Supporting Information

Wireless bipolar electrode based textile electrofluidics: towards novel micro-total-analysis systems

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The supporting information provides three videos showing: redox reactions on the surface of polyester braided structure carrying a bipolar electrode in centre, the formation of ion depletion zone near the cathodic end of bipolar electrode inside braid and subsequent concentration enrichment of fluorescein and finally, the simultaneous separation, isolation and enrichment of negatively charged fluorescein from a mixture containing rhodium-B. It also provides a calibration curve for fluorescent intensity measurement on braid surface, an example of gaussian fit to the fluorescence intensity profile of the focused band of a fluorescein marker (FL) and data acquisition (concentration factor) determination using MATLAB code.

S1. Wireless electrochemistry on polyester braided structure

Video V1 shows oxidation and reduction reactions on the centre of the braided structure near both edges of bipolar electrode (BPE). The appearance of red colour indicates acidic pH due to oxidation at anodic end (right) of BPE and dark green colour represents basic pH due to reduction at cathodic pole (left) of BPE. Here, length of the braided structure is 4 cm connected between two buffer reservoirs and carrying a 4mm long platinum wire in the centre of the braid in axial direction, which act as BPE, when 200 V electric potential was applied to driving electrodes. One drop of universal pH was added to 2.5 mM TRIS-CHES (pH 8.8) electrolyte solution to visualize the electrochemical reactions at the BPE driving localised pH changes at the surface of textile. The video was recorded at 60 fps using a handheld mobile device, iPhone 11 Pro with 12 MP built-in camera, and is shown here at 32X speed. In this video we can see, on application of electric field across driving electrodes, the electrolyte solution along with pH indicator migrate from anodic reservoir along the braid channel due to cathodic electroosmotic flow (EOF). The pH indicator changes its colour locally as it approaches the position of the BPE (platinum wire) in the centre of braid due to oxidation and reduction reaction, and the poles of BPE arrange in the polarity opposite to the driving electrodes. This can be understood by noticing the same red colour due to oxidation at the driving electrode in anodic reservoir. The dark green colour due to reduction at BPE is hard to distinguish from the green colour of the electrolyte solution at pH 8.8, however it's still noticeable.

S2. Fluorescein concentration enrichment

Video V2 shows formation of ion depletion zone, appearance of focused FL band and migration and enrichment of focused band with time on the surface of braided structure for a single experiment. Here, a 4 cm long polyester braided structure with 4 mm BPE in centre connected between two buffer reservoirs; the reservoirs and braid channel are filled with 0.2 5µM FL in 2.5 mM TRIS-CHES buffer solution. Videos were captured at 30 fps using a fluorescent microscope with built in camera and presented here at 16X speed. In this video, we can see the appearance of an ion depletion zone (IDZ) appear near the cathodic

edge of BPE on application of 400 V across the driving electrodes, which subsequently sweeps the charged ions near the vicinity of BPE and later, a distinct FL band forms in the anodic section of braid near this IDZ due to electric field gradient focusing. The focused sample band gets brighter and travels towards the anodic reservoir. The increase in fluorescent intensity was indicative of an increase in the concentration of the ions within the sample zone as a result of the interplay between the electrokinetic force of negatively charged FL which dominates over the electroosmotic drag force of bulk solution.

S3. Calibration curve for concentration of fluorescein

For calibration curve, individual braided structures were dipped in a series of solutions containing different concentrations of the fluorescein in 2.5 mM TRIS-CHES. These braided structures were then imaged using fluorescence microscope at 3 different positions. The lighting conditions, magnification settings and working distance of the microscope used to obtain the calibration curve were as similar as possible to those used for the electrofluidic experiments. The average fluorescence intensities were obtained from the fluorescence micrographs using ImageJ software for each concentration.

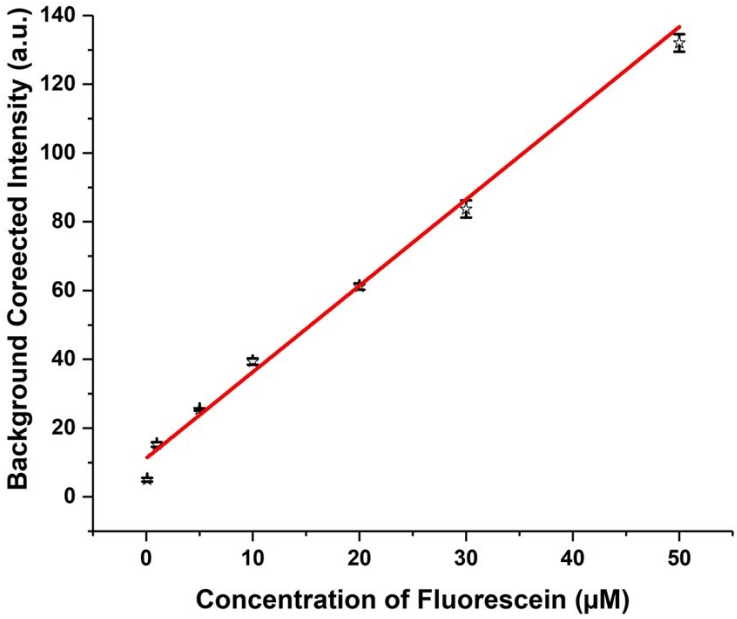

Figure S1. Calibration curve for fluorescein. Background corrected average intensities of fluorescein plotted as a function of molar concentration. Red line is the best linear fit to the data and error bars indicate the standard deviations obtained from intensities at different position on braid, R^2 0.96.

