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Photoelectrochemistry on a planar, interdigitated electrochemical cell

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ABSTRACT

The construction of a planar electrochemical cell with two interdigitated electrodes, with a finger spacing of 25–100 μm and 200–50 pairs of fingers and gaps, respectively, is described. The working electrode consists of n-type semiconducting titanium dioxide (anatase). Under UV light, photocurrents are produced. No reduction in the photocurrent due to *iR* drop was observed, even in electrolytes of low ionic strength. This makes the device an interesting candidate for photoelectrocatalytic purification of drinking water. An example is shown in the degradation of the azo-dye AO7 (acid orange 7). *k*(photoelectrocatalysis) on a device with 50 finger pairs was 3.29×10^{-6}/s and the corresponding kinetic parameter, $p = kFV/i_{\text{photo}}$ (rate constant normalized to unit volume and photocurrent), was 404 M^{-1}.

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1. Introduction

The removal of organic material of biological and industrial origin (bacteria, hormones, endocrine disruptors, textile dyes, dioxines, etc.) from waste- and drinking water is of enormous importance nowadays. There is a great need for new technologies serving this purpose. One way could be photocatalysis, involving semiconducting oxides under illumination. In a heterogeneous photocatalytic process, a solid semiconductor catalyst such as titanium dioxide or other transition metal oxides irradiated with near UV light generate strongly oxidizing species (valence band holes, ·OH radicals) which can destroy organic species present in the aqueous phase in contact with the surface of the catalyst. However, there are limitations to the efficiency of the process that are partly due to the semiconductor and partly due to various recombination processes and the technical outline of the reactor in which the reaction is carried out.

Early work [1] showed how photocatalysis in semiconductor particulate suspensions can be understood on the basis of photoelectrochemical analysis of macroscopic electrodes. From this analysis it became also clear that the main obstacle to efficient use of absorbed photons is the recombination of photogenerated charge carriers, and that this can be suppressed by applying electrical bias to the semiconducting photocatalyst which must be immobilized on an electrically conducting substrate. This has also the advantage of making separation of the catalyst from the reaction mixture (the liquid phase) superfluous, once the reaction is over. Recent work on organic impurity degradation using TiO_2 [2–6] and WO_3 thin film electrodes [7–9] validated these ideas.

In an electrochemical cell, *iR* drop is one of the factors limiting high current throughput at moderate bias. Especially in electrochemical treatment of additives (pollutants) in drinking water, having low ionic strength, where addition of supporting electrolyte is prohibited for obvious reasons, ways must be devised for limiting *iR* drop. (Typical tap water conductivities of 528 μS/cm were observed in our laboratory.) One way is to use a parallel plate reactor with two opposite electrodes and a small space between them where the electrolyte is passed through. For practical reasons (if the electrodes are large) this distance may not fall below 1 mm, if energy investment for pumping is to be minimized. Such a reactor with 10 cm × 10 cm plates and a 1 mm gap, with a conductivity cell constant, κ, of 0.001 cm^{-1} was recently demonstrated [2], and had an electrolyte resistance for tap water (500 μS/cm) of 2 Ω, leading to an *iR* drop of only 40 mV for current densities of around 3×10^{-4} A/cm^2 as observed for photocurrents on TiO_2 produced by solar irradiation. However, as the pressure build-up is considerable in a module consisting of many such cells, another way is suggested in the present work: both electrodes are positioned in the same plane so that no flow restrictions apply. However, when large currents are to be passed through a planar device, it must be taken into account that the conductivity cell constant of such an arrangement is considerable higher than that of an equivalent arrangement where the electrodes face each other. In order to ensure low *iR* drop between the electrodes, they are arranged in an interdigitated way. Electrochemical cells with various planar interdigitated arrangements have been used for redox sensing and other electroanalytical

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applications [10–13]. Such structures can lead to very low cell constants if a large number of thin fingers and small spacing are used [14].

2. Experimental

The schematics of a typical planar device with an interdigitated part of 0.5 cm × 1.5 cm are shown in Fig. 1. Fingers extending from the left side towards the centre consist of photoactive TiO_2 deposited on platinum, whereas the finger family starting on the right side is the Pt counter electrode. Electric contacts were taken at the bottom to which the Pt collector areas are extended. The devices were fabricated by standard lithographic techniques using contact masks. The masks were prepared in the following way: the pattern was drawn as vector graphics on computer file using the program "Adobe Illustrator". The patterns were then transferred either onto transparent plastic sheets using a laser printer, or directly onto photographic film. As these foils were subsequentially used as contact masks in the lithographic processes, high accuracy combined with mechanical stability for repeated use was sought, giving preference to patterns on photographic film. An example is given in the optical microscope pictures in Fig. 2 with details of the pair of masks used for the "100–25" device, *i.e.*, a device with 100 μm wide Pt and TiO_2 fingers and a 25 μm gap between the finger families: a mask used for Pt deposition (a) and a mask used during TiO_2 deposition (b). The optical pictures show the excellent sharpness of the lithographic masks.

