

# Carbon monoxide tolerant catalysts for fuel cells

PhD Thesis

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# Introduction

Exploiting renewable energy sources is one of today's most important tasks, but their availability varies both in space and time. Therefore, it is necessary to store the produced energy until further use. Hydrogen can be produced indirectly from any renewable energy source: first electricity is generated from primary energy sources and then it is converted to hydrogen by electrolysis of water. The hydrogen can then be stored and, when necessary, converted into electrical energy in fuel cells with high efficiency.

Particularly, there is great potential for Polymer Electrolyte Membrane (PEM) fuel cells. Considering their size, relatively low operating temperature and fast start-up, they are capable of powering different devices, from phones to transport devices, auxiliary power units and combined heat and power units. A huge market is therefore waiting for a breakthrough in production of affordable PEM fuel cells with long service life, which depends critically on progress in electrocatalyst development.

The PEM fuel cells operate with platinum catalyst, which is a major item of the high production costs. Accordingly, the main direction of research and development is to reduce the amount of platinum used (eventually to zero, by developing platinum-free electrodes), and to increase the stability and lifetime of catalysts and the electrodes made from them.

According to the literature, the stability of the electrodes can be improved primarily by increasing the stability of the catalyst support. Active carbon is an excellent catalyst support because of its highly advantageous properties, but in an acidic environment (due to the proton exchange membrane in the PEM fuel cells), especially if the electrode potential fluctuates as a result of rapid load changes (e.g. as source of energy in a vehicle), it tends to corrode. Therefore it is important to research materials that are more resistant to these stresses.

The carbon monoxide sensitivity of the catalyst is also an important problem on the anode side where the fuel is oxidized. At present, most of the hydrogen used as fuel is obtained from the reforming of hydrocarbons, thus it always contains carbon monoxide as impurity. Therefore, the development of anode side catalysts with increased carbon monoxide tolerance is an important task.

Members of the Renewable Energy Research Group in IMEC RCNS, HAS work on solving the problems described above have shown promising results in their previous works.<sup>1,2</sup> I joined this research during my PhD.

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<sup>1</sup> D. Gubán, I. Borbáth, Z. Pászti, I.E. Sajó, E. Drotár, M. Hegedűs, A. Tompos, *APPLIED CATALYSIS B-ENVIRONMENTAL* **174**: pp. 455-470. (2015)

<sup>2</sup> D. Gubán, Z. Pászti, I. Borbáth, I. Bakos, E. Drotár, I. Sajó, A. Tompos, *PERIODICA POLYTECHNICA-CHEMICAL ENGINEERING* **60**: (1) pp. 29-39. (2016)

# Objectives

The main purpose of my PhD work was to produce a catalyst with good carbon monoxide tolerance and good stability as an anode-side electrode in a hydrogen-fueled PEM fuel cell.

Based on the research group's previous results and literature, I have aimed to develop a method for the preparation of titanium-molybdenum mixed oxide-activated carbon composite materials as a support for platinum electrocatalysts.

Nowadays, there are still several theories in the literature to interpret the relationship between the carbon monoxide oxidation pre-peak appearing on  $\text{CO}_{\text{ads}}$ -stripping voltammograms and the carbon monoxide tolerance. Therefore, detailed investigation of the electrooxidation properties of carbon monoxide was found to be necessary in order to reveal possible explanations for the phenomena described in the literature and observed by me. A model catalyst was also developed to discover and understand these relationships, which could help to design a better catalyst.

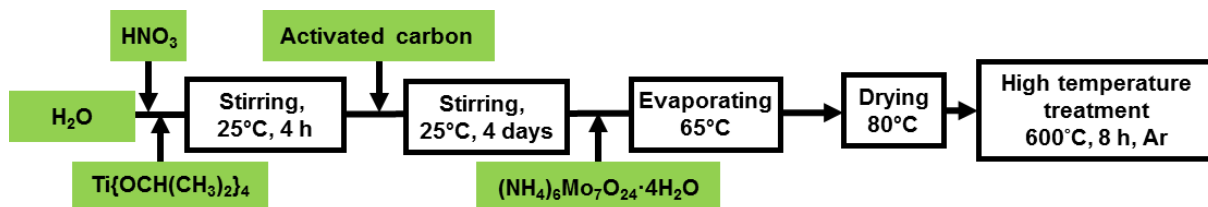
Another aim of my work was to investigate the durability of the catalyst, especially with regard to its effect on the catalytic activity.

Last but not least, my aim was to compare the catalytic properties of my catalyst to those of commercially available carbon supported platinum, which is the most commonly used catalyst in hydrogen-fueled PEM fuel cells and carbon supported platinum ruthenium which is referred in the literature as the state-of-the-art carbon monoxide tolerant catalyst. Therefore, it was an important task to test the catalysts in a fuel cell test apparatus.

