



Universiteit
Leiden
The Netherlands

High-pressure STM studies of oxidation catalysis

Bobaru, Ş.C.

Citation

Bobaru, Ş. C. (2006, October 25). *High-pressure STM studies of oxidation catalysis*. Retrieved from <https://hdl.handle.net/1887/4952>

Version: Corrected Publisher's Version

License: [Licence agreement concerning inclusion of doctoral thesis in the Institutional Repository of the University of Leiden](#)

Downloaded from: <https://hdl.handle.net/1887/4952>

Note: To cite this publication please use the final published version (if applicable).

High-Pressure STM
Studies of Oxidation Catalysis

S. C. Bobaru

High-Pressure STM Studies of Oxidation Catalysis

PROEFSCHRIFT

TER VERKRIJGING VAN
DE GRAAD VAN DOCTOR AAN DE UNIVERSITEIT LEIDEN,
OP GEZAG VAN DE RECTOR MAGNIFICUS DR. D.D. BREIMER,
HOGLERAAR IN DE FACULTEIT DER WISKUNDE EN
NATUURWETENSCHAPPEN EN DIE DER GENEESKUNDE,
VOLGENS BESLUIT VAN HET COLLEGE VOOR PROMOTIES
TE VERDEDIGEN OP WOENSDAG 25 OKTOBER 2006
KLOKKE 16:15 UUR

DOOR

ȘTEFANIA CARMEN BOBARU

GEBOREN TE DRĂGOEȘTI (ROEMENIË)
IN 1975

Promotiecommissie

Promotor: Prof .dr. Joost Frenken

Referent: Prof. dr. B. E. Nieuwenhuys

Overige leden: Prof. dr. J. A. Moulijn (DelftChemTech-Technische
Universiteit Delft)

Dr. J. C. van den Heuvel (Universiteit van Amsterdam)

Prof. dr. M. T. M. Koper

Prof. dr. P. H. Kes

Prof. dr. J. Aarts

High-pressure STM studies of oxidation catalysis
Ștefania Carmen Bobaru

ISBN: 90-9021165-9

ISBN-10: 90-9021165-9

ISBN-13: 978-90-9021165-7

A digital version of this thesis can be downloaded from
<http://physics.leidenuniv.nl/sections/cm/ip>
<https://openaccess.leidenuniv.nl>

The work described in this thesis was performed at the Kamerlingh Onnes Laboratory of Leiden University. This work is part of the research programme of the Stichting voor Fundamenteel Onderzoek der Materie (FOM), which is financially supported by the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO).

“Remember to play after every storm”
Matthew J. T. Stepanek

*In the memory of my grandmothers:
Margareta and Constantina
To whom I owe being who I am*

Contents

1 Introduction	1
1.1 Heterogeneous catalysis	1
1.2 The three-way catalyst	2
1.3 Surface-science studies	2
1.4 Scanning Tunneling Microscopy and catalysis studies	4
1.4.1 Introduction	4
1.4.4 Our set-up	5
1.5 This thesis	8
1.6 References	10
2 Introduction to the theory concerning CO oxidation over platinum group metals	11
2.1 CO interaction with platinum group metals	11
2.2 CO oxidation over platinum group metals	12
2.3 Oscillatory CO oxidation over platinum group metals	15
2.4 Summary	17
2.5 References	18
3 CO oxidation over palladium surfaces	19
3.1 Motivation	19
3.2 Relation between the efficiency and the crystal structure of a palladium catalyst :a literature overview	19
3.3 Electronic and structural information about Pd	20
3.4 Experimental	21
3.5 Results and discussion	21
3.6 Summary for CO oxidation on Pd(100)	26
3.7 CO oxidation over high-Miller-index palladium surfaces	27
3.7.1 Motivation	27
3.7.2 Vicinal surfaces-an introduction	28
3.7.3 CO oxidation on Pd(1.1.17)	29
3.7.4 CO oxidation on Pd(553)	38
3.8 Conclusions	40
3.9 References	42
4 New insights into the oscillatory behaviour of CO oxidation over platinum group metals	45
4.1 Introduction	45
4.2 Traditional models for reaction oscillations	46
4.3 Comparison with experimental observations	49
4.4 The role of roughness	52
4.5 Conclusions	55
4.6 References	56

5 CO oxidation on Pt(111): overlayers, oxidation and reaction oscillations	59
5.1 Introduction	59
5.3 Previous work on Pt(111)	60
5.3.1 Existence and stability of surface platinum oxides-a literature survey	60
5.3.2 Interaction of Pt(111) with CO	61
5.4 Experimental	62
5.5 Results and discussion	62
5.5.1 CO adsorption on Pt(111) at ambient pressure	62
5.5.2 STM images combined with reaction kinetics	64
5.5.3 I-V Spectroscopy	71
5.5.4 Bistability and oscillations in the reaction kinetics	72
5.6 Conclusions	75
5.7 References	77
6 Oxidation of Pt(100)	79
6.1 The quasi-hexagonal reconstruction	79
6.2 Experimental	81
6.3 Results and discussion	82
6.3.1 Interaction of CO with Pt(100)	82
6.3.2 Interaction of O ₂ with Pt(100)	87
6.3.3 Pt(100) in a CO+O ₂ mixture, during CO oxidation	89
6.3.4 Bistability and hysteresis	98
6.4 Conclusions	101
6.5 References	102
A.I NO reduction by CO on Pt(100)	103
A.I.1 Introduction	103
A.I.2 Results and discussions	104
A.I.3 Conclusions	110
A.I.4 References	111
A.II Ethylene oxidation over Ag(111) and Pt(111)	113
A.II.1 General	113
A.II.2 partial oxidation of ethylene over Ag(111)	114
A.II.3 Total oxidation of ethylene on Pt(111)	118
A.II.4 Conclusions	120
A.II.5 References	122
Summary	123
Samenvatting	125

List of publication	127
Curriculum Vitae	129