S4. Gaussian fit to the intensity profile of fluorescein

Figure S2 displays an example of the Gaussian fit to the fluorescence intensity distribution of the FL focused band on the braided structure. Experimental fluorescence the signals extracted from fluorescence video image captures and background corrected by subtracting it from the intensities of braid channel filled with electrolyte solution only. A Gaussian distribution was then applied the peak intensity of this Gaussian fitted curve used to calculate the maximum intensity in the focused band.

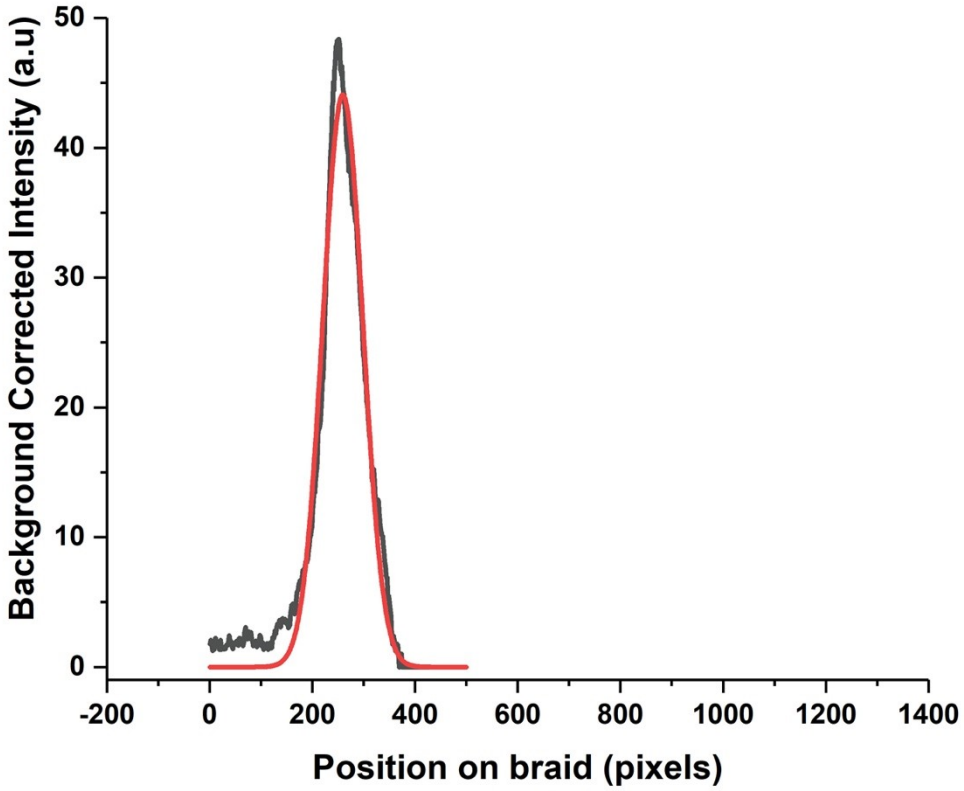

Figure S2. Width averaged intensity profile of the fluorescein focused band as a function of axial position on braid obtained from a single redox focusing experiment at 300 s, after applying 400V voltage across a 4 cm long braid channel (housing a 4mm platinum wire in centre) connected between two buffer reservoirs carrying driving electrodes. Both reservoirs and braid channel filled with 0.25 µM fluorescein in 2.5 mM TRIS-CHES buffer solution. Solid black line represents the background corrected experiment intensity data and maximum measured intensity in the focused band. Solid red line is a Gaussian fit to the data, used to calculate the maximum concentration of sample in the focused band. Dotted lines indicate the axial position of bipolar electrode (platinum wire) inside the braid channel.

S5. Data acquisition and MATLAB code

The following steps were taken in a MATLAB script for quantification of data from recorded videos. First, the individual image frames were extracted out of the videos recorded by the fluorescent microscope camera. The individual image frames were processed in sequence for the quantification of pixel intensity as a measure of the fluorescent intensity recorded by the microscope. Two supporting images were required before the processing of the video: the saturated image (braided structure with the fluorescent dye of a known concentration) (C_S) and the blank image (braided structure, saturated with buffer only). To do this, images were read with the imread function, converted to grayscale images with rgb2gray function, and finally converted to double precision values using the double function.

From the blank image, the background fluorescence intensity (IB) was determined by finding the pixel of maximum intensity. For the saturated image, the background contribution determined from the blank image was subtracted. Following this, any intensity number below 0 was converted into a NaN. The averaged fluorescent intensity of the saturated image (IS) was determined by width averaging (column-wise) throughout all of the lateral image pixels (excluding NaN values using nanmean function), followed by horizontal averaging, which resulted in a single pixel value. For example, if the image has dimension of 1024 rows by 1280 columns, the vertical averaging results in an array of 1 row by 1280 columns which is then further averaged laterally.

