Electronic Supplementary Material (ESI) for Analytical Methods.

Supporting Information

Comparison of Signal-enhancement Strategies for Carbamazepine Detection in Undiluted Human Saliva Using an Electrochemical Sensor with Stencil-Printed Carbon Electrodes

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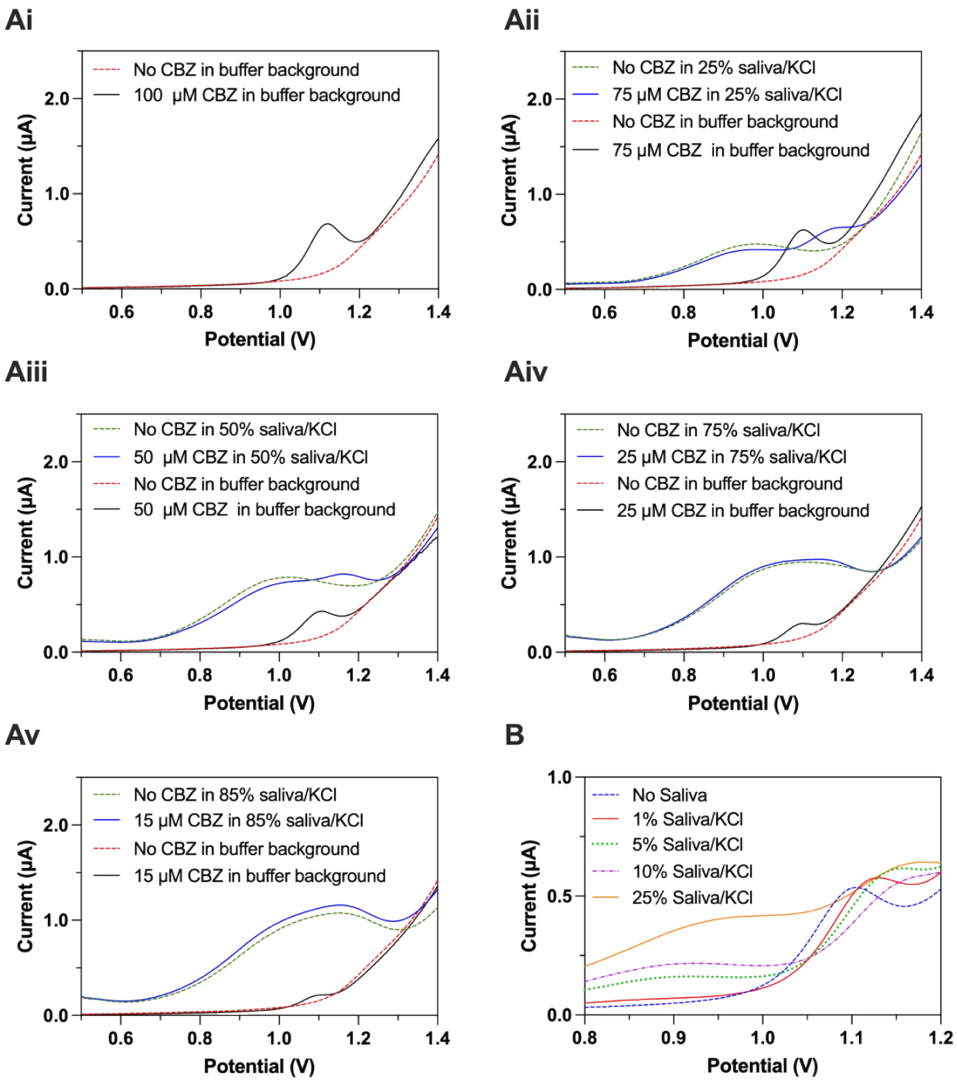
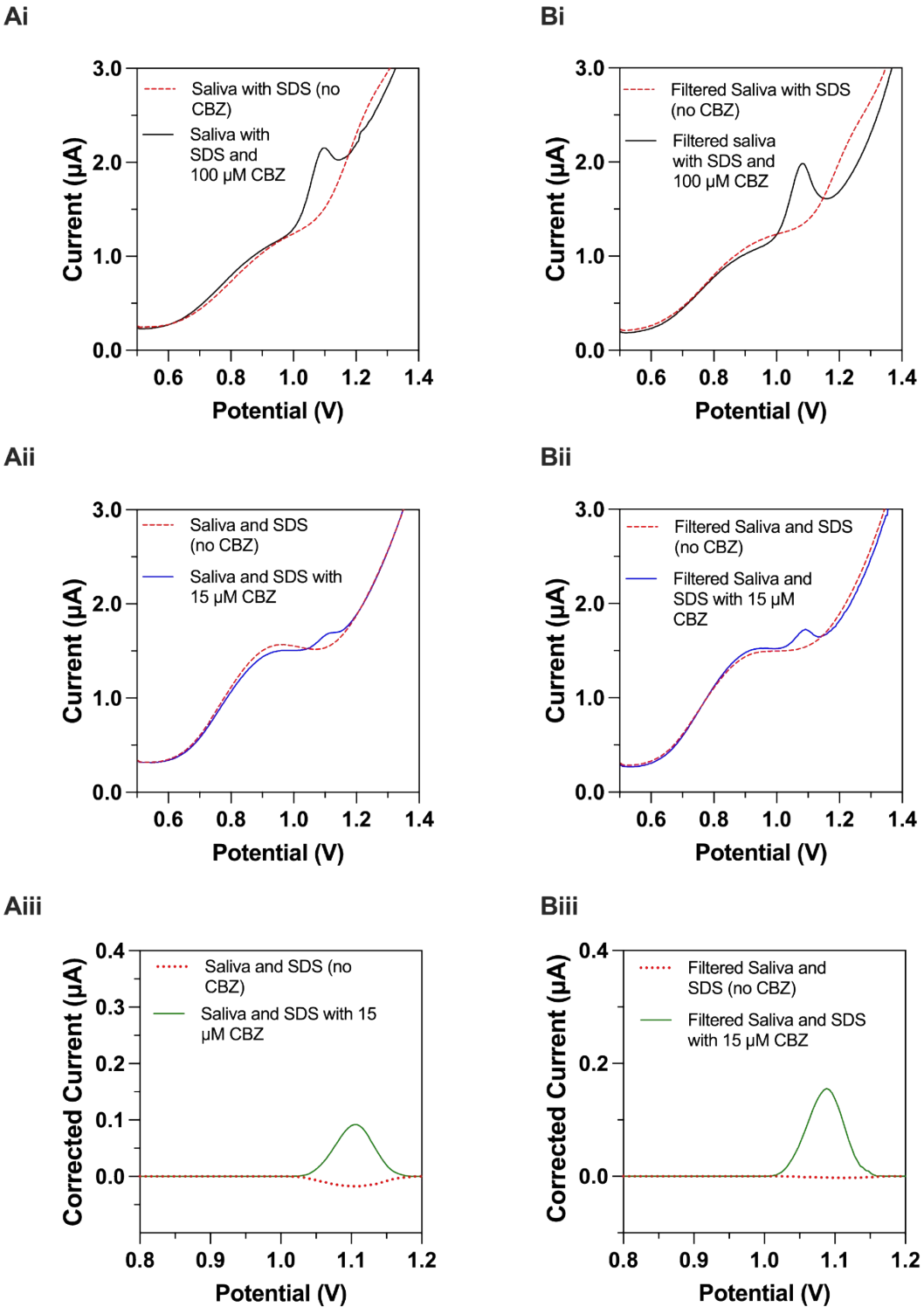


