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Abstract

Towards Next-Generation Glucose Sensors: Reactively Sputtered Nanostructured Nickel Nitrides for CMOS Integration †

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Abstract: A facile and clean-room compatible approach to Ni nitride (Ni_xN_y) synthesis is proposed, based on a reactive sputtering deposition technique. Ni_xN_y thin films were deposited at different N_2 partial pressures, after which their electrocatalytic properties towards glucose oxidation were investigated. Relative to the bare Ni, Ni_xN_y thin films sputtered at low N_2 partial pressures exhibited a decreased linear range (0–5 mM) and sensitivity. Contrarily, Ni_xN_y thin films sputtered at high N_2 partial pressures displayed an increase in sensitivity of ≈30% in the same linear range (0–10 mM) as for Ni. The Ni_xN_y films showed a clear morphological change from a flat thin film (Ni) to a faceted nanostructure whose characteristic dimensions decreased with increasing N_2.

Keywords: glucose; non-enzymatic sensing; nickel nitride

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1. Introduction

Detecting glucose levels accurately and sensitively in biofluids is vital for the proper clinical management of diabetes, personal health, and fitness [1]. Despite remarkable efforts towards the development of minimally invasive, miniaturized and reliable sensors for glucose monitoring, most devices continue to rely on an enzyme to mediate the electrooxidation of glucose [1]. Limited by poorly stable as well as pH- and oxygen-sensitive enzymes, this particular field is in dire need of novel non-enzymatic solutions [1]. Recently, transition metal nitrides have attracted increasing interest as catalysts for glucose electrooxidation. Of these, nickel nitrates are particularly intriguing due to their resistance to chloride poisoning [2], biocompatibility, electrical conductivity, thermal and chemical stability [2]. The most common methods of synthesis are via ammonolysis in a NH_3 atmosphere at high temperatures (>300 °C for crystal phases) or via a solvothermal method involving toxic species [3]. In contrast, an RF magnetron sputtering system does not require any harmful chemicals and can be performed at room temperature [3], thus being more compatible for the fabrication of integrated sensors. Moreover, by adjusting the sputtering parameters of thin films with different crystal phases and significantly different catalytic activities can be obtained. Although promising, this method is yet to be applied in the development of non-enzymatic glucose sensors in view of an easy integration with electronics.

2. Materials and Methods

All electrochemical measurements were taken by means of an *Autolab* potentiostat (PGSTAT204, Metrohm, Kontich, Belgium) in a 3-electrode configuration. The reference electrode was a standard Ag/AgCl/**KCl_{sat}** electrode (Sigma Aldrich, St. Louis, MO, USA), the counter electrode was a Pt coil (Sigma Aldrich). The working electrode was a 7 mm × 7 mm Ni_xN_y on borosilicate glass electrode connected a tantalum clip (Redoxme).

3. Discussion

The electrical resistivity of the Ni film was 100 $\mu\Omega\cdot cm^{-1}$, in accordance with other works [4]. Both Ni_xN_y thin films displayed similar resistivities of roughly 300 $\mu\Omega\cdot cm^{-1}$. Our electrochemical study showed that the electrical measurements were not a predictor for the glucose electrocatalytic activity.

Morphological characterization: Unlike for the Ni_xN_y thin films, the pure Ni samples grew with a Volmer Weber growth, leading to partially connected grains. Cubic-like structures fully covered the surface of the Ni_xN_y (20%) and Ni_xN_y (50%) samples, with particularly smaller nanostructures present in the latter (Figure 1).

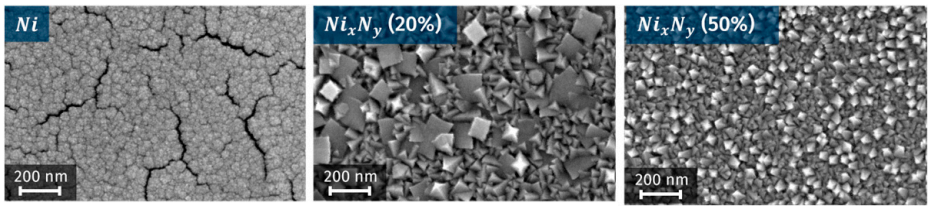

Figure 1. (From **left** to **right**) SEM images of Ni, Ni_xN_y (20%), Ni_xN_y (50%).

Electrochemical characterization: For all samples, we extended the potential window to more cathodic and anodic potentials and observed no significant effect on the glucose oxidation current. Thus, we kept the potential window at [−0.2 V to 0.8 V]. Although the bare Ni was the least resistive, the Ni_xN_y (50%) sample exhibited a 30% lower Ni(II)/Ni(III) peak separation ΔV relatively to bare Ni and a 30% overall increase in sensitivity towards glucose. The Ni_xN_y (20%) sample showed a linear range limited to 5 mM (instead of 10 Mm) and more than double the ΔV (Figure 2).

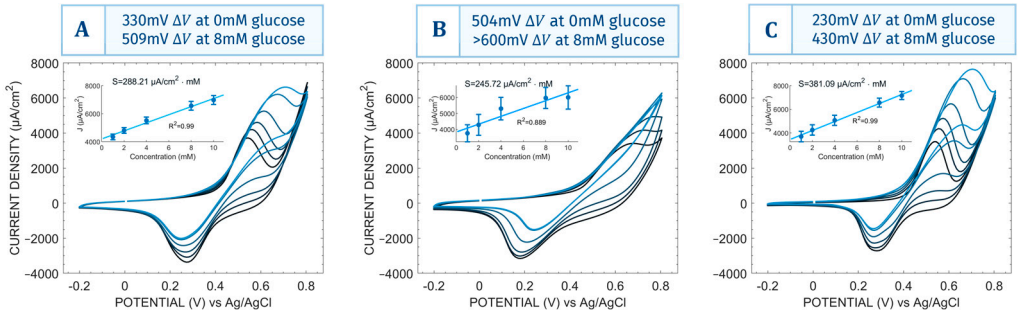

Figure 2. Cyclic voltammograms of Ni (**A**) Ni_xN_y 20% (**B**), Ni_xN_y 50% (**C**) in 0.1 M KOH at 0, 1, 2, 4, 8, 10 mM of glucose.

4. Conclusions

This work depicts the first steps in the development of reactively sputtered Ni_xN_y thin films for non-enzymatic glucose sensing. We observed that a 50% partial pressure of N2 during sputtering significantly increases the electrocatalytic activity. Our results set the groundwork for the fabrication of fully integrated glucose sensors without the need for high temperatures or toxic chemicals.

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