Supporting Information

Nanopaper based Artificial Tongue: Ratiometric Fluorescent Sensor Array on Bacterial Nanocellulose for Chemical Discrimination Applications

Samira. Abbasi-Moayed^a, Hamed Golmohammadi^b and M. Reza Hormozi-Nezhad^{a,b*}

^aChemistry Department, Sharif University of Technology, Tehran, 11155-9516, Iran

^b Chemistry and Chemical Engineering Research Center of Iran, 14335-186, Tehran, Iran

^cInstitute for Nanoscience and Nanotechnology, Sharif University of Technology, Tehran, 11155-9516, Iran

*Corresponding author: (M. R. H. N) email: <u>hormozi@sharif.edu</u>



(b)



Fig. S1: (a) The creation of test zones with hydrophobic barrier (black areas) and with desired patterns (multiwall and two-dimensional cuvette patterns) through direct-printing a toner layer onto the dried BC nanopaper films using an office laser printer. (b) The fabricated sample holder for recording fluorescence spectra of the developed NRFSA platforms

(a)



Fig. S2 (a) The SEM image and (b) EDS analysis of the CDs₃-RhB nanohybrid immobilized on BC nanopaper (CDs₃: CA-Gly).



Fig. S3: The effect of various heavy metal ions (1: Hg (II), 2: Pb (II), 3: Cd (II), 4: Fe (III), 5: Cu (II)) at different concentrations (0-100 μ M) on the fluorescence intensity of RhB immobilized on the BC nanopaper platforms.





RhB



b)



Fig. S4: (a) The fluorescence emission of RhB and three different types of CDs immobilized on the BC nanopaper platforms and (b) their corresponding red and blue values within 24 h.



Fig. S5: The fluorescence emission difference (Δ I) and their corresponding images of three sensor elements on the traditional paper and BC nanopaper (a) SE1 (CDs₁-RhB) (b) SE2 (CDs₂-RhB) (c) SE3 (CDs₃-RhB) in the presence of heavy metal ions (60µM) (1: Blank 2: Hg(II) 3:Pb(II) 4: Cd(II) 5: Fe(III) 6:Cu(II))



Increasing the CDs volume

Fig. S6: Different mixture ratios of CDs and RhB in the developed NRFSAs. The ratio of R/ B= 0.5 was chosen for blank at three sensor elements (SE₁: CDs₁-RhB, SE₂: CDs₂-RhB, SE₃: CDs₃-RhB).



Fig. S7: Two-dimensional Canonical score-plot for the discrimination of heavy metal ions (60 μ M) using the developed NRFSA at different pH values. ((a) pH 3, (b) pH 7, (c) pH 10).



Fig. S8: The difference fluorescence emission (ΔI) of the developed NRFSAs for discrimination of heavy metal ions at different concentrations (a) 10 μ M, (b) 20 μ M, (c) 40 μ M, (d) 60 μ M, (e) 80 μ M , (f) 100 μ M.



Fig. S9: The fluorescence spectra of the (a) BC nanopaper, (b) RhB and (c) three different types of CDs (CA-EDA (1), CA-Urea (2), CA-Gly (3)) on the surface of BC nanopaper platforms.



Wavelength (nm)

(b)Pb(II)



Wavelength (nm)





Wavelength (nm)

(d)Fe(III)



Wavelength (nm)





Wavelength (nm)

Fig. S10: The fluorescence spectra of the developed NRFSA with different sensor elements (SE1, SE2 and SE3) in the presence of different concentrations of (a) Hg(II), (b) Pb(II), (c) Cd(II), (d) Fe(III), (e) Cu(II).



Fig. S11: Three-dimensional Canonical score-plot for discrimination of heavy metal ions using the developed NRFSA (data matrix containing the ratio of fluorescence intensity at 570 and 450 for three different sensor elements for five different metal ions) All of the experiments were performed in triplicate. The concentration ranges of heavy metal ions were 5-20 μ M in LDA analysis.





Fig. S12: (a) The color emissions of the NRFSAs (1) and their corresponding color difference map (2) for different interferences species (b) Three-dimensional Canonical score-plot for the discrimination of heavy metal ions (40-100 μ M) using the developed NRFSA (interferences: Na⁺, K⁺, Ca²⁺, NO₃⁻, SO₄²⁻, PO₄³⁻, Mg²⁺, SO₃²⁻ (1 mM) and Zn²⁺, Fe²⁺ (500 μ M)).



Fig. S13: Two-dimensional Canonical score-plot (a) and HCA dendrogram with Ward linkage (b) for Hg (II), Cd (II) and Pb (II), their binary and ternary mixtures for the discrimination of heavy metal ions using the developed NRFSA.



a)

b)

Fig. S14: Three-dimensional LDA plot for discrimination of five heavy metal ions at concentration range between $20-100\mu$ M in (a) river water sample b) fish sample (test set) using the developed NRFSA.