

Article

Supplementary Information: Controlled Insertion of Silver Nanoparticles in LbL Nanostructures: Fine-Tuning the Sensing Units of an Impedimetric E-Tongue

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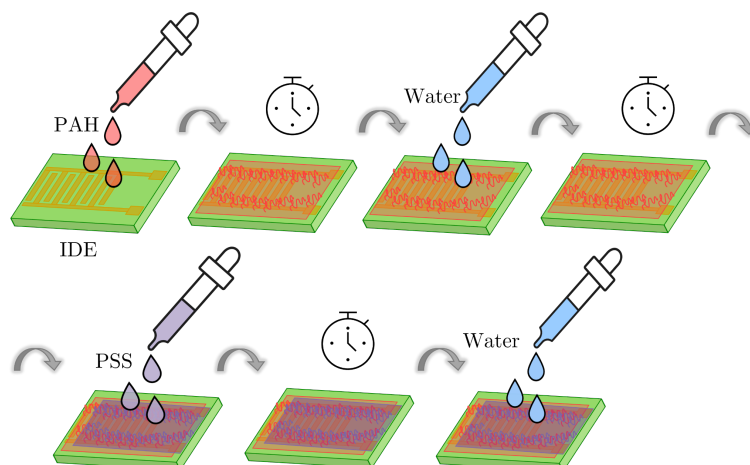


Figure S1. The layer-by-layer (LbL) technique employed in this study involves the application of a drop of one solution containing positive polyelectrolytes onto the interdigitated electrode (IDE) surface for 8 minutes. The excess solution is then removed using a syringe, and the surface is thoroughly washed with ultrapure water to eliminate loosely bound material. Following this, the surface is allowed to dry for 10 minutes. Subsequently, the process is repeated for the negative polyelectrolyte. This iterative process is repeated until the desired number of deposited bilayers is achieved.

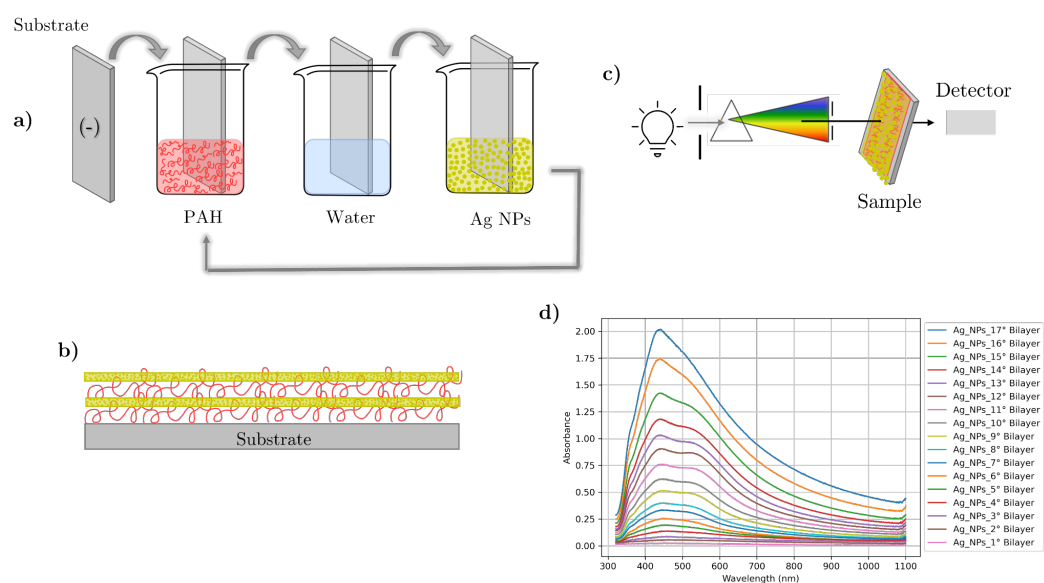


Figure S2. Adsorption kinetics procedure. (a) Cleaned quartz plates undergo an immersion in the PAH solution, followed by rinsing in pure water, and then immersion in the AgNPs solution for varying duration, resulting in the formation of a multilayered LbL structure (b). (c) UV-Vis absorption spectroscopy is employed at each deposition step, confirming material adsorption by the characteristic absorption band of AgNPs at approximately 420 nm (d).

