

Author's Accepted Manuscript

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PII: S0956-5663(18)30161-1
DOI: <https://doi.org/10.1016/j.bios.2018.02.060>
Reference: BIOS10322

To appear in: *Biosensors and Bioelectronic*

Received date: 13 November 2017
Revised date: 11 February 2018
Accepted date: 26 February 2018

Cite this article as: Feriel Boussema, Andrew J. Gross, Fatma Hmida, Brahim Ayed, Hatem Majdoub, Serge Cosnier, Abderrazak Maaref and Michael Holzinger, Dawson-type polyoxometalate nanoclusters confined in a carbon nanotube matrix as efficient redox mediators for enzymatic glucose biofuel cell anodes and glucose biosensors, *Biosensors and Bioelectronic*, <https://doi.org/10.1016/j.bios.2018.02.060>

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Dawson-type polyoxometalate nanoclusters confined in a carbon nanotube matrix as efficient redox mediators for enzymatic glucose biofuel cell anodes and glucose biosensors

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Abstract: Two new inorganic-organic hybrid materials based on heteropolyoxometalates (POMs) : $(C_4H_{10}N)_6[P_2Mo_{18}O_{62}] \cdot 4H_2O$ (P_2Mo_{18}) and $(C_6H_8NO)_4[H_2P_2W_{18}O_{62}] \cdot 6H_2O$ (P_2W_{18}) are reported as mediators for electron transfer between FAD-dependent glucose dehydrogenase (FAD-GDH) and a multiwalled carbon nanotube (MWCNT) matrix for glucose biofuel cell and biosensor applications. These polyoxometalates were chosen due to their promising redox behavior in a potential range for mediated electron transfer with the glucose oxidizing enzyme, FAD-GDH. P_2Mo_{18} and P_2W_{18} were immobilized on 1-pyrenemethylamine (PMA) functionalized MWCNT deposits. After immobilization of FAD-GDH, the P_2W_{18} -modified MWCNT electrode demonstrated mediated electron transfer and provided a catalytic current density of 0.34 mAcm^{-2} at 0.2 V vs SCE with an open circuit potential (OCP) of -0.08 V vs SCE. A 10-fold increase in catalytic current to 4.7 mAcm^{-2} at 0.2 V vs SCE and a slightly lower OCP of -0.10 V vs SCE was observed for an equivalent electrode modified with P_2Mo_{18} . The apparent superiority of P_2Mo_{18} is related, at least in part, to its improved incorporation in the MWCNT matrix compared to P_2W_{18} . Both POM-modified bioanodes showed exceptional stabilities with 45% of their initial performances remaining after 15 days. The mediated electron transfer capacities of the POMs were also evaluated in a glucose sensor setup and showed very satisfying performances for glucose detection, including a sensitivity of $0.198 \text{ mA mol}^{-1} \text{ cm}^{-2}$, a satisfying linear range between 1 mmol L^{-1} and 20 mmol L^{-1} , and good reproducibility for the P_2Mo_{18} electrode.

Keywords: Electron transfer mediator, Dawson type polyoxometalates, glucose biofuel cells, glucose dehydrogenase, multi walled carbon nanotubes

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