Colloids and Surfaces A 552 (2018) 124–129

Contents lists available at ScienceDirect

Colloids and Surfaces A

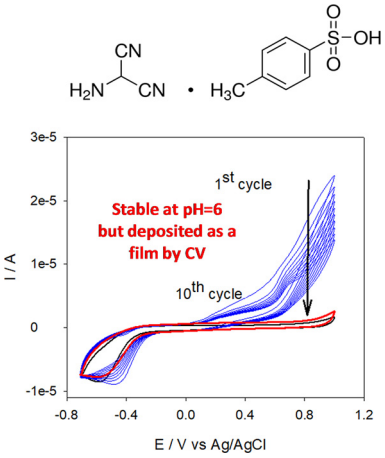
journal homepage: www.elsevier.com/locate/colsurfa

Electrochemical deposition of aminomalonitrile based films

Vincent Ball[a,b,*], Rou Jun Toh[c,d], Nicolas H. Voelcker[c,d], H. Thissen[c], Richard A. Evans[c]

[a] Université de Strasbourg, Faculté de Chirurgie Dentaire, 8 rue Sainte Elisabeth, 67000, Strasbourg, France
[b] Institut National de la Santé et de la Recherche Médicale, Unité de Recherche 1121, 11 rue Humann, 67085, Strasbourg Cedex, France
[c] CSIRO Manufacturing, Research Way, Clayton, 3168, Victoria, Australia
[d] Monash University, Monash Institute of Pharmaceutical Sciences, 381 Royal Parade, Parkville, 3052, Victoria, Australia

GRAPHICAL ABSTRACT



ARTICLE INFO

Keywords:
Aminomalonitrile
Cyclic voltammetry
Electrochemical deposition

ABSTRACT

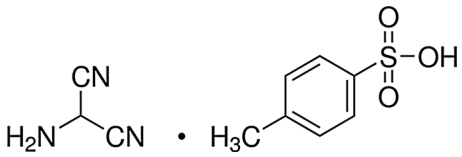
Versatile coatings based on the polymerization of aminomalonitrile (AMN) start to attract attention in the field of surface science owing to their biocompatibility and also in relation to the use of a precursor molecule being considered as a fundamental building block in prebiotic chemistry. As other emerging versatile coating methods, the polymerization of AMN produces not only coatings on almost all kinds of known materials but also the deposition of a useless precipitate in solution when AMN is dissolved at pH = 8.5. To circonvent this drawback, at least on conductive substrates, it is shown that AMN coatings can be obtained in a scan rate dependant manner on amorphous carbon electrodes by means of cyclic voltammetry at pH = 6.0 in conditions where AMN is stable in solution. The composition of the electrodeposited AMN coatings is compared with that of the solution processed coatings by means of X-ray photoelectron spectroscopy.

1. Introduction

In the last ten years huge research efforts have been devoted to design thin films able to adhere on a large repertoire of materials, after the pioneering article by Lee et al. [1] describing the versatile deposition of “polydopamine” films on all kinds of known materials from an

* Corresponding author at: Université de Strasbourg, Faculté de Chirurgie Dentaire, 8 rue Sainte Elisabeth, 67000, Strasbourg, France.
E-mail address: vball@unistra.fr (V. Ball).

https://doi.org/10.1016/j.colsurfa.2018.05.028
Received 21 February 2018; Received in revised form 10 May 2018; Accepted 10 May 2018
Available online 12 May 2018
0927-7757/ © 2018 Elsevier B.V. All rights reserved.



Scheme 1. Structure of aminomalonitrile p-toluene sulfonate used in this study.

aerated dopamine solution at pH = 8.5. The idea behind this concept was the fact that dopamine contains simultaneously a catechol and an amine group, similarly to the adjacent L-DOPA and L-Lysine amino acid residues in the mussel adhesion foot proteins [2]. These chemical groups allow strong metal coordination with metals under acidic conditions, where the catechol groups are prevalent. Under basic conditions, as in sea water, the catechol groups are oxidized into quinones which allow to establish covalent bonds with nucleophiles.

The repertoire of versatile surface chemistry was further extended with polyphenol based coatings [3–5] which are also based on an empiric observation, namely that tea cups and red wine glasses are covered with a coloured film after the consumption of tea and wine respectively. Finally, it has been demonstrated that it is possible to deposit coatings from aminomalonitrile (2-aminopropanedinitrile) (Scheme 1) solutions in basic solutions on the surface of almost all known materials [6,7].

In all cases, from dopamine, polyphenols or aminomalononitrile (AMN) based solutions, the deposition of films on interfaces is accompanied by the formation of insoluble precipitates in solution, inducing a huge loss of active molecules. In the case of dopamine [8–10] and polyphenols, mostly for tannic acid and pyrocatechol [11], it has been demonstrated that electrodeposition on the surface of conductive materials induces the formation of a conformal coating without formation of modified monomers in the bulk of the solution. It is the aim of the present research to demonstrate that AMN based coatings can also be deposited on the surface of amorphous carbon electrodes under potential scanning with respect to a reference electrode in a potential window compatible with the use of water as a solvent. It will also be demonstrated by means of electrochemical impedance spectroscopy, that the properties of the obtained films depend markedly on the potential scan rate for a given number of potential sweep cycles. The electrodeposited AMN based coatings could find various applications as the protection against corrosion and as substrates for the immobilisation of nanoparticles or biomolecules on conductive substrates

2. Materials and methods

All chemicals used in the present study were purchased and used without further purification. The aqueous solutions were prepared from Milli Q water (Millipore, Molsheim, France) with a resistivity of 18.2 MΩ cm.

