

23 September 1996

PHYSICS LETTERS A

Physics Letters A 221 (1996) 141-143

On the behavior of the cathodically polarized Pd/D system: a response to Vigier's comments

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Received 23 May 1996; accepted for publication 24 May 1996 Communicated by P.R. Holland

Abstract

Electrodes prepared by Pd/D codeposition exhibit highly expanded surfaces which achieve high degrees of D/Pd loading within seconds. In this communication, morphology of the Pd electrode, the structure of the interphase, and selected thermal effects are discussed.

In response to Professor Vigier's comments, we assemble in this Letter other observations of the behavior of the Pd/D system when under cathodic polarization. This material is provided with the hope that such data may contribute to unraveling the mysteries of the Fleischmann-Pons effect announced on 23 March 1989. Here, we address: (1) morphology of the Pd electrode, (2) structure of the interphase, and (3) selected thermal effects. Only electrodes prepared by the codeposition technique, i.e., by palladium electrodeposited in the presence of evolving deuterium, are considered [1].

(1) Electrode morphology. Electrodes prepared by codeposition exhibit highly expanded surfaces consisting of small spherical nodules, Fig. 1a. A high degree of deuterium loading, with the atomic ratio D/Pd > 1, is obtained within seconds [2]. This be-

havior appears to be similar to that observed by Celani et al. (cited in Ref. [3]).

(2) Structure of the interphase. The metal side of the electrode/electrolyte interphase comprises, at least, two layers of vastly different deuterium content [4]. The presence of D_2^+ species in the interphase, at moderate cathodic overpotentials, has been suggested [4], supporting the tight orbit model. Moreover, the interphase is an active participant in the rate of charging, the intensity of emanating X-rays [5] as well as other manifestations, e.g., tritium production and its distribution between the electrolyte and vapor phases [6].

(3) Thermal effects. Observed thermal effects cover a wide range: from electrolyte cooling to the melting of the Pd electrode [7]. Electrolyte cooling in the initial stages of codeposition has been observed [1]. With further charging, the electrode temperature (measured by a thermocouple) exceeds that of the electrolyte by a few degrees. However, when viewed

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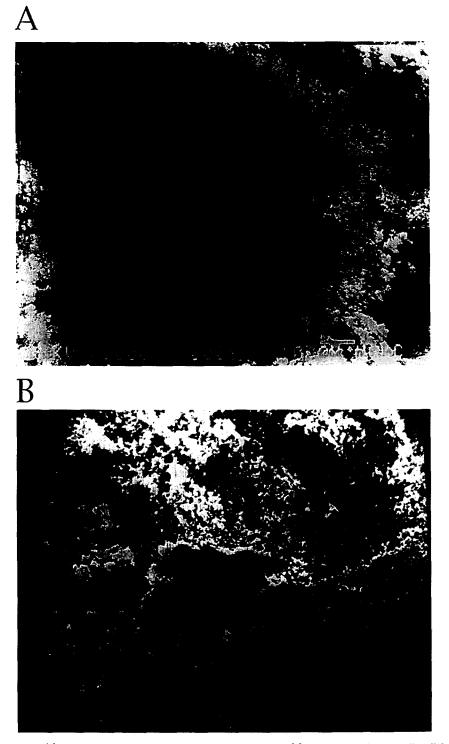


Fig. 1. SEM photographs of (a) a Pd electrode freshly prepared by co-deposition and (b) Pd film deposited on cell wall following melting of the Pd electrode during D_2O electrolysis.

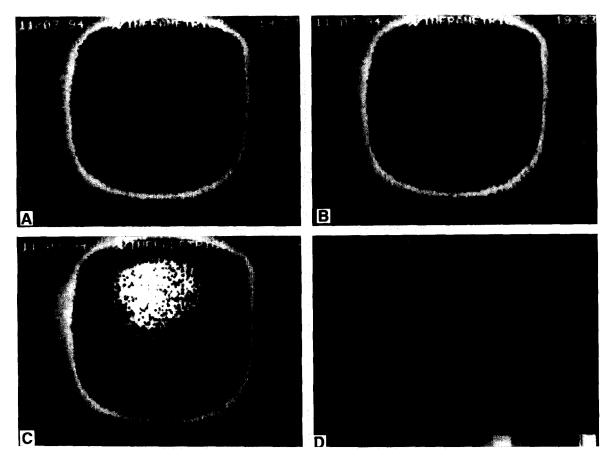


Fig. 2. Temperature distribution of the Pd electrode, prepared by co-deposition on a Ni screen, during cathodic polarization as recorded by an infrared camera. (a) Appearance of "hot spots"; (b), (c) merging of hot spots to form larger temperature zones; (d) graphical representation of temperature distribution.

with an infrared camera, the chaotic appearance/disappearance of hot spots is observed, followed by merging in larger islands that often exhibit oscillations in size, Figs. 2a and 2b. The most extreme temperature rise, exceeding the melting point of Pd, was observed once. In this case, molten Pd was deposited on the wall of the electrolytic cell in the form of a thin film, Fig. 1b.

In selecting the topics for inquiries into the Fleischmann-Pons effect, we kept in mind the closing sentence in Born's 1943 address to the Durham Philosophical Society [8]: "My advice to those who wish to learn the art of scientific prophecy is not to rely on abstract reason, but to decipher the secret language of Nature from Nature's documents, the facts of experience."

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