

Diffusion kinetics of the glucose/glucose oxidase system in swift heavy ion track-based biosensors



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ABSTRACT

For understanding of the diffusion kinetics and their optimization in swift heavy ion track-based biosensors, recently a diffusion simulation was performed. This simulation aimed at yielding the degree of enrichment of the enzymatic reaction products in the highly confined space of the etched ion tracks. A bunch of curves was obtained for the description of such sensors that depend only on the ratio of the diffusion coefficient of the products to that of the analyte within the tracks. As hitherto none of these two diffusion coefficients is accurately known, the present work was undertaken. The results of this paper allow one to quantify the previous simulation and hence yield realistic predictions of glucose-based biosensors.

At this occasion, also the influence of the etched track radius on the diffusion coefficients was measured and compared with earlier prediction.

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1. Introduction

There are quite a number of strategies to use swift heavy ion tracks for producing biosensors [1]. One is e.g., the pore blocking concept introduced by Siwy et al. [2], which makes use of the blocking of an individual nanopore tip in a thin polymeric foil, by trapping there at a suitable sensor molecule *S* a traversing adequate analyte molecule *A* in a carrier liquid: $A + S \rightarrow AS$. Alternatively, one can e.g., make use of the principle chemical reaction: $A + S \rightarrow P$, with *A* being the analyte under consideration, *S* being a suitable (preferentially enzymatic) material and *P* being the products of that reaction. Specifically, if the charge states of *A* and *P* differ, one can exploit these differences for the sensing of *A*, as they result in differences of the sensor's resistivity. In order to maximize these differences, the sensing reaction should preferentially take place in confinement, such as given within very long and very narrow etched ion tracks (where the aspect ratios $F = \text{length/diameter}$ may reach the order of ~ 1000 or so) in e.g.,

thin polymeric foils [3]. Here, the confinement leads to an enrichment of the products *P* as long as their production rate within the tracks $R_{\text{production}}$ (determined by the rate of analyte indiffusion $R_{\text{anal,indiff}}$ and the enzymatic reaction activity η) exceeds their diffusional loss $R_{\text{prod,out}}$ from the tracks.

As the overall product enrichment during the finally achieved dynamic equilibrium: $R_{\text{production}} = R_{\text{prod,outdiff}}$ depends on all above-mentioned parameters, a corresponding diffusion simulation was performed [4]. The simulation was based on the simplifying assumption that, in spite of the nanometer-sized etched tracks, nanofluidic effects do not play any role in this case as the bonding of enzymes on the track wall largely neutralizes all surface charges, hence rendering nanofluidics obsolete here (the validity of this assumption is examined in greater detail in another work [5]). A bunch of curves with generalized dimensions (to eliminate *F* in the calculation) was obtained for the description of such sensors that depended on the parameters η (for simplicity assumed there to be 1), $D_{\text{anal,indiff}}$, and $D_{\text{prod,outdiff}}$, with $D_{\text{anal,indiff}}$ and $D_{\text{prod,outdiff}}$ being the diffusion coefficients of indiffusing analyte and outdiffusing products, respectively. However, as hitherto the latter diffusion coefficients were not yet properly known, it could not yet be decided unambiguously which of the curves represents reality.

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For this sake, the present work was undertaken. As a representative example, we selected the system: [A = glucose (G) solution] + [S = glucose oxidase (GOx), fixed to the etched track walls] → [P = gluconic acid (GA) solution]. The obtained diffusion coefficients for the migration of {A into P} and {P into A} allow one to quantify the previous simulation for that system and yield realistic predictions of glucose-based biosensors. At that occasion we also recorded the diffusion coefficients of A and P in water and of water in P, and of solutions of the enzyme (S = GOx) in water. Finally, we also determined the dependence of the diffusion coefficients of P in H₂O on the radius of the etched tracks.

2. Experimental

For the first measuring series (“A” in Table 1), 12 μm thick PET foils were obtained from P.Apel (JNRI Dubna, Russia) which were irradiated with 250 MeV Xe ions up to fluences of $4 \times 10^6 \text{ cm}^{-2}$. They were etched a few years ago from both sides with 2 M NaOH at ~45 °C until cylindrical etched ion tracks of about ~100 nm radius were obtained. The track etching was performed at temperatures somewhat lower than usual in order to define properly the track diameter well by stopping in time and not to “overshoot” the desired diameter. In another measuring series (“B” in Table 1), 19 μm PET foils, irradiated in ca. 2014 by 170 MeV Xe ions up to a fluence of $1 \times 10^7 \text{ cm}^{-2}$, were etched with 4 M NaOH at room temperature for different times, to obtain various average track radii between 0 and 50 nm.

For both the track etching and the subsequent diffusion measurements the foils were inserted into the center of a specially designed measuring chamber (designed after the example of R. Spohr; pers. Commun., 2005, and Ref. [6]), that separated it into two neighbouring compartments. The compartments used here have volumes of ~1 cm³ each (however, especially for biological work with small quantities of expensive materials also chambers with volumes of ~0.1 cm³ are available). Additionally, to minimize the adsorption of biomaterials onto the chamber walls that might lead to contaminations, the chamber walls were made of Teflon.