AMN in the form of p-toluenesulfonate salt (Sigma, ref. 221147, see Scheme 1) was dissolved at a concentration of 10 mg.mL^{-1} in 50 mM sodium phosphate buffer (Sodium dihydrogen phosphate dihydrate, Sigma ref. 71,500) with pH adjusted to 6.0 with concentrated sodium hydroxide. The pH of the solutions was controlled with a Hi221 pH meter (Hanna instruments) fitted with a calibrated glass electrode. The calibration was performed with standard buffers at pH 4.0 and 7.0.

Potassium hexacyanoferrate (II) trihydrate ($K_4Fe(CN)_6$, $3H_2O$, Sigma, ref. 3987) was dissolved at 1 mM in the sodium phosphate buffer at pH 6.0 as an electrochemical probe.

For the aim of CV experiments, the AMN solutions were put in contact with amorphous carbon electrodes (ref. CHI 104, CHInstruments, Houston) which were polished with a SiC disk and successively on 1 and 0.1 µm Al_20_3 slurries (Escil, Villeurbane, France) before sonication in ultra-pure water (2 sonication steps lasting 2 min each). The cleanliness of the electrode was checked before the deposition of the AMN based films by performing a CV scan between -0.7 and + 1.0 V versus the Ag/AgCl reference electrode (ref. CHI 111, CHInstruments) in the presence of buffer containing 1 mM $K_4Fe(CN)_6$. The current was measured between the amorphous carbon working electrode and a Pt wire (ref. CHI 115, CHInstruments). For the electrochemical deposition experiments the number of potential sweep cycles as well as the potential scan rate were used as the experimental parameters. All experiments were performed at (20 ± 2)°C

Electrochemical impedance spectroscopy (EIS) was performed with the same device at constant direct potential of +0.283 V vs the Ag/AgCl reference electrode, corresponding to the oxidation potential peak of $K_4Fe(CN)_6$ on the pristine carbon electrode. This potential was superimposed with a sinusoidal potential 5 mV in amplitude with a frequency varying between 10^5 and 10^{-2} Hz. 12 measurements were made in each frequency decade. The obtained Nyquist plots, the opposite of the imaginary part of the impedance (-Z") plotted as a function of the real part of the impedance (Z'), were analyzed using the "eisanalyzer" software, freely available on the internet.

UV-visible spectra were performed on AMN solutions at 10 mg.mL^{-1} in the presence of 50 mM sodium phosphate buffer at pH = 8.5 (conditions used to deposit AMN based films [6,7]) and at pH = 6.0 in order to check the absence of change in the later case over time scales typical of those used in the electrodeposition experiments. To that aim the spectra of the AMN solutions, were measured after 20 fold dilution in sodium phosphate buffer (at the required pH) between 200 and 750 nm using the sodium phosphate buffer as the reference with a double beam Safas mc^2 spectrophotometer (Safas, Monaco).

X-ray photoelectron spectroscopy (XPS) analysis was carried out using an AXIS Ultra DLD spectrometer (Kratos Analytical, Manchester, UK) with a monochromated Al Kα source at a power of 168 W (14 kV x 12 mA), a hemispherical analyser operating in the fixed analyser transmission mode, and the standard aperture (analysis area: 0.3 mm x 0.7 mm). Survey spectra were acquired at a pass energy of 160 eV. Data processing was performed using CasaXPS processing software version 2.3.15 (Casa Software Ltd., Teignmouth, UK). All elements present were identified from survey spectra. The atomic concentrations of the detected elements were calculated using integral peak intensities and the sensitivity factors supplied by the manufacturer.

For XPS measurements, a glassy carbon plate (ref. 37022, Alfa Aesar) was used as the working electrode. Before the deposition of AMN based films, the glassy carbon surface was cleaned by polishing with 0.05 µm Al_2O_3 slurry. The electrode was then rinsed thoroughly with ultra-pure water and sonicated in ultra-pure water and acetonitrile (each sonication step lasting 5 min). Electrochemical deposition experiments were performed with cyclic voltammetry at a scan rate of 50 mV·s^{-1}. A Pt wire and Ag/AgCl electrode were employed as the counter and reference electrode respectively. Moreover, two chemical deposition experiments were used for control; glassy carbon plates were immersed in 10 mg·mL^{-1} AMN solutions at: (1) pH 8.5 for 24 *h*, as previously reported [6,7]; and (2) at pH 6.0 for 12 min, which is the approximate time required for 10 cycles of cyclic voltammetry at a scan rate of 50 mV·s^{-1}. All samples were rinsed thoroughly with Milli-Q water and dried using a stream of high purity nitrogen before being subjected to XPS characterization.

3. Results and discussions

A set of conditions were first defined in order to get a stable AMN solution, ie pH conditions in which AMN does not polymerise or self-assemble because this would inevitably lead to the spontaneaous formation of films at interfaces [6,7] and to the appearance of brown coloured species and finally precipitates in solution. Indeed, the aim of the present investigation is to produce films via electrochemical methods from stable AMN solutions as has been done from dopamine solutions [8–10]. When AMN is dissolved at pH = 6 in the presence of 50 mM sodium phosphate no observable change in the UV–vis spectrum

Download English Version:

https://daneshyari.com/en/article/6977309

Download Persian Version:

https://daneshyari.com/article/6977309

Daneshyari.com