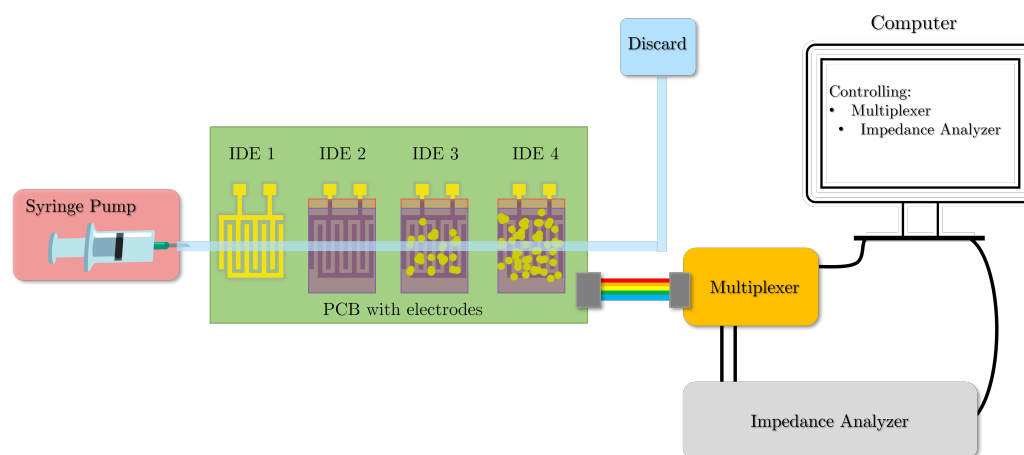


Figure S3. Schema of the microfluidic e-tongue device. Four IDEs are linearly arranged on a gold-plated printed circuit board (PCB). Each IDE comprises four pairs of digits, measuring 5 mm in length, 0.2 mm in width, and spaced 0.2 mm apart. The microchannel dimensions are 500 μm in width, 500 μm in height, and 4 cm in length. The syringe pumps the analyte through the microchannel that passes through four sensory units. The multiplexer switches between each IDE in such a way that the impedance analyzer performs measurements on each sensing unit individually, in triplicate, and with minimal human intervention.

E-tongue applied to discern basic flavors

Figure S4 presents impedance curves obtained for each sensing unit (IDE 1 to IDE 4) corresponding to each basic taste, highlighting distinct behaviors exhibited by all sensing units for the five basic tastes (saltiness, sourness, sweetness, bitterness, and umami). Notably, caffeine (bitterness) and sucrose (sweetness) exhibit similar curves across all sensing units, which aligns with expectations since caffeine is a weak electrolyte and sucrose is a non-electrolyte. Furthermore, measurements were conducted in deionized water both before and after the analytes, as depicted in Figure S5, confirming the absence of potential cross-contamination.

The visual analysis of Figure S4 suggests the formation of a unique fingerprint for each analyte, a crucial characteristic in e-tongue analysis. To enhance discrimination, PCA analyses and *k-means* clustering were performed for triplicated measurements. Figure S6 displays the score plot of PC1 vs PC2, representing a total variance of 99.1%, with PC1 capturing 90.9% of the information, while PC2 describes 8.2%. The evaluation of *k-means* indicates a very robust silhouette coefficient ($SC = 0.96$), forming five groups, each corresponding to a distinct basic taste pattern analyzed. Furthermore, Figure S6 suggests another classification using PC1: (i) weak and non-electrolytes (sucrose and caffeine) all present negative PC1 score values, while (ii) electrolytes (L-glutamic acid, hydrochloric acid, and sodium chloride) have positive scores. Figure S7 presents the score plot using the very same device after 1 year stored without any special care, demonstrating the long-term stability of the sensor.

