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Supplemental information

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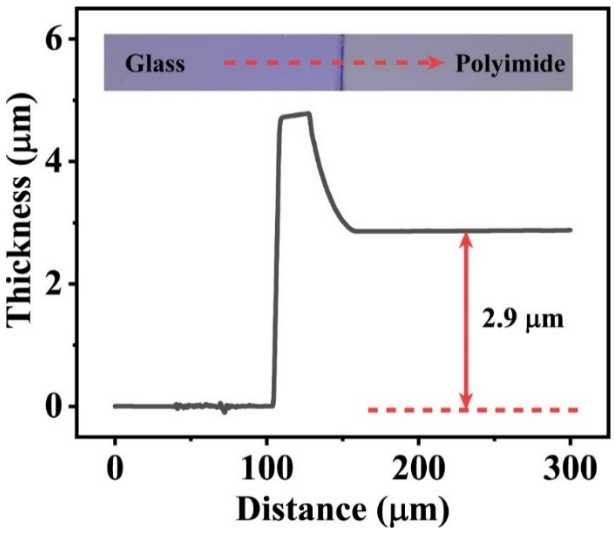

Figure S1. Height profile of spin-coated PI film measured by surface profiler, showing a thickness of ~2.9 μm. Related to Figure 1.
Inset shows an optical image of the sample.

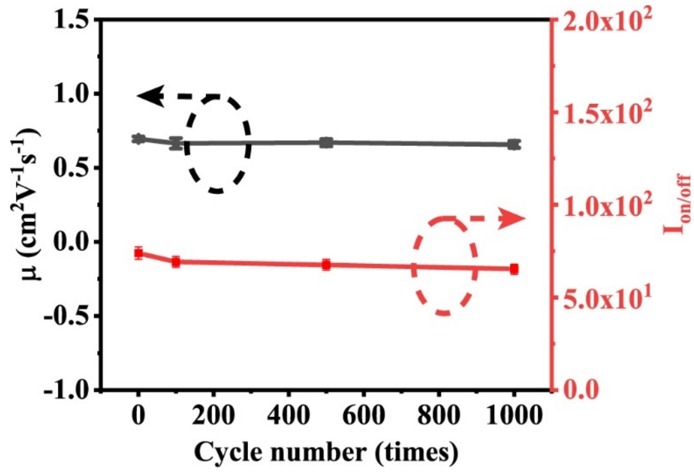

Figure S2. The extracted mobility (*μ*) and current on/off ratio as a function of bending cycles of the flexible bio-FETs with a bending radius of 1 cm, which shows minor performance degradation. Related to Figure 2.

Error bar stands for standard deviations of three tests.

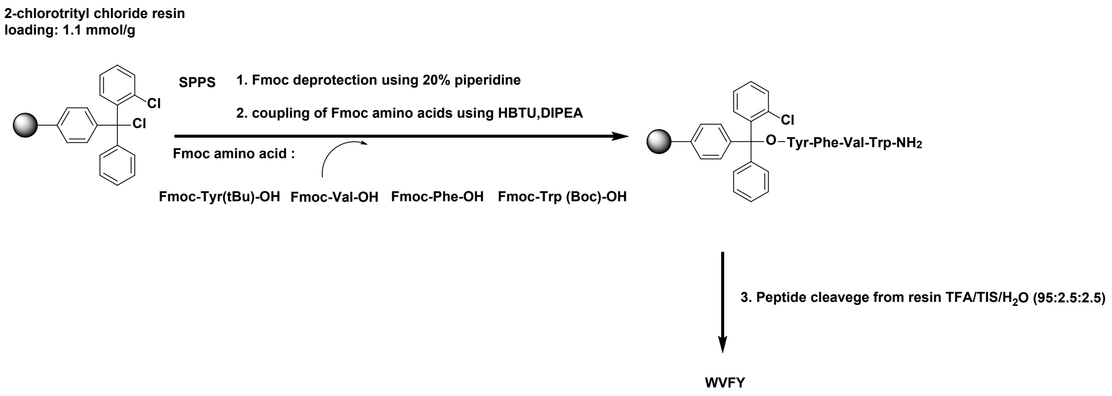

Figure S3. Solid phase peptide synthesis (SPPS) route of WVFY peptides. Related to Figure 3.