# Applied methods

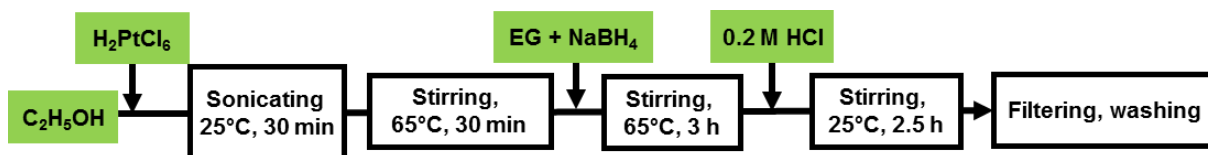
## *Preparation of the support materials and the related Pt electrocatalysts*

$\text{Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  mixed-oxide – carbon composite materials were prepared by a modified room temperature multi-step sol-gel based method which was followed by a high-temperature heat-treatment. The main features of the optimized preparation procedure of  $\text{Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  composite support materials are briefly summarized in Fig. 1.



**Fig. 1** Flow chart for preparing  $\text{Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  composite materials by using multi-step synthesis route.

Ti<sub>(1-x)</sub>Mo<sub>x</sub>O<sub>2</sub>-C composite materials were loaded with Pt via a modified NaBH<sub>4</sub> assisted ethylene-glycol reduction-precipitation method (see Fig. 2).



**Fig. 2** Flow chart for Pt loading of the Ti<sub>(1-x)</sub>Mo<sub>x</sub>O<sub>2</sub>-C composite supports.

### *Physicochemical characterization of the support materials and the related Pt catalysts*

Composite materials obtained at the end of room temperature preparation were investigated by temperature programmed reduction (TPR) and X-ray photoelectron spectroscopy (XPS) with *in-situ* heat treatments in order to determine the optimal temperature of the heat treatment for the preparation.

The structure of composite materials (the formation of rutile or anatase TiO<sub>2</sub> phase and the degree of Mo incorporation) before and after the high-temperature heat treatment were investigated the by X- ray diffraction (XRD) technique.

After the platinum loading the catalysts were examined by transmission electron microscopy (TEM) in order to specify the particle size distribution of platinum nanoparticles. The Pt electrocatalysts were also investigated by energy-dispersive X-ray spectroscopy (EDX) and XPS to define the bulk and the surface compositions, respectively.

### *Electrochemical characterization and single fuel cell measurements*

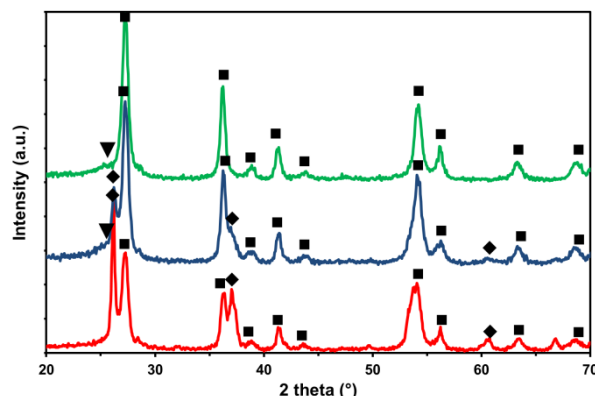
The electrocatalytic properties (carbon monoxide and hydrogen oxidation reactions) of the catalysts were measured by various electrochemical techniques (cyclic voltammetry, CO<sub>ads</sub>-stripping voltammetry) and fuel cell test measurements, compared to commercially available carbon supported platinum (40 and 20 wt.% Pt/C (Quintech)) and the state-of-the-art CO tolerant carbon supported platinum ruthenium (PtRu/C, Pt= 20 wt.%, Ru= 10 wt.% (Quintech)) catalysts as references.

Stability is an important requirement for fuel cell catalysts. Therefore, the effect of the number of polarization cycles on the physical-chemical and catalytic properties of Pt/Ti<sub>(1-x)</sub>Mo<sub>x</sub>O<sub>2</sub>-C and the commercial PtRu/C electrocatalysts were studied by prolonged cyclic polarization combined with (i) CO<sub>ads</sub>-stripping voltammetry, (ii) hydrogen oxidation reaction on rotating disk electrode, as well as (iii) TEM and XPS techniques.

To further clarify the relationship between the molybdenum oxidation state and the CO tolerant behavior of the Pt/Ti<sub>(1-x)</sub>Mo<sub>x</sub>O<sub>2</sub>-C electrocatalysts, the electrochemical and XPS experiments were performed with molybdenum-modified platinum model surfaces.