Masks with different finger widths (100, 50, 25 μm) and interfinger distances (100, 50, 25 μm) were prepared, with a finger length of 5 mm. Devices with patterns deposited over a length of 15 mm and equal finger spacing of 25–100 μm led to 200–50 pairs of fingers, respectively, and the same number of pairs of gaps (insulating parts made of SiO_2 between the finger families), covering a total operating area of 0.75 cm^2, of which 0.1875 cm^2 (one fourth) was rendered photoactive due to coverage with titanium dioxide.

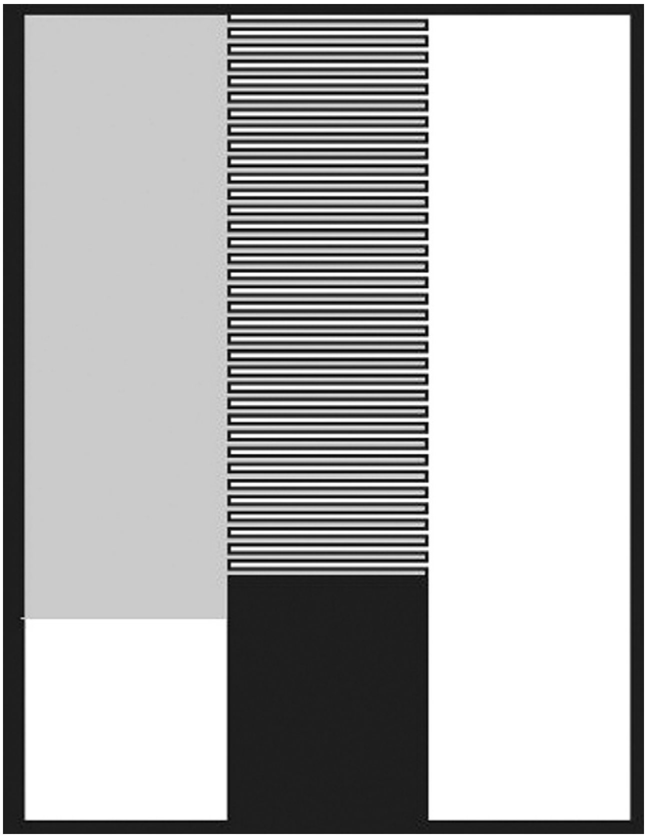

Fig. 1. Schematics of an interdigitated device (in scale: interdigitated area 1.5 cm × 0.5 cm). Black part – insulating area (SiO_2), light grey part – titanium dioxide, white part – Pt. Bottom left and right – area for applying external contacts.

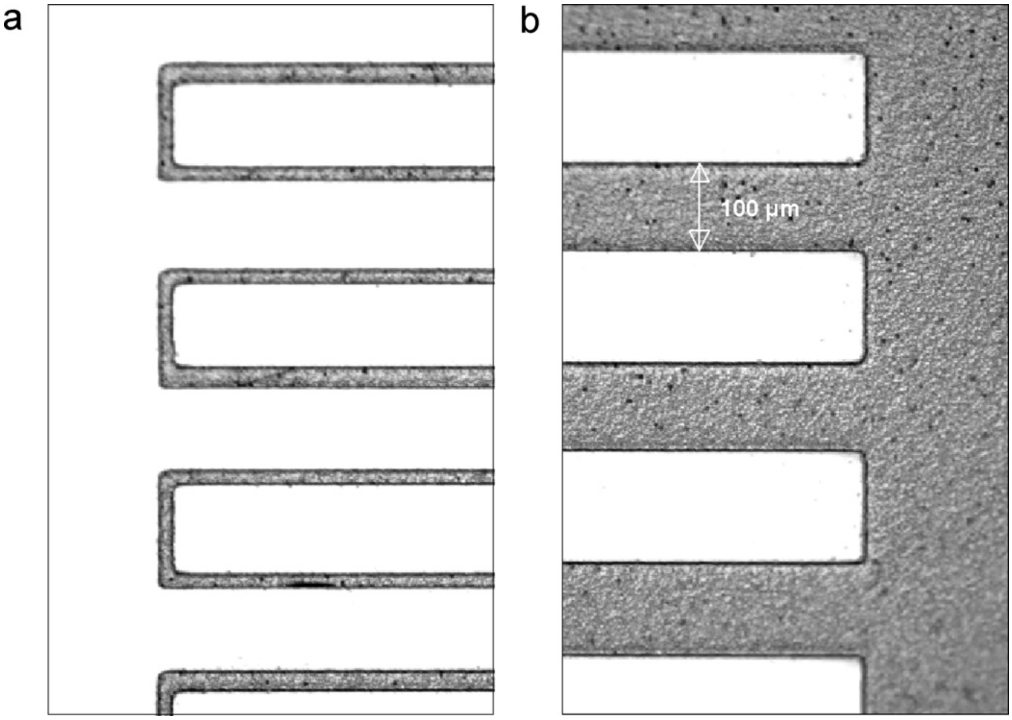

Fig. 2. Inspection of the emulsion side of developed photographic film containing the masks. Details (optical microscope) of a pair of masks used for depositing a "100–25" device (100 μm wide Pt and TiO_2 fingers and 25 μm gap between the finger families). Left – mask used for Pt deposition, right – mask used for revealing TiO_2.

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