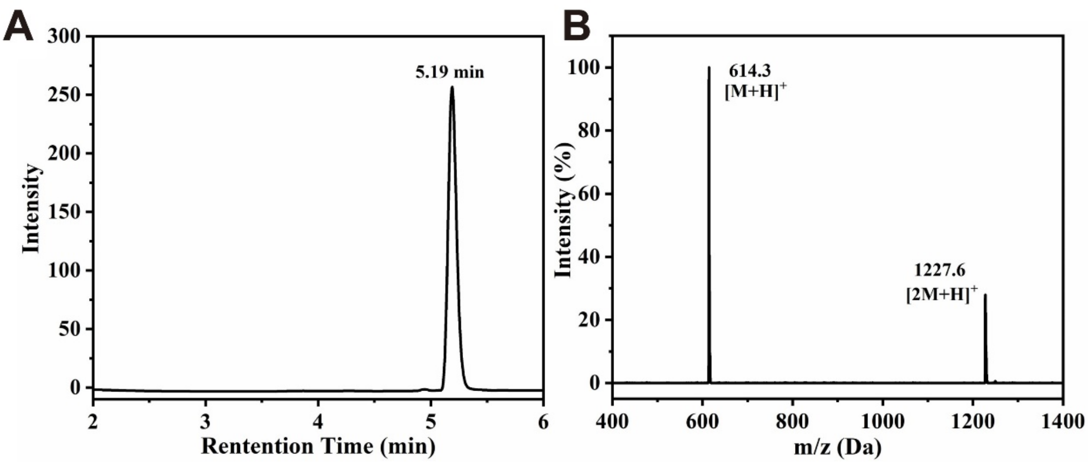

Figure S4. LC-MS spectrum of the purified tetrapeptide WVFY. Related to Figure 3.
The retention time is 5.19 min; the corresponding molecular weight is 613.3 g mol^{-1}; and the purity of WVFY is higher than 96%. MS: calcd. M = 613.3, obsvd. $(M+H)^+$ = 614.3.