# Theses

**T1. Method for the preparation of titanium-molybdenum mixed-oxide-activated carbon composite materials was developed. It was shown that in case of the Ti/Mo=80/20 atomic composition, the molybdenum is fully incorporated into the rutile-TiO<sub>2</sub> structure. In the case of samples containing more added molybdenum the presence of non-incorporated, segregated molybdenum oxide was demonstrated. [1-2]**



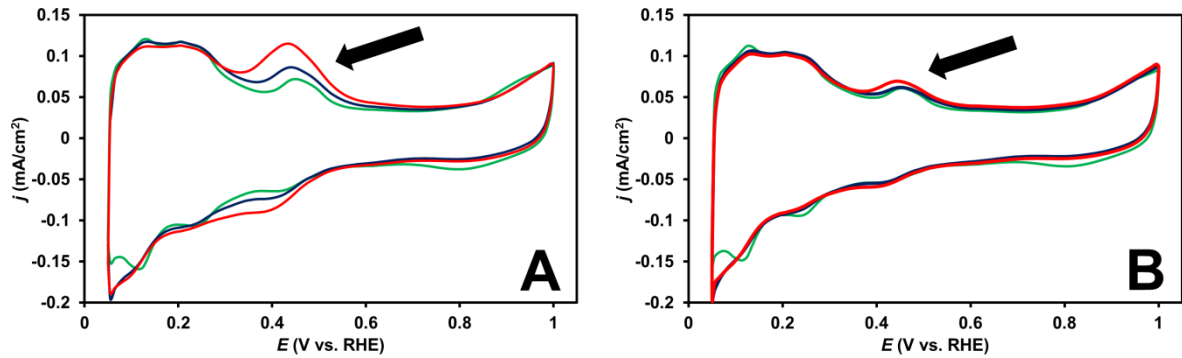
**Fig. 3** XRD patterns of  $\text{Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  samples after high-temperature treatment. Green line:  $\text{Ti}_{0.8}\text{Mo}_{0.2}\text{O}_2\text{-C}$ ; blue line:  $\text{Ti}_{0.7}\text{Mo}_{0.3}\text{O}_2\text{-C}$ ; red line:  $\text{Ti}_{0.6}\text{Mo}_{0.4}\text{O}_2\text{-C}$ . ■ - rutile; ▼ - anatase; ● -  $\text{MoO}_3$ ; ◆ -  $\text{MoO}_2$ .

**Table 1** Structural properties of the  $\text{Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  (C= 25 wt.%) composite materials after the high-temperature treatment (600 °C, 8 h, Ar) determined by XRD analysis

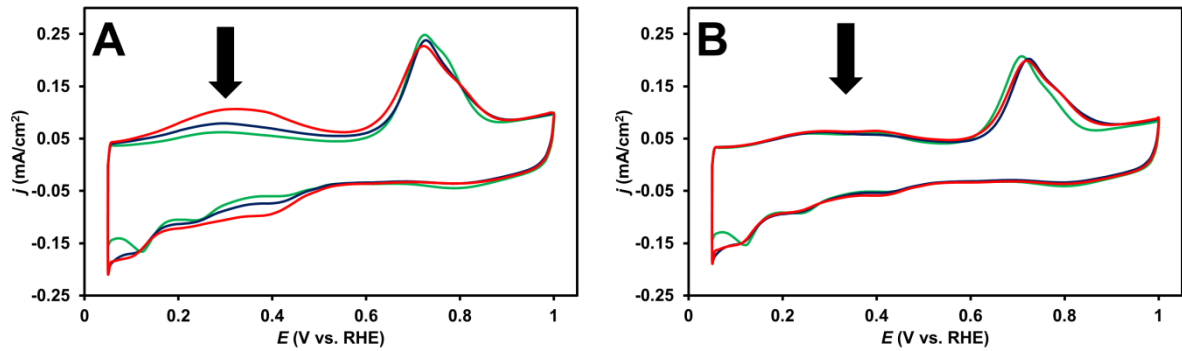
Samples	Phase (wt.%)			Lattice parameters (Å)	Mo substitution (at.%)
	Rutile	Anatase	$\text{MoO}_3/\text{MoO}_2$		
Reference $\text{TiO}_2$	100	0	-	$a= 4.593, c= 2.959$	-
$\text{Ti}_{0.8}\text{Mo}_{0.2}\text{O}_2\text{-C}$	90	10	0/0	$a= 4.640, c= 2.935$	21
$\text{Ti}_{0.7}\text{Mo}_{0.3}\text{O}_2\text{-C}$	86	10	0/4	$a= 4.645, c= 2.932$	25
$\text{Ti}_{0.6}\text{Mo}_{0.4}\text{O}_2\text{-C}$	80	5	0/15	$a= 4.655, c= 2.928$	30

The phase composition of the  $\text{Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  ( $x= 0.2\text{-}0.4$ ) composite materials was investigated by XRD measurements (Fig. 3). According to the change of the unit cell parameters compared to the pure rutile- $\text{TiO}_2$  lattice (see Table 1) the estimated degree of Mo incorporation was about 21-30 %. The presence of the diffraction peaks characteristic to the  $\text{MoO}_2$  phase (Fig. 3) indicates that a certain fraction of Mo remains outside of the titania lattice. It was demonstrated that the amount of non-incorporated  $\text{MoO}_2$  phase increases with increasing nominal molybdenum content and incorporation of molybdenum under the used conditions has a maximum.