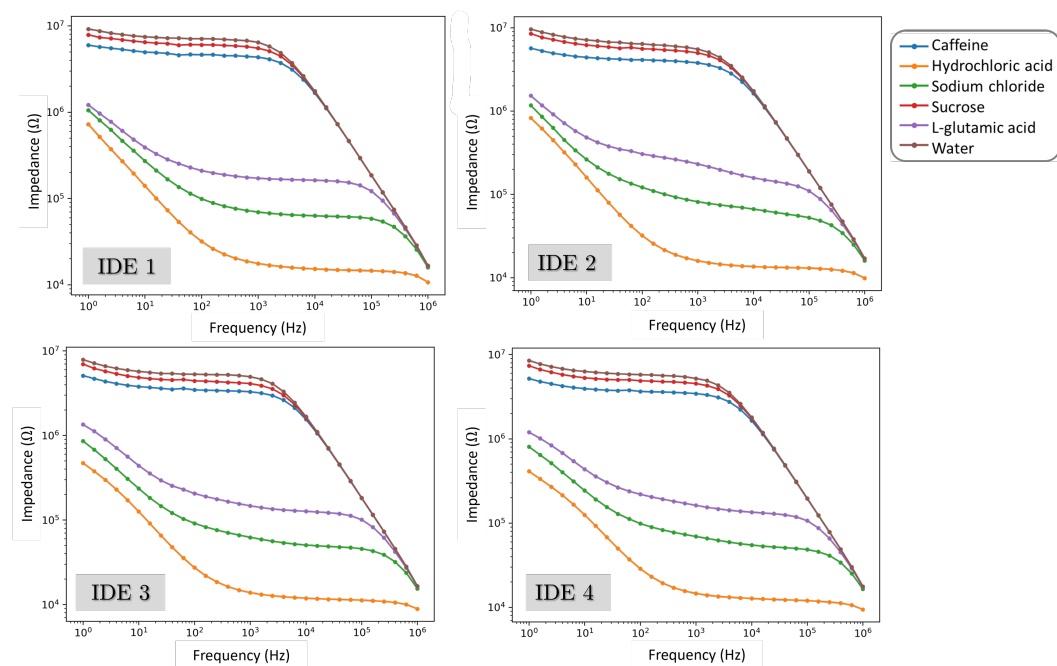


Figure S4. Impedance of the four sensing units, as a function of the variation in excitation frequency, for each basic palate.

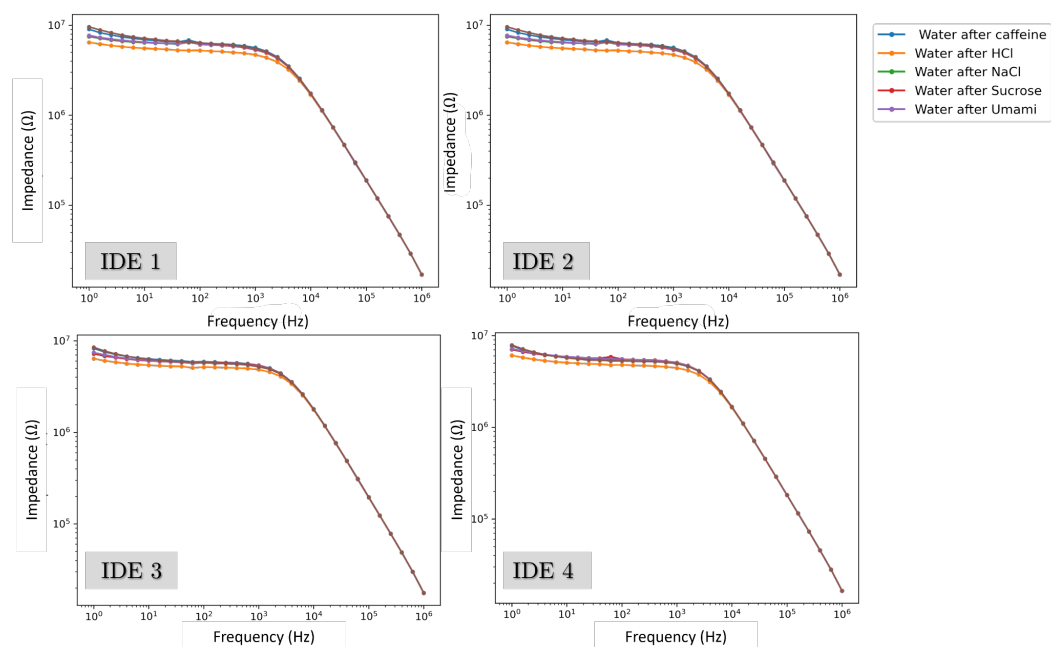


Figure S5. The impedance of the four sensing units was measured as a function of the excitation frequency variation after testing each basic palate with water as the analyte. These results demonstrate the absence of cross-contamination.