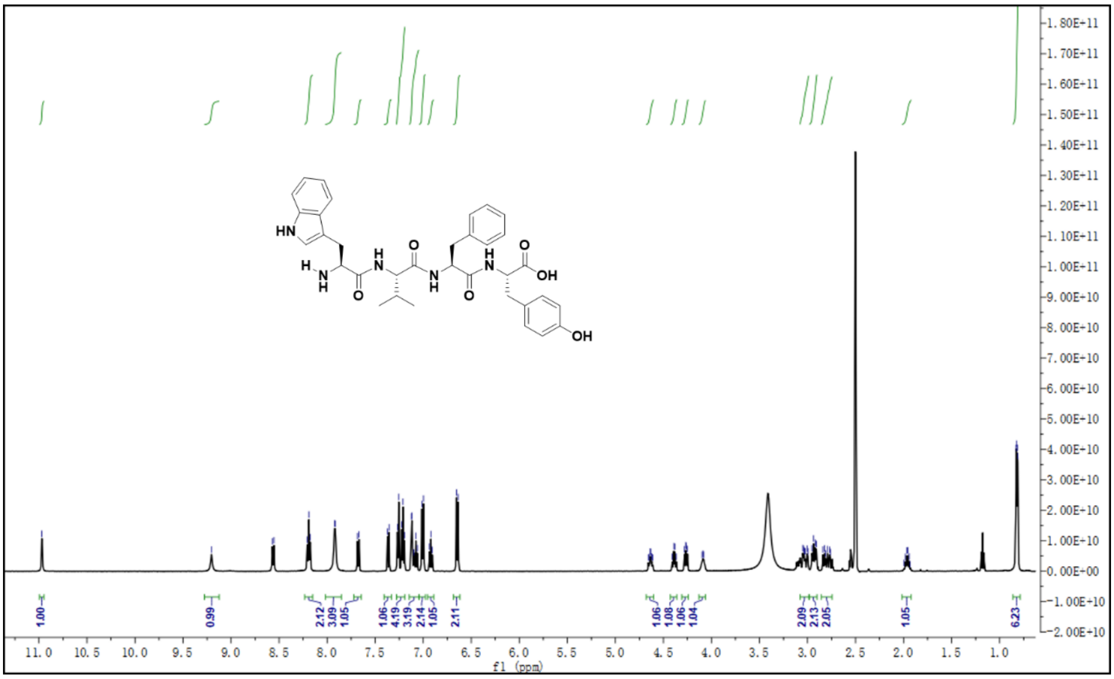

Figure S5. ^{1}H NMR spectrum of the tetrapeptide WVFY. Related to Figure 3.

δ10.97 ppm (s, 1H, NH in tryptophan), δ9.21 ppm (s, 1H, OH in tyrosine), δ8.23 – 8.15 ppm (m, 2H, NH), δ8.02 – 7.85 ppm (m, 3H, NH), δ7.68 ppm (d, 1H, CH in tryptophan), δ7.36 ppm (d, 1H, CH in tryptophan), δ7.28 – 7.19 ppm (m, 4H, CH in phenylalanine/tryptophan), δ7.15 – 7.04 ppm (m, 3H, CH in phenylalanine), δ7.01 ppm (d, 2H, CH in tyrosine), δ6.92 ppm (t, 1H, CH in tryptophan), δ6.65 ppm (d, 2H, CH in tyrosine), δ4.63 ppm (td, 1H, CH in phenylalanine), δ4.43 – 4.36 ppm (m, 1H, CH in tyrosine), δ4.26 ppm (dd, 1H, CH in valine), δ4.13 – 4.06 ppm (m, 1H, CH in tryptophan), δ3.08 – 2.99 ppm (m, 2H, CH_2 in tryptophan), δ2.93 ppm (dd, 2H, CH_2 in phenylalanine), δ2.86 – 2.74 ppm (m, 2H, CH_2 in tyrosine), δ1.97 ppm (dt, 1H, CH in valine), δ0.82 ppm (dd, 6H, $2CH_3$ in valine).

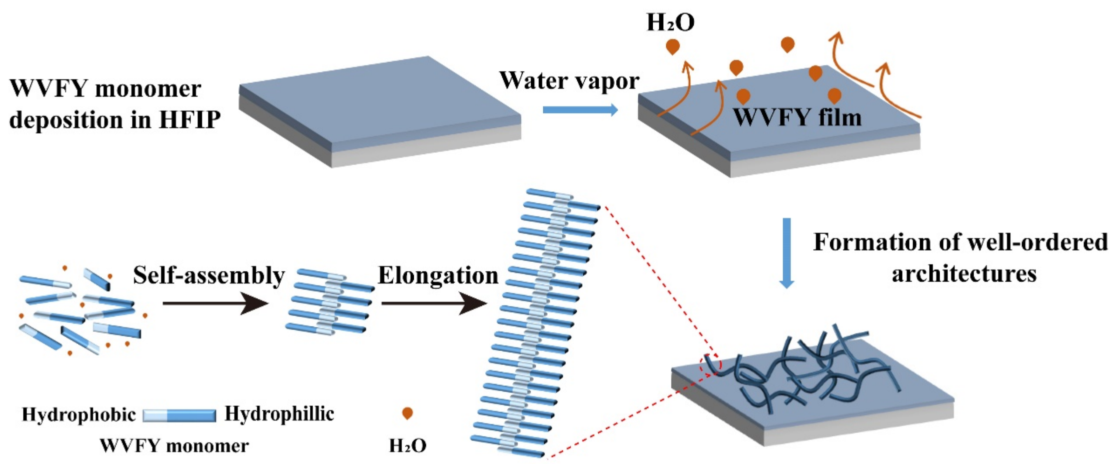

Figure S6. Schematic illustration of the self-assembly process of WVFY peptide nanostructures. Related to Figure 3.