**T2.** It was shown that the stability of the samples with different molybdenum content depends on the degree of Mo incorporation and does not depend on the amount of added molybdenum. [2]



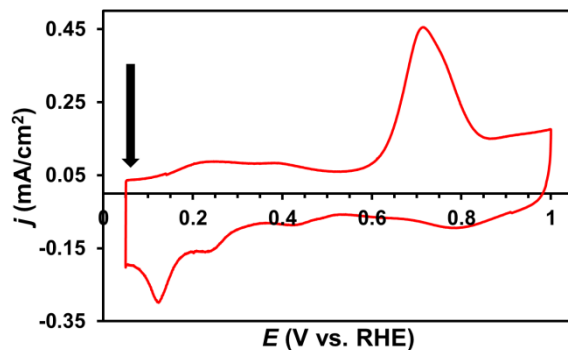
**Fig. 4** Cyclic voltammograms of 20 wt.%  $\text{Pt/Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  catalysts obtained before (A) and after the “pre-leaching” test for 30 cycles (B). Green line:  $\text{Pt/Ti}_{0.8}\text{Mo}_{0.2}\text{O}_2\text{-C}$ ; blue line:  $\text{Pt/Ti}_{0.7}\text{Mo}_{0.3}\text{O}_2\text{-C}$ ; red line:  $\text{Pt/Ti}_{0.6}\text{Mo}_{0.4}\text{O}_2\text{-C}$ . Recorded in 0.5 M  $\text{H}_2\text{SO}_4$  at 10 mV/s,  $T = 25^\circ\text{C}$ .



**Fig. 5** Argon purged  $\text{CO}_{\text{ads}}$  stripping voltammograms of 20 wt.%  $\text{Pt/Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  catalysts before (A) and after (B) the “pre-leaching” procedure. Green line:  $\text{Pt/Ti}_{0.8}\text{Mo}_{0.2}\text{O}_2\text{-C}$ ; blue line:  $\text{Pt/Ti}_{0.7}\text{Mo}_{0.3}\text{O}_2\text{-C}$ ; red line:  $\text{Pt/Ti}_{0.6}\text{Mo}_{0.4}\text{O}_2\text{-C}$ . Recorded in 0.5 M  $\text{H}_2\text{SO}_4$  at 10 mV/s,  $T = 25^\circ\text{C}$ . The “pre-leaching” was carried out for 30 cycles at 100 mV/s.

Figures 4 and 5 show that the degree of changes of the surface of the 20 wt.%  $\text{Pt/Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  catalysts observed due to prolonged cyclic polarization (30 cycles, 50-1000 mV, 100 mV/s) correlates with the content of non-incorporated molybdenum of the samples.

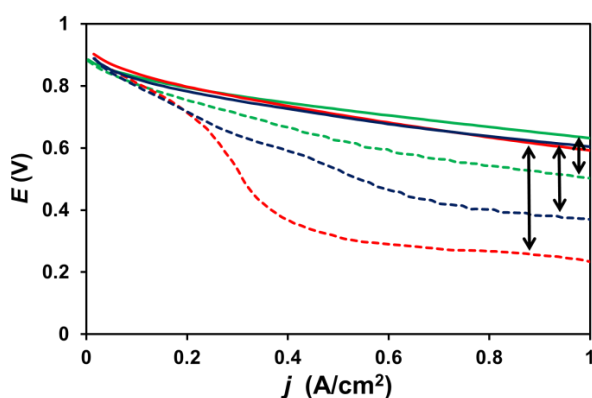
**T3.** It was demonstrated that the electrooxidation of CO adsorbed on the surface of  $\text{Pt/Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  catalysts starts at an exceptionally low potential value (ca. 50 mV). This can be explained by the formation of active sites consisted of Pt and reducible ionic Mo surface species of mixed oxide. [1]



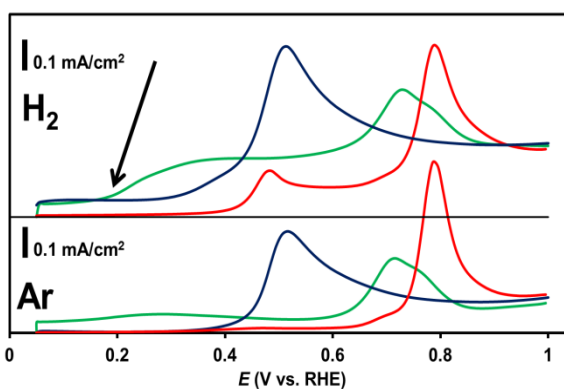
**Fig. 6** Argon purged  $\text{CO}_{\text{ads}}$  stripping voltammograms of 40 wt.%  $\text{Pt/Ti}_{0.7}\text{Mo}_{0.3}\text{O}_2\text{-C}$  catalyst. Obtained in 0.5 M  $\text{H}_2\text{SO}_4$  at 10 mV/s.

As shown in Fig. 6 the increase of CO electrooxidation current was observed on the composite supported catalyst at exceptionally low potential values (ca. 50 mV).