Supporting Information Figure 1: (A) Representative raw curves of a CBZ in buffer concentration series compared to dilution with a saliva/KCl solution at different percentages (i) 100 µM CBZ in 100% buffer, (ii) 75 µM CBZ in 100% buffer vs. 75 µM CBZ in 25% saliva/KCl solution, (iii) 50 µM CBZ in 100% buffer vs. 50 µM CBZ in 50% saliva/KCl solution, (iv) 25 µM CBZ in 100% buffer vs. 25 µM CBZ 75% saliva/KCl solution, and (v) 15 µM CBZ in 100% buffer vs. 15 µM CBZ 85% saliva/KCl solution. Comparison of the two dilution series shows the substantial effect of the saliva background on CBZ peak magnitude. (B) Representative raw curves of 75 µM CBZ with different concentrations of saliva from 0 to 25% saliva/KCl solution. Note that there is an increase in background signal and a shift of the peak to higher potentials as the percentage of saliva is increased.



Supporting Information Figure 2: (A) CBZ detection in saliva with SDS addition (0.4 mM and 0.1 M KCl) vs. (B) in filtered saliva (3 kDa and larger components removed) and SDS addition (0.4 mM and 0.1 M KCl). Each panel displays representative curves for (i) 100 µM CBZ and no CBZ (raw curves), (ii) 15 µM

CBZ and no CBZ (raw curves), or (iii) 15 μM CBZ and no CBZ (background-corrected curves). For these experiments, the dilution of saliva was less than 8%.

Supporting Information Table 1: Oxygen plasma treatment of the stencil-printed carbon electrodes affects the contact angle of water on the carbon surface and the flow time of fluid through the electrode-containing channel. The contact angle substantially decreased after oxygen plasma treatment times of 30 seconds and 1 minute relative to the initial higher contact angle measured on the electrode with no treatment. This change in the electrode surface properties from hydrophobic to more hydrophilic with increasing oxygen plasma treatment time correlated with increasing wettability of the electrodes when exposed to fluid as assessed by decreasing fluid flow time through the flow channel past the electrodes.

Oxygen plasma treatment time	Average contact angle (degrees) $N = 4$	Average flow time (seconds) $N = 3$* or 4
No treatment	82±18	72±28
30 seconds	65±13	13±3
1 minute	23±7	9±6

* One of the no treatment replicates did not flow within 3 minutes and was not included in the average and standard deviation.

Experimental: During the fabrication of the polymeric laminate-based devices with stencil-printed electrodes, and after stencil-printing and curing of the carbon electrodes (working and counter), the carbon electrodes on polyester were exposed to oxygen plasma in a plasma asher (PE50, Plasma Etch, Inc., Carson City, NV) at 50 Watts with a chamber pressure of 350 mTorr for periods of 30 seconds and 1 minute. The Ag/AgCl ink was then stencil printed on the polyester sheets and cured in an oven set to 100 ºC for 15 minutes and the electrochemical cells assembled as described above. Flow time was measured as the time for deionized water (12 μL) to flow past the second carbon electrode (~8.3 mm from the inlet) and performed in replicate (N = 3 or 4). Contact angle measurements (FTA 135, First Ten Angstroms, Inc., Newark, CA) were made using 1 μL deionized water droplets placed on stencil-printed carbon surfaces and subjected to oxygen plasma treatment (or not) as specified (N = 4).