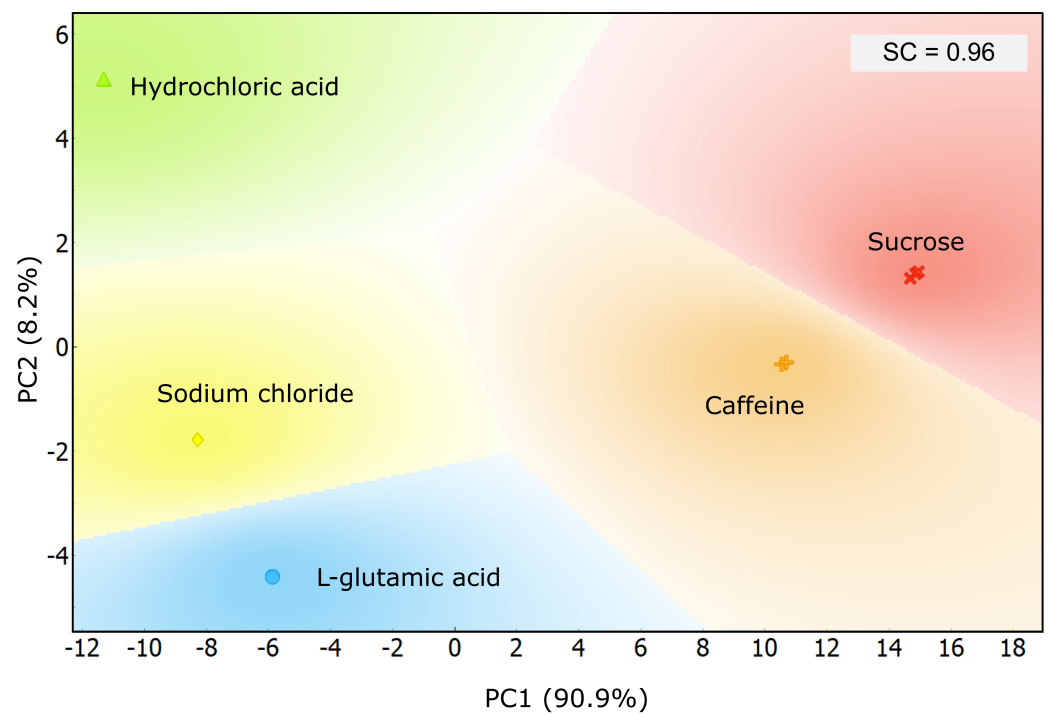


Figure S6. PCA score plot for caffeine (bitterness), hydrochloric acid (sourness), L-glutamic acid (umami), sodium chloride (saltiness), and sucrose (sweetness).

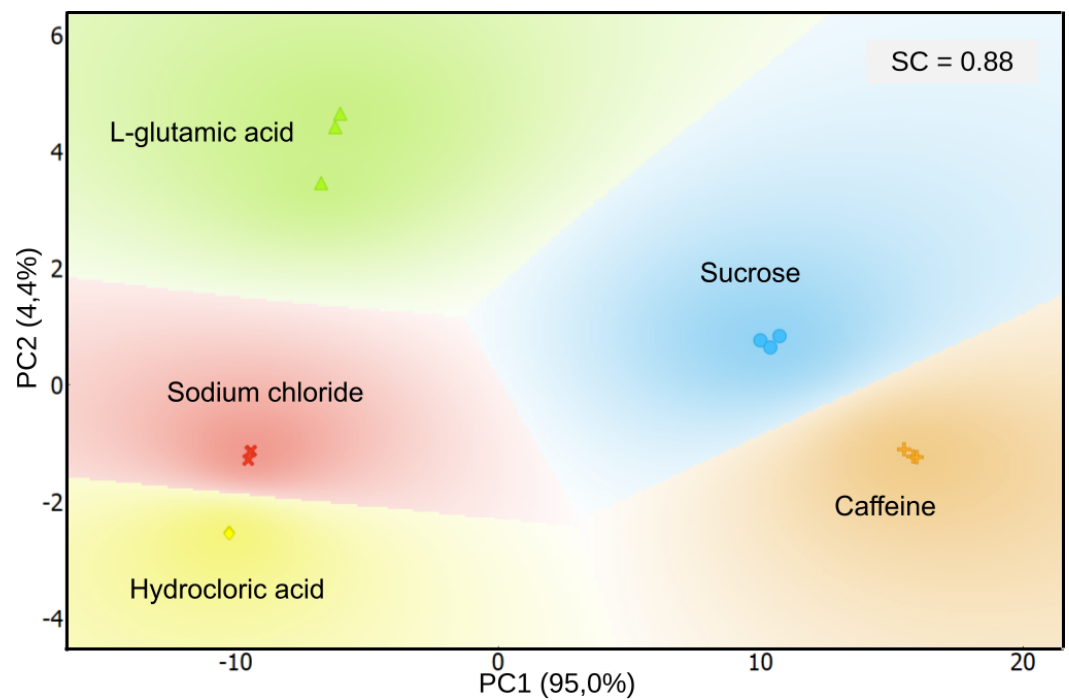


Figure S7. PCA score plot obtained from measurements using the same sensor device used in the measurements presented in S6, after 1 year stored without any special care.