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A potentiometric tattoo sensor for monitoring ammonium in sweat

Tomàs Guinovart,[ab] Amay J. Bandodkar,[a] Joshua R. Windmiller,[ac] Francisco J. Andrade*[b] and Joseph Wang*[a]

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The development and analytical characterization of a novel ion-selective potentiometric cell in a temporary-transfer tattoo platform for monitoring ammonium levels in sweat is presented. The fabrication of this skin-worn sensor, which is based on a screen-printed design, incorporates all-solid-state potentiometric sensor technology for both the working and reference electrodes, in connection to ammonium-selective polymeric membrane based on the nonactin ionophore. The resulting tattooed potentiometric sensor exhibits a working range between 10^{-4} M to 0.1 M, well within the physiological levels of ammonium in sweat. Testing under stringent mechanical stress expected on the epidermis shows that the analytical performance is not affected by factors such as stretching or bending. Since the levels of ammonium are related to the breakdown of proteins, the new wearable potentiometric tattoo sensor offers considerable promise for monitoring sport performance or detecting metabolic disorders in healthcare. Such combination of the epidermal integration, screen-printed technology and potentiometric sensing represents an attractive path towards non-invasive monitoring of a variety of electrolytes in human perspiration.

1. Introduction

Ammonium is present in blood mostly as a result of the breakdown of proteins.[1] For this reason, the determination of the levels of this cation in plasma provides extremely relevant physiological information related to the metabolic state of the individual, dietary conditions, or even liver malfunctions. During exercise, for example, ammonium concentration varies when changing from aerobic to anaerobic state.[2,3] Also, it has been shown that diets with low levels of carbohydrates may lead to temporary increased levels of ammonia in plasma, which is sometimes evidenced as an ammonia smell in sweat. Last, but not least, since the liver converts ammonia to urea prior to its excretion, high ammonia levels can be used as markers of hepatic disorders, such as hepatitis or cirrhosis.[4] All in all, monitoring the levels of ammonium may provide a wealth of information, which ranges from the improvement of the sport performance and monitoring metabolic state to the screening of the health status of individuals. Unfortunately, monitoring ammonium in plasma requires the collection of blood samples, which is a serious limitation during exercise or military activities.

On the other hand, ammonium is excreted through sweat by non-ionic diffusion from plasma,[5] and many studies have shown that the ammonium levels in sweat can be directly correlated with its concentration in plasma. Czarnowski *et al.* examined the relationship between ammonia and urea levels in plasma and ammonium concentration in sweat. Their studies suggest that ammonia in plasma is the main source of ammonium in sweat.[6] Subsequent studies of ammonium secretion *via* sweat have been carried out through physical exercise such as running, where sweat was collected with gauze pads. The measurements in the collected sweat of the total amount of nitrogen excreted (urea, ammonium and amino acid loss) concluded that the difference of nitrogen loss by comparing both methods is relatively small.[7] Other tests performed under submaximal cycling exercise (and comparing the results of ammonium with both, urea and lactic acid) suggested that ammonium is secreted through sweat during short-term exercise on the onset of sweating.[8] Yuan *et al.* studied the effect of one year non-specific program on the ammonia threshold, which was uniquely correlated to endurance time.[9] Also, studies performed with rugby players, where the ammonium levels in sweat were monitored before, during and after the game, showed a significant increase while playing the match. This was also correlated with the concentration of ammonia in blood, which showed an increment almost three times higher.[10] Last, but not least, studies of the ammonium levels in sweat were also performed with a low-carbohydrate (LC) and normal diet, in both cases combined with non-exhausting exercise. The clear

[a]Department of Nanoengineering, University of California San Diego, La Jolla, CA, 92093-0448, USA. E-mail: josephwang@ucsd.edu
[b]Departament de Química Orgànica, Analítica i Nanosensors, Universitat Rovira i Virgili, C/Marcel·lí Domingo s/n, Tarragona, 43007, Spain. E-mail: franciscojavier.andrade@urv.cat
[c]Electrozyme LLC, Executive Square (Suite 485), San Diego, CA 92037-4225, USA