All foil manipulations – i.e., both the etching and the subsequent diffusion examinations – were recorded by either a classical Amperemeter or an oscilloscope, combined with a pulse generator. In the first case, 2.15 V DC was applied across the foil via two Pt electrodes, submerged in each of the two compartments (see Fig. 1). In the latter case, 5 V AC was applied to two Ag/AgCl

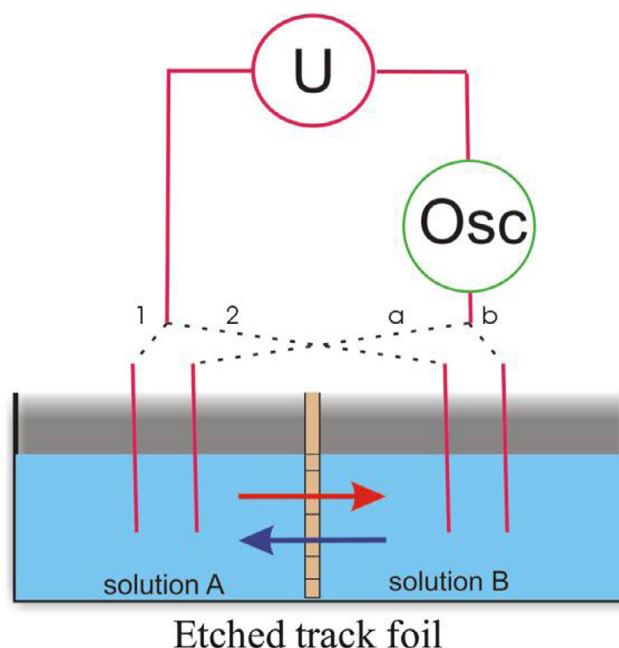


Fig. 1. The measuring principle with its setup. U = voltage source, Osc = oscilloscope in current-measuring configuration. In experiment Nr. 2 of the first series, Osc was replaced by an amperemeter. The arrows show the directions of the diffusion of solutions A and B across the foils. Depending on the type of examination (see text), the foils either contained as-etched tracks or etched tracks clad with the enzyme GOx. The weak dashed lines between the red cables indicate the different possibilities of interconnection for the different experiments. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

electrodes in both compartments each, and measuring the current passing through the foil in the “Transient Recorder” mode of a Velleman PCSGU250 system which is a pulse generator/oscilloscope combination. Tests revealed that the different measuring strategies did not have any influence on the measuring results. Also, the influence of possible polarisation induced by the DC measurement was negligible.

For the diffusion examinations, the compartments were filled with those aqueous biomaterial solutions that were important for this work, namely glucose (here: 0.1 M solution), GOx (here:

Table 1
Overview about the different experiments performed with the setup of Fig. 1, and their results (average of ~2 to 3 measurements each). Several foils with different etched tracks (as-etched and enzyme-clad) were used. For details please consult the text.

SeriesNr.	Measuring Technique and foil type	Solution A	Solution B	Track type, radius [nm]	Diffusion result [cm ² s ⁻¹]
A,1	Oscilloscope; etched track foil only	Glucose	Water	etched, ~100 etched, ~50	$D_{\text{glucose}} \gg 1 \times 10^{-8}$ $D_{\text{glucose}} = 1.7 \times 10^{-9}$ $D_{\text{water}} = 1.2 \times 10^{-8}$
A,2	Oscilloscope; etched track foil only	Gluconic Acid	Water	etched, ~50	$D_{\text{gluc/H}_2\text{O interdiffusion}} = 4.0 \times 10^{-9}$ $D_{\text{gluconic acid}} = 7.0 \times 10^{-9}$ $D_{\text{water}} = 1.5 \times 10^{-7}$
A,3a	Ampere-meter; etched track foil only	Glucose	Gluconic Acid	etched, ~100	$D_{\text{gluconic acid /H}_2\text{O interdiff}} = 1.4 \times 10^{-7}$ $D_{\text{glucose}} = 3 \times 10^{-9}$
A,3b Bio-sensor	Oscilloscope; etched track foil with GOx	Gluconic Acid	Water	etched, ~100	$D_{\text{gluconic acid}} = 4 \times 10^{-9}$ $D_{\text{gluconic acid}} = 9 \times 10^{-9}$
A,4 Bio-sensor	Oscilloscope; etched track foil with GOx	Glucose	GOx	etched, ~100	$D_{\text{glucose}} \gg 1 \times 10^{-8}$ $D_{\text{GOx}} = 4.5 \times 10^{-9}$
B,1	Oscilloscope; etched track foil only	Gluconic Acid	Water	unetched, 0 etched, 7.5 etched, 13 etched, 17 etched, 30 etched, 50	$D_{\text{interdiffusion gluc. \& GOx}} = 3.2 \times 10^{-9}$ $D_{\text{GOx}} = 1.14 \times 10^{-9}$ $D_{\text{gluconic acid}} = 2.5 \times 10^{-9}$ $D_{\text{gluconic acid}} = 2.3 \times 10^{-9}$ $D_{\text{gluconic acid}} = 3.0 \times 10^{-9}$ $D_{\text{gluconic acid}} = 5.2 \times 10^{-9}$ $D_{\text{gluconic acid}} = 5.4 \times 10^{-9}$

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