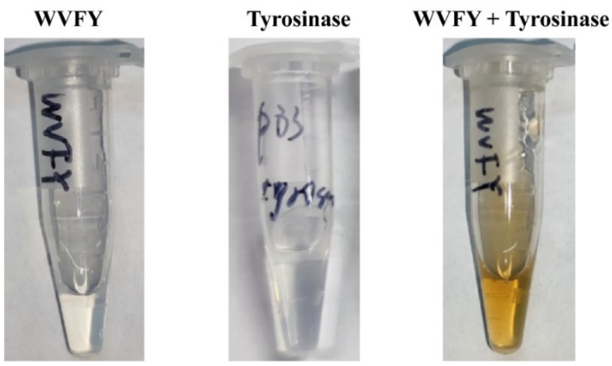

Figure S7. Optical images of WVFY solution before and after oxidized by tyrosinase. Related to Figure 3.

The result indicates that WVFY was transformed.

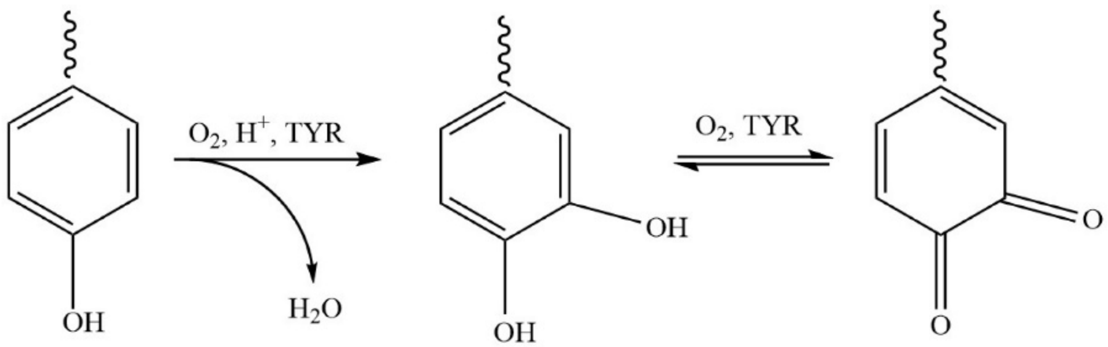

Figure S8. Chemical reaction of WVFY with tyrosinase (TYR). Related to Figure 4.

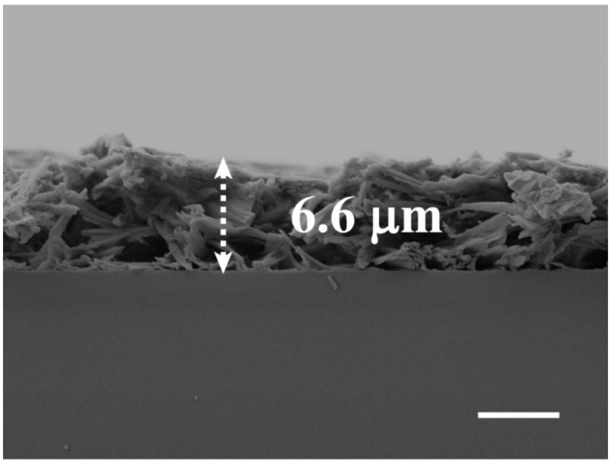

Figure S9. Cross-sectional SEM image of self-assembled WVFY peptides nanostructures, showing a thickness of ~6.6 µm. Scale bar: 5 µm. Related to Figure 3.