For extracting individual frames out of the video, first a video object is created using the videoReader function. This function creates an object that allows for querying information from the video. In this case we give the name of v to the video object. The total video length is extracted using v.Duration function. From here, we define the number of frames to process by defining the spacing between individual frames, for example 10 second intervals. The number of frames defines the number of iterations to run the script.

The script used for analysing one image frame at a time has described here. For each iteration, need to define the v.CurrentTime, which gives the instruction of readFrame function to look at that particular time (the time corresponding of the iteration number multiplied by the spacing between individual frames). Then, the individual frame, including its time stamp, was extracted for processing. The intensity of the selected image frame was first subtracted by the background contribution,

and values equal or below 0 intensity were converted into NaN. To track the lateral displacement of the fluorescent along the different frames, pixel coordinates of the fluorescent band centre were located. The averaged fluorescent intensity of the individual image frame is determined by width averaging as said before. Plotting of the intensity along the image width results in the Figure S2, black spectrum. On the same array a Gaussian fit was applied using the function fit (gauss1). From the gauss fit the maxima (C_M), the full width at half maximum (FWHM), and the fitting coefficients were extracted. An exception was also included in case the gauss fit does not converge to a minimum, for cases where the fluorescent band was not present in the image or the fluorescent band was partially shown in the image frame. In that case, the gauss fit maxima was assigned as the image maximum intensity, and the gauss fitting coefficient as well as the FWHM are assigned to zeros.

The concentration of the concentrated fluorescent band value is calculated by the following equation,

$$C_{value} = C_S \frac{C_M}{I_S}$$

where C_S is the concertation used in the saturated image, G_M is the gauss fitted intensity of the focused band for the individual frame and I_S is the intensity of the saturated image.

After the iteration of the script for all images, we then collect relevant values and save to excel sheets for further evaluation.

S6. Separation, isolation and enrichment from a mixture

Video V3 shows separation of anionic FL from the zwitterionic rhodamine B (Rh-B) in a mixture with TRIS-CHES buffer solution with simultaneous isolation and enrichment of FL for a single experiment. Here, a 0.5 µL aliquot of sample mixture micropipetted on 4 cm long braid channel (4 mm BPE inside) at 5mm distance from the anodic reservoir. The sample mixture consisted of 0.07 µM FL and 1 mM Rh-B in 2.5 mM TRIS/CHES electrolyte solution and the braid channel and both buffer reservoirs were pre-soak and filled respectively with electrolyte solution. Note that, the driving voltage, 400 V was applied for 600 s before application of the sample mixture, ensuring the electrochemical reactions start proceeding at the BPE with the formation of IDZ at the cathodic edge of BPE. The video was captured at 30 fps using the same fluorescent microscope as mentioned in main manuscript with switchable red and green light filters and presented here at 8X speed. It can be seen in this video as soon as the sample mixture is applied on the braid electrofluidic setup, the two solutes anionic FL (green) and zwitterionic Rh-B (red) separate from each other on the basis of their different electrophoretic mobilities and migrate towards the cathodic reservoir along with the cathodic EOF. However, when two solutes approach to the position of BPE, the anionic FL isolate from the remaining mixture due to redox focusing and the zwitterionic Rh-B travel downstream the braid channel. As the experiment proceeds, isolated FL band gets brighter due to increase in concentration of FL ions and remains immobilize.

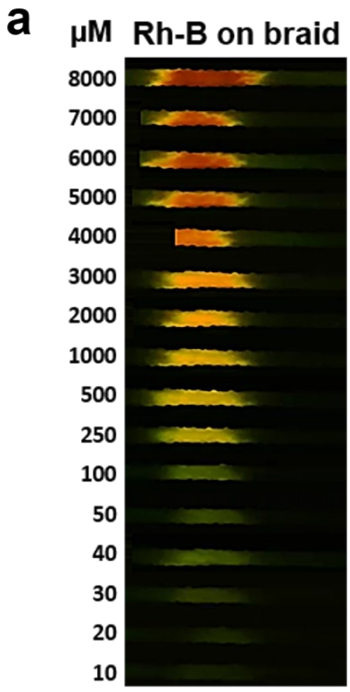

Figure S3. Fluorescent intensities of Rh-B at different concentrations. (a) different colours of Rh-B at different concentrations in 2.5 mM TRIS-CHES buffer solution on the surface of polyester braided structure, when observed with green light emission filter (510 nm) of optical microscope.

The green fringes at the end of the red colour Rh-B band are due to the low concentration of Rh-B ions at the band edges, which appear green with green colour emission filter. To further confirm that no FL ions pass beyond BPE, red light emission filter was switched at 3 and 4 s in this video to monitor the position of Rh-B ions on the braided structure. Moreover, to verify that the green fringes at the end of Rh-B band were the result of low concentration Rh-B ions, the fluorescent intensities of the different concentrations of Rh-B (in 2.5 mM TRIS-CHES) were captured on the braid surface with green colour emission filter as shown in S3 (a).