**T4.** It was proved by electrochemical and single fuel cell test measurements that the  $\text{Pt/Ti}_{(1-x)}\text{Mo}_x\text{O}_2\text{-C}$  catalysts have more advantageous properties in low-potential carbon monoxide oxidation reaction compared to the state-of-the-art CO-tolerant commercially available PtRu/C catalyst. The  $\text{Ti}_{0.8}\text{Mo}_{0.2}\text{O}_2\text{-C}$  composite supported Pt electrocatalysts showed significantly less degradation compared to the PtRu/C. [1-2, 5]

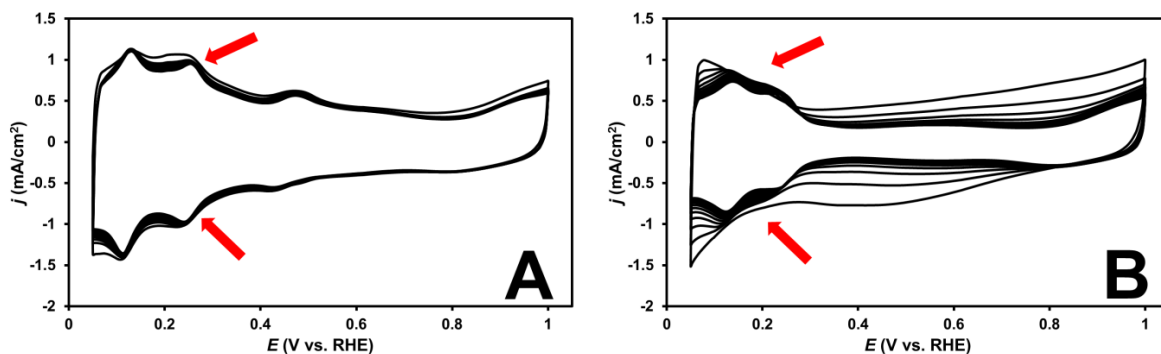


**Fig. 7** Cell potential vs. current density plots for single cells with pure  $\text{H}_2$  (solid lines) and 100 ppm  $\text{CO/H}_2$  fuel on the 40 wt.%  $\text{Pt/Ti}_{0.7}\text{Mo}_{0.3}\text{O}_2\text{-C}$  (dashed green line), PtRu/C (dashed blue line), Pt/C (dashed red line).



**Fig. 8**  $\text{CO}_{\text{ads}}$ -stripping voltammograms obtained after Ar-purging or  $\text{H}_2$  purging in the anodic scan on the 20 wt.%  $\text{Pt/Ti}_{0.8}\text{Mo}_{0.2}\text{O}_2\text{-C}$  (green line), PtRu/C (blue line) és Pt/C (red line) catalysts. Recorded in 0.5 M  $\text{H}_2\text{SO}_4$  solution between 50 and 1000 mV potential limits with 10 mV/s sweep rate,  $T=25^\circ\text{C}$ .

According to the voltage loss ( $\Delta E$ ) at 1 A/cm<sup>2</sup> current density measured during experiments done in a single fuel-cell test device using as a fuel pure and 100 ppm CO containing H<sub>2</sub> (Fig. 7) the order of CO tolerance (reversed order of activity decrease due to CO poisoning) of the catalysts was as follows: 40 wt.% Pt/Ti<sub>0.7</sub>Mo<sub>0.3</sub>O<sub>2</sub>-C ( $\Delta E$ = 0.13 V) > PtRu/C ( $\Delta E$ = 0.23 V) > Pt/C ( $\Delta E$ = 0.37 V).



