EUROPEAN SYNCHROTRON RADIATION FACILITY
INSTALLATION EUROPEENNE DE RAYONNEMENT SYNCHROTRON

Experiment Report Form

	Experiment title: Stability of chlorinated RuO2(110) model catalyst in the Sumitomo Process	**Experiment number**: SI-1944
Beamline: ID03	**Date of experiment**: from: 23.9.2009 to:29.9.2009	**Date of report**: 22.2.2010
Shifts: 18	**Local contact(s)**: Dr. Olivier Balmes	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

Stefan Zweidinger* ***Physikalisch-Chemisches Institut, Justus Liebig University Giessen, Germany***; Dr. Jan Philipp Hofmann* ***Physikalisch-Chemisches Institut, Justus Liebig University Giessen, Germany***; Dr. Olivier Balmes* LOCAL CONTACT (IS03)

Prof. Dr. Edvin Lundgren* *Dept. of Synchrotron Radiation Research, University of Lund, Sweden*

Prof. Dr. Herbert Over* ***Physikalisch-Chemisches Institut, Justus Liebig University Giessen, Germany***

Report: submitted to Journal of Catalysis: Paper has been accepted on 16.2.2010.

In-situ Studies of the Oxidation of HCl over RuO_2 Model Catalysts: Stability and Reactivity

S. Zweidinger [a)], J. P. Hofmann [a)], O. Balmes [b)], E. Lundgren [c)], and H. Over[a)*]

Abstract

Structure-activity experiments were performed for the HCl oxidation reaction (Deacon-like process) over RuO_2 model catalysts – chlorinated RuO_2(110) and RuO_2(100) - applying in-situ surfaces x-ray diffraction (SXRD) combined with on-line mass spectrometry. The studied model catalysts turned out to be long-term

stable under reaction conditions with gas feed ratios p(HCl):p(O_2) ranging from 1:4 to 4:1 in the mbar pressure regime and temperatures as high as 685 K. Even pure HCl exposure in the mbar regime was not able to reduce RuO_2 below 600 K; above 650 K chemical reduction of the oxide sets in. Under strongly oxidizing reaction conditions the (surface) oxides grow slowly in thickness. On-line reactivity experiments of both types of model catalysts in a batch reactor yield a mean turn-over frequency (TOF) of 0.6 Cl_2 molecules per second and active site for the HCl oxidation at 650 K and initial partial pressures of p(HCl) = 2 mbar and p(O_2) = 0.5 mbar. The HCl-oxidation over RuO_2 is therefore considered to be structure insensitive.

Graphical Abstract:

Graphical Abstract

In-situ surface x-ray diffraction reveals that RuO_2-model catalysts are long-term stable for the HCl oxidation reaction by oxygen with a mean TOF of 0.6 Cl_2/s using a batch reactor.