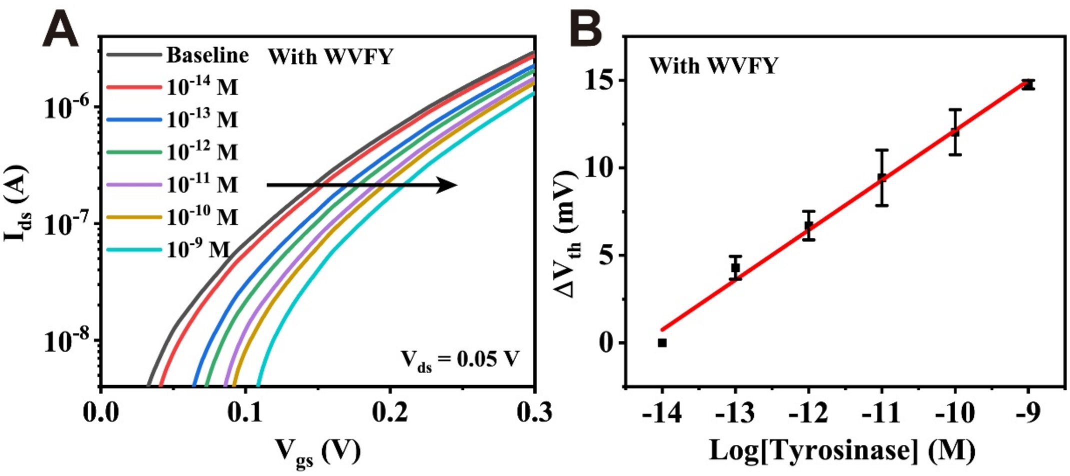

Figure S10. Sensitivity of rigid Al_2O_3/In_2O_3 bio-FETs functionalized with self-assembled WVFY peptides for tyrosinase (TYR) sensing. Related to Figure 4.

(A) Family of typical transfer curves for the device measured in 0.1× PBS solution containing different concentrations of tyrosinase (C_{TYR}): 0.1× PBS (black), 10^{-14} M (red), 10^{-13} M (deep blue), 10^{-12} M (green), 10^{-11} M (purple), 10^{-10} M (ginger), and 10^{-9} M (sky blue), showing a positive shift with the increase of C_{TYR}. (B) A corresponding linear relationship between the ΔV_{th} and C_{TYR} in logarithmic scale. Error bar stands for standard deviations of three tests with $p < 0.05$.

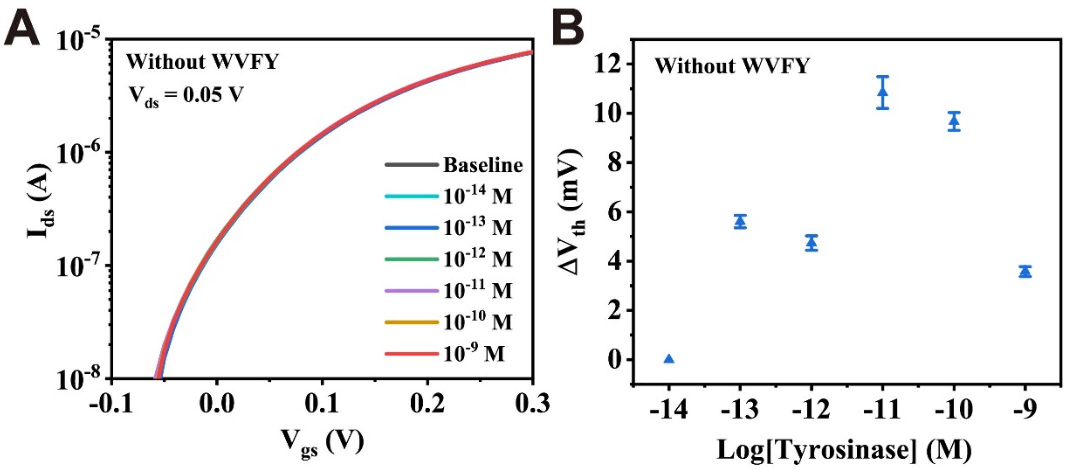

Figure S11. Response of rigid Al_2O_3/In_2O_3 bio-FETs without WVFY peptides to tyrosinase. Related to Figure 4.