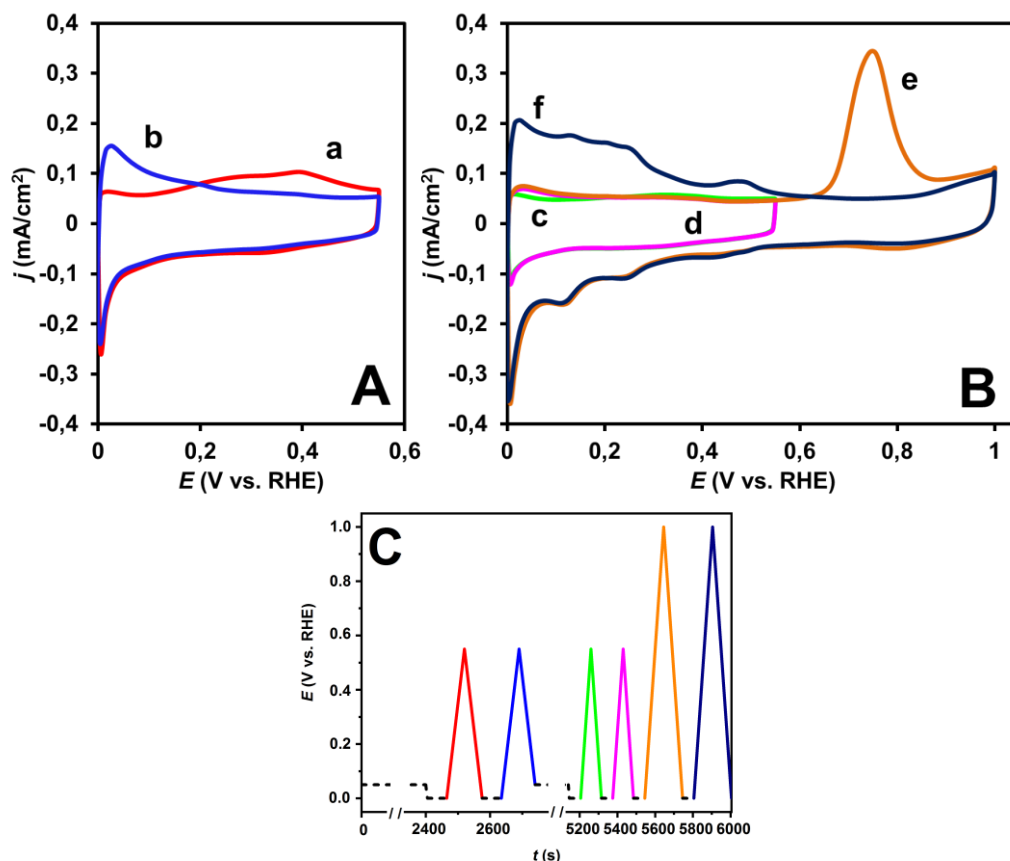
**Fig. 9** Cyclic voltammograms of the Pt/Ti<sub>0.8</sub>Mo<sub>0.2</sub>O<sub>2</sub>-C (A) and the PtRu/C (B) electrocatalysts; 500 cycles recorded after the 10 cycles of conditioning by displaying every 50<sup>th</sup> cycles (the arrows indicate the direction of changes observed upon increase of the number of the cycles). Recorded in 0.5 M H<sub>2</sub>SO<sub>4</sub> at 100 mV/s, T= 25 °C.

An increase of the anodic current at around 200 mV observed during H<sub>2</sub>-purged CO<sub>ads</sub> stripping measurement on the 20 wt.% Pt/Ti<sub>0.8</sub>Mo<sub>0.2</sub>O<sub>2</sub>-C catalyst (see Fig. 8) means that the onset potential of hydrogen oxidation is shifted toward less positive potentials in comparison with Pt/C and PtRu/C (around 400 and 300 mV, respectively); this result also demonstrates the good carbon monoxide tolerance of the composite supported catalysts.

Fig. 9.A shows that during 500 polarization cycles of the stability test on the 20 wt.% Pt/Ti<sub>0.8</sub>Mo<sub>0.2</sub>O<sub>2</sub>-C catalyst only a relatively small change in the shape of the voltammogram was observed. In contrast, as a result of ruthenium dissolution, after 500 cycles a pronounced change of the voltammogram shape of the PtRu/C catalyst was observed (Fig. 9.B): it begins to resemble the shape characteristic for a typical Pt/C electrocatalyst; degradation of Ru-containing catalyst results in loss of CO-tolerance.



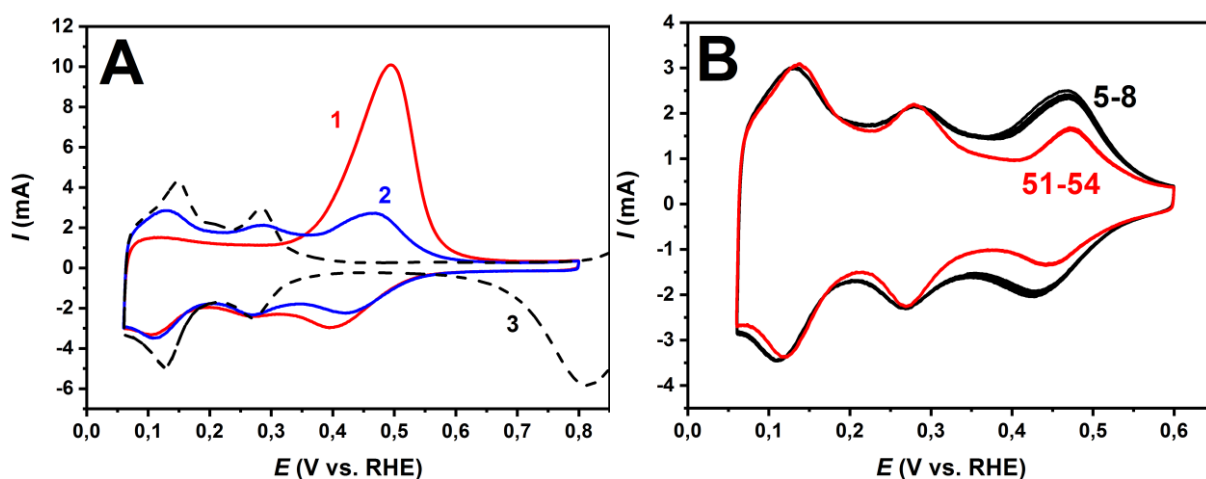
**T5.** It was shown that the so-called CO oxidation pre-peak can be assigned to at least two oxidation processes: to the oxidation of the carbon monoxide weakly adsorbed on the platinum located in atomic closeness to Mo surface species and to the oxidation of molybdenum surface species. Due to the oxidation of molybdenum (polarization to a potential of more positive than 400 mV), the ability of the catalyst to oxidize the carbon monoxide at low potential is eliminated, and it can be restored only after the removal of the carbon monoxide layer covering the entire platinum surface. [3]



