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## Chemical switches and logic gates based on surface modified semiconductors

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### Abstract

Photoelectrochemical properties of multicomponent photoelectrodes based on titanium dioxide and cadmium sulfide powders modified with hexacyanoferrate complexes have been examined. Photocurrent responses were recorded as functions of applied potential and photon energy. Surprisingly, the photocurrent can be switched between positive and negative values as a result of potential or photon energy changes. This new effect called PhotoElectrochemical Photocurrent Switching (PEPS) opens a possibility of new chemical switches and logic gates construction. Boolean logic analysis and a tentative mechanism of the device are discussed. *To cite this article: K. Szaciłowski, W. Macyk, C. R. Chimie 9 (2006).*

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### Résumé

Les propriétés photoélectrochimiques des photoélectrodes à composants multiples à base de poudres d'oxyde de titane et de sulfure de cadmium, modifiées avec des complexes hexacyanoferrate, ont été examinées. Les réponses photovoltaïques ont été observées en fonction du potentiel ou de l'énergie photonique appliquée. De manière surprenante, la réponse photovoltaïque peut passer d'une valeur positive à une valeur négative par suite de modifications du potentiel ou de l'énergie photonique. Ce nouvel effet, appelé *PhotoElectrochemical Photocurrent Switching* (PEPS), ouvre la voie à de nouveaux interrupteurs chimiques ainsi qu'à la construction de portes logiques. On fait appel à l'analyse logique de Boole et on propose un mécanisme possible.

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## 1. Introduction

Simple inorganic pigments: *titanium white*, *cadmium yellow* and *Prussian blue* are known for centuries. The oldest in the family, Prussian blue (PB), was first described by M. Diesbach in 1704 [1,2]. Initially it was used as a blue pigment, but recently has found applications as a chemosensor [3], ion exchanger [4], in electrochromic displays [5], as molecular magnet [6], and in some semiconductor devices [4]. Two other materials studied here: titanium dioxide and cadmium sulfide are well known wide bandgap semiconductors, widely used as photocatalysts [7,8]. A special attention is paid to titanium dioxide as the material potentially suitable for solar energy conversion [9,10].

Photochemical properties of TiO<sub>2</sub> surface modified with cyanometallates have been studied extensively [11–16]. Vrachnou et al. [11,12] found that the surface complexes show characteristic metal to metal charge transfer transitions (MMCT, Fe<sup>II</sup>→Ti<sup>IV</sup>) in visible range and excitation within new bands results in photocurrent generation. Other important contributions concerned composite materials consisting of titanium dioxide particles embedded in polymeric cyanoferrate matrices [17–20]. These materials showed some interesting properties—the photocurrent direction depends on the photoelectrode potential. This behavior was explained in terms of electrostatic barriers within the hexacyanoferrate matrix. Surprisingly, contrary to the reports from the Grätzel laboratory [11,12] the interactions between TiO<sub>2</sub> particles and cyanoferrate matrix do not result in an MMCT (Fe<sup>II</sup>→Ti<sup>IV</sup>) absorption band and hence no sensitization of the semiconductor is observed. According to our knowledge there is no report on switching of photocurrent direction caused by change of the incident photon energy in similar systems. Similar phenomenon was reported for very different systems composed of helical peptides (attached to gold electrode) carrying various chromophores [21]. Systems described in this paper are much simpler, do not require organic electron donor neither acceptor and show photocurrents ca. four orders of magnitude higher.

Recently much attention is focused on chemical systems capable of performing logic operations [22–44], especially in solid state [25,45,46]. The research in the field of molecular logic is stimulated by natural limits of classical, silicon-based electronics [47]. This contribution describes new phenomena in systems contain-

ing semiconductor particles and hexacyanoferrate moieties, both as isolated ions and as polymeric films. The photoelectrochemical properties of the studied systems are interpreted in terms of chemical switches.

## 2. Results and discussion

Modification of semiconductor materials with cyanoferrate complexes changes their optical properties: impregnation of TiO<sub>2</sub> with [Fe(CN)<sub>6</sub>]<sup>4-</sup> ions results in the MMCT (Fe<sup>II</sup>→Ti<sup>IV</sup>) transition at 450 nm (Fig. 1a). This indicates formation of a stable “surface complex” containing the Ti<sup>IV</sup>-N≡C-Fe<sup>II</sup>(CN)<sub>5</sub> moieties. Deposition of the Prussian blue layer on semiconductor surfaces results in materials exhibiting strong MMCT

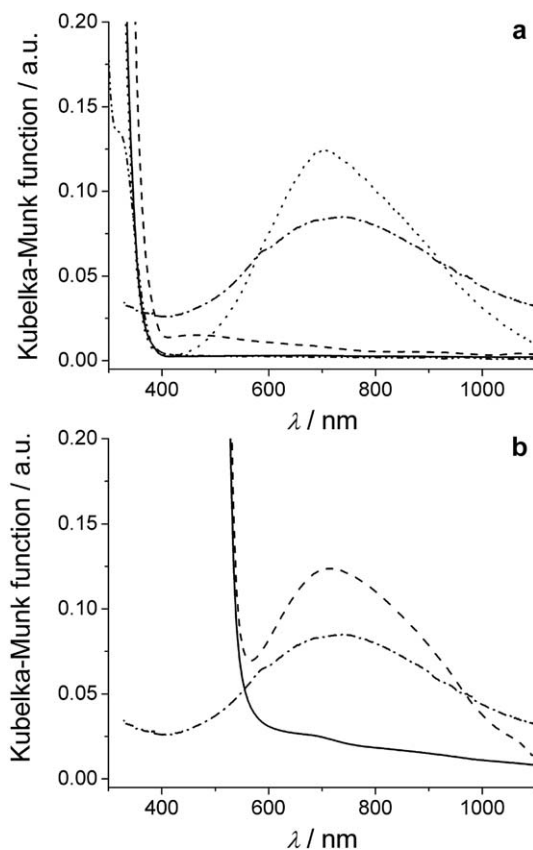


Fig. 1. Diffuse reflectance spectra of modified TiO<sub>2</sub> materials and modifying compounds (a): TiO<sub>2</sub> (—), [Fe(CN)<sub>6</sub>]<sup>4-</sup>@TiO<sub>2</sub> (---), PB@TiO<sub>2</sub> (.....), PB@SiO<sub>2</sub> (-.-.-), and aqueous solution of K<sub>4</sub>[Fe(CN)<sub>6</sub>] (-.-.-, absorbance scale). Diffuse reflectance spectra of modified CdS materials and modifying compounds (b): CdS (—), PB/CdS (---), and PB@SiO<sub>2</sub> (-.-.-).

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