996, 41, 263-276 with permission from The Royal Society of Chemistry).

These correlation demonstrated that the oxygen atoms from the surface and in some cases (V<sub>2</sub>O<sub>5</sub>, MoO<sub>3</sub>,...) from the bulk were involved in the catalytic

mechanism of the oxidation reaction. It confirmed the catalytic cycle reported in 1954 by Mars and Van Krevelen for the oxidation of aromatic compounds in which the substrate is oxidized with oxygen surface atoms in a first step and the partially reduced surface is re-oxidized with gaseous oxygen in a second step. This mechanism well known as Mars Van Krevelen mechanism is described in the two following equations:



Where R is the substrate and Cat-ox and Cat-red correspond to the oxidized and reduced oxide catalyst respectively.

In the isotopic exchange studies performed in University of Poitiers, two general reaction types have been investigated: (i) homomolecular exchange in which a mixture of labeled molecules is scrambled over a surface, and (ii) heterolytic exchange in which a labeled molecule is scrambled with the lattice atom of the catalyst isotopes. From the curves representing the evolution of the partial pressures of the different isotopomers of the exchanged molecules, information on the

exchange mechanism and on surface and bulk diffusion coefficient of active species can be obtained. Coupling this technique with spectroscopic characterization techniques such as FTIR or NMR gives complementary data on the nature of the active species or the active sites in the exchange process. The studies enables an optimization of the catalyst design developed in processes investigated in the No-Waste project like the pollution abatement or sustainable production of energy.



Picture of the No-Waste workshop 2016 organized in El Jadida, Morocco in which a keynote was given on isotopic exchange studies over catalysts involved in environmental processes for clean air.

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