**Fig. 10** The CO<sub>ads</sub> stripping voltammograms of the Pt/Ti<sub>0.6</sub>Mo<sub>0.4</sub>O<sub>2</sub>-C catalyst obtained after Ar purging using different potential limits (a, b); (c) shows the potential program (the colour of the insert lines indicating the potential cycling conditions applied during the measurements corresponds to the colour of voltammograms shown on the figure). Obtained in 0.5 M H<sub>2</sub>SO<sub>4</sub> at 10 mV/s, T = 25 °C.

As emerges from curve **a** in Fig. 10.A removal of weakly adsorbed CO results in hydrogen adsorption and oxidation on the released platinum surface (Fig. 10.A **b** curve). During the successive polarizations according to the potential program (Fig. 10.C), after the second CO saturation the activity of the surface could not be restored (Fig. 10.B, curve **c** and **d**) due to another process occurring in addition to the CO oxidation, which presumably is connected to the change of molybdenum oxidation state.

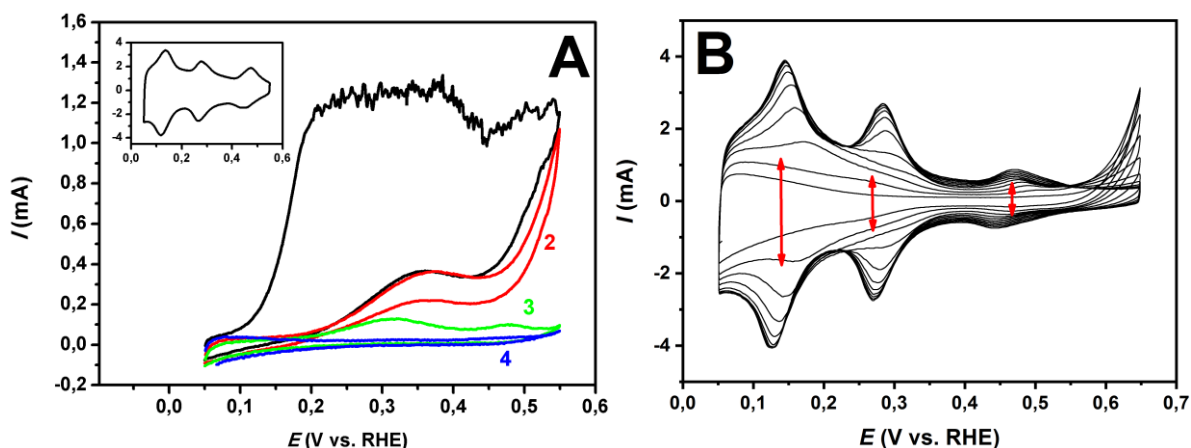
**T6.** Upon investigation of molybdenum modified platinum model surfaces, it was demonstrated that approx. 20-25 % of the initial molybdenum monolayer deposited on the platinum electrode is irreversibly bound to the surface even after oxidation to (VI) (by polarization to a potential of more positive than 550 mV). This deposited Mo partial monolayer is enough to significantly increase the carbon monoxide tolerance of the Pt surface. [4]



**Fig. 11** (A) Cyclic voltammograms of a Mo-Pt/Pt electrode after Mo-deposition at 50 mV for 5 min; CVs were measured in pure Mo-free 0.5 M  $\text{H}_2\text{SO}_4$ . Curve 1: 1st cycle, curve 2: 3rd cycle, curve 3 (dashed line): CV of the bare Pt/Pt electrode in 0.5 M  $\text{H}_2\text{SO}_4$ . (B) Continuation of the cyclic polarization of the Mo-Pt/Pt electrode in 0.5 M  $\text{H}_2\text{SO}_4$ . Numbers of the cycles are indicated.

As emerges from the cyclic voltammograms presented in Fig. 11.A, even after oxidation into the +6 state a certain part (20-25 %) of the initial Mo monolayer irreversibly deposited on the Pt/Pt electrode at 50 mV remains stable on the surface even after prolonged cyclic polarization (Fig. 11.B). This irreversibly adsorbed Mo surface species can be reduced back into the +4 state even after polarization up to 1000 mV.

