Palladium Alloy Catalysts Synthesized by Sacrificial Support Method for the Electrooxidation of Ethylene Glycol in Alkaline Environment

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Direct alcohol fuel cells offer very high energy densities. They therefore are most advantageously used for applications requiring long up-times and in remote areas, where weight and volume savings compared to primary batteries are highest. Unfortunately this requires also that the fuel is transported to the place of usage. Today already direct methanol fuel cells are used for this purpose. Though the transport of methanol in certified fuel cell cartridges is possible, the logistic for larger deployments using such cartridges is still challenging. Bulk transport of the fuel would be preferred, is however restricted mostly because of the high flammability of methanol.

Ethylene glycol offers practical the same energy density as methanol and can be transported with much less restrictions. Furthermore, the breaking of the C-C bond in ethylene glycol is easier than in ethanol [1]. Using commercially Pd based catalyst from Acta CO_2 current efficiencies of up to 60% were found in single cell testing [2]. However, reaction kinetics are still sluggish and palladium though less expensive than platinum might be still too expensive.

The sacrificial support method (SSM) developed at UNM can be used for preparation of highly active and durable electroctalysts for anode and cathode side of MEA [3-7].

In this study we investigated the properties of palladium base metal alloys in particular of Pd-Cu catalyst for the ethylene glycol oxidation reaction. The modified SSM was used for preparation of unsupported Pd-Cu materials.

Figure 1 shows morphology of PdCu catalysts with well-developed 3-D porous structure. The surface area of prepared material was found as ~50 m^2 g^{-1}.

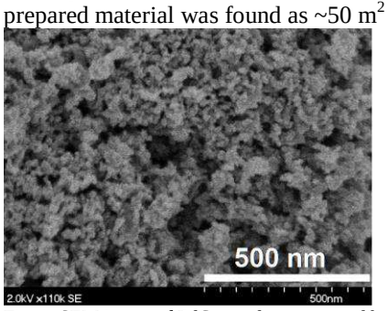

Fig. 1. SEM image of PdCu catalysts prepared by SSM

Samples of the UNM and Acta Hypermec® catalysts were analyzed at the Fraunhofer ICT using a flow-through DEMS cell [8].

Cyclic voltammetric and chronoamperometric measurements were performed with sprayed electrodes of UNM or Acta catalyst or sputtered palladium, respectivel, in 1 mM ethylene glycol, 0.1 M KOH solution at ambient temperature. During these measurements mass spectrometer signals for m/z = 15, 22 and 44 were monitored to monitor the evolution of CO_2 (m/z = 22 and 44),, acetaldehyde (m/z = 15 and 44) or other volatile organic compounds (m/z 15 only). In order to be able to quantify the CO_2 release CO_2 stripping measurements were performed to calibrate the CO_2 capture efficiency.

Under potentiodynamic conditions the PdCu catalyst achieved an integral CO_2 current efficiency of 86% for the positive half sweep of the CV (cf. fig. 2)

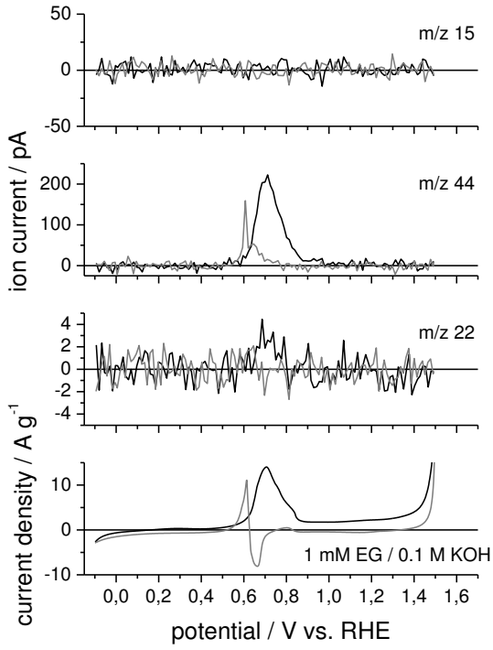

Fig 2: Oxidation of 1 mM EG / 0.1 M KOH in CV. Scan rate 10 mV s-1; 3rd cycle and corresponding signals for m/z 15, 44 and 22 are displayed

Under potentiostatic conditions the CO_2 current efficiency of the PdCU catalyst was determined with 36% which is significantly higher than for the other palladium catalyst.

Electrode	CO_2 current efficiency @ 0.7 V vs. RHE
UNM PdCu	36 %
Acta Hypermec	ca. 10%
Sputtered Pd	0%

Further investigations are ongoing including fuel cell tests.

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