**T7.** It was shown that only Pt model surface containing Mo (IV) is active in low potential CO oxidation. The irreversibly bound molybdenum oxidized to the (VI) state at a potential more positive than 550 mV can be re-reduced and the activity of the Pt-Mo catalytic surfaces can be restored. However, the reduction requires clean, CO-free Pt surface. [4]



**Fig. 12** (A) Cyclic voltammograms of Mo-Pt/Pt electrode (Mo coverage is about 25%) with low sweep rate (0.5 mV/s) in CO saturated and CO-purged 0.5 M H<sub>2</sub>SO<sub>4</sub>. Numbers indicate the number of cycles. Insert: CV of the same electrode in Ar-purged H<sub>2</sub>SO<sub>4</sub> with 10 mV/s sweep rate. (B) Cyclic voltammograms of the CO-covered Mo-Pt/Pt electrode in pure Ar-purged 0.5 M H<sub>2</sub>SO<sub>4</sub>. Red coloured arrows indicate the direction of changes.

Figure 12.A shows that the irreversibly deposited Mo partial monolayer (20-25 %) is enough to change significantly the CO poisoning properties of the Pt surface. However, during the subsequent cyclic polarization the CO tolerance of the Mo-Pt/Pt electrode was totally lost after the third sweep of the cyclic polarization from 50 mV to 550 mV (see curve 4 in Fig. 12.A). Since the upper potential limit 550 mV is higher than the potential needed for the Mo(IV)→Mo(VI) transition, this observation suggests that only Pt modified by Mo(IV) species is active in the low-potential CO oxidation reaction. This behaviour is quite similar to that observed on the 20 wt.% Pt/Ti<sub>0.6</sub>Mo<sub>0.4</sub>O<sub>2</sub>-C catalyst. However, Mo(VI)-oxide remaining on the surface can be reactivated by reduction of molybdenum into the +4 oxidation state and thus the catalyst activity can be restored, but this reduction requires clean, CO-free Pt surface (Fig. 12.B).

# Publications

MTMT identifier: 10040794

## PUBLICATIONS RELATED TO THE THESIS

1. D. Gubán, A. Tompos, I. Bakos, Á. Vass, Z. Pászti, E. Gy. Szabó, I. E. Sajó, I. Borbáth: Preparation of CO-tolerant anode electrocatalysts for polymer electrolyte membrane fuel cells. *INTERNATIONAL JOURNAL OF HYDROGEN ENERGY* **42**:(19) pp. 13741-13753. (2017) **IF: 4.229**
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3. Á. Vass, I. Borbáth, I. Bakos, Z. Pászti, I. E. Sajó, A. Tompos: Novel Pt Electrocatalysts: Multifunctional Composite Supports for Enhanced Corrosion Resistance and Improved CO Tolerance. *TOPICS IN CATALYSIS* **61**: (12-13) pp. 1300-1312. (2018) **IF: 2.439**
4. I. Bakos, I. Borbáth, Á. Vass, Z. Pászti, A. Tompos: Design and investigation of molybdenum modified platinum surfaces for modeling of CO tolerant electrocatalysts. *TOPICS IN CATALYSIS* **61**: (14) pp. 1385–1395. (2018) **IF: 2.439**
5. Á. Vass, I. Borbáth, I. Bakos, Z. Pászti, Gy. Sáfrán, A. Tompos: Stability issues of CO tolerant Pt-based electrocatalysts for polymer electrolyte membrane fuel cells: comparison of Pt/Ti<sub>0.8</sub>Mo<sub>0.2</sub>O<sub>2</sub>-C with PtRu/C. *REACTION KINETICS MECHANISMS AND CATALYSIS* **126**: (2) pp. 679–699. (2019). **IF(2017/2018): 1.515**

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## LECTURES RELATED TO THE THESIS

1. Á. Vass, I. Borbáth, I. Bakos, E. Gy. Szabó, A. Tompos: Preparation and characterization of Ti<sub>(1-x)</sub>Mo<sub>x</sub>O<sub>2</sub>-C/Pt electrocatalysts for PEM fuel cell. The 13<sup>th</sup> Pannonian International Symposium on Catalysis, Siófok, 19-23 September 2016
2. Vass Á., Tompos A., Bakos I., Pászti Z., Szabó E. Gy., Sajó I. E., Borbáth I.: CO-toleráns elektrokatalizátorok előállítása és vizsgálata. XXXIX. Kémiai Előadói Napok, Szeged, 17-19 October 2016
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4. Á. Vass, I. Borbáth, I. Bakos, Z. Pászti, P. Németh, I. E. Sajó, A. Tompos: CO tolerant behavior of Ti-Mo mixed oxide-carbon composite supported Pt electrocatalyst. 13<sup>th</sup> European Congress on Catalysis- A Bridge to the future (EUROPACAT 2017) Florence, Italy, 27-31 August 2017 – poster
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9. Vass Á., Tompos A.: Szén-monoxid-tűrő katalizátorok tüzelőanyag-elemekhez. Elektrokémiai Munkabizottsági ülés, Budapest, 19 September 2018
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11. Vass Á.: Szén-monoxid-tűrő katalizátorok tüzelőanyag-elemekhez. Katalízis Munkabizottsági ülés, Szeged, 11 December 2018

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12. Vass Á., Berki P., Hernádi K.: MWCNT/WO<sub>3</sub> kompozit anyagok előállítása és vizsgálata. XXXVI. Kémiai Előadói Napok, Szeged, 28-30 October 2013
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