(A) Transfer characteristics of bio-FET with various C_{TYR}, indicating no obvious response. (B) The relationship between ΔV_{th} as a function of C_{TYR}, showing a random correlation. Error bars indicate the standard deviations of three repeated tests.

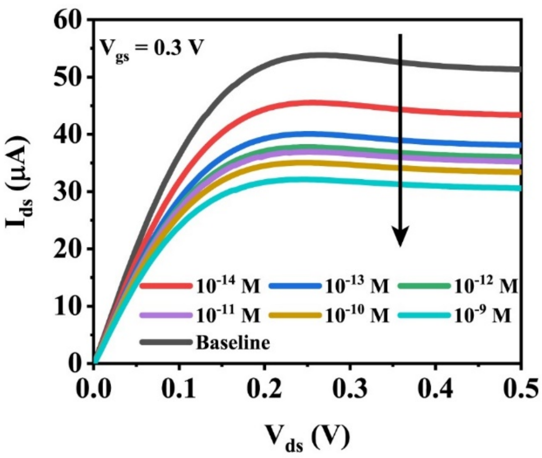

Figure S12. Family of output curves of WVFY modified Al_2O_3/In_2O_3 bio-FETs on rigid substrates with various C_{TYR}. Related to Figure 4.

The saturation current reduced with the accumulation of tyrosinase. The gate bias was 0.3 V.

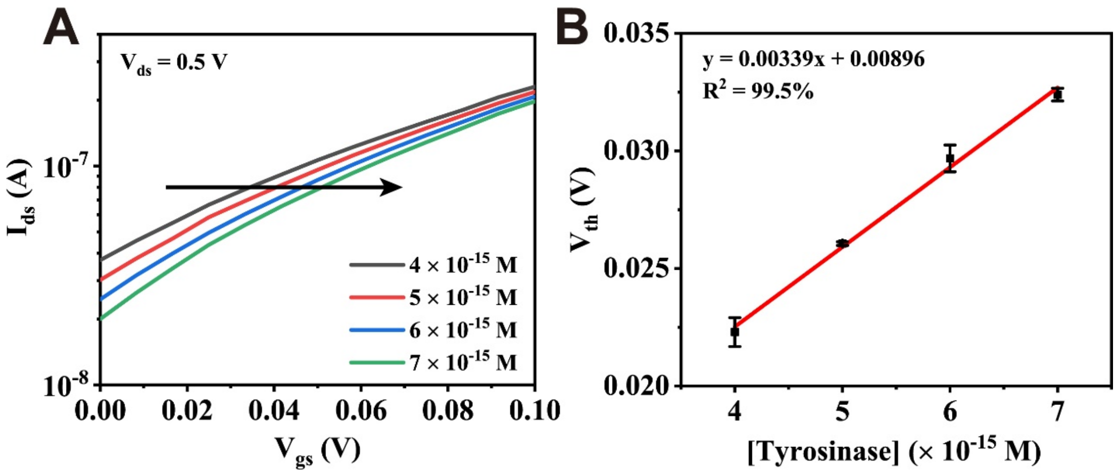

Figure S13. Limit of detection calculation of WVFY modified Al_2O_3/In_2O_3 devices. Related to Figure 4.

(A) Typical transfer curves for tyrosinase sensing in a very low concentration regime from 4 × 10^{-15} to 7 × 10^{-15} M. (B) The V_{th} shift as a function of C_{TYR} in logarithmic scale, demonstrating a linear equation of $y = 0.00339x + 0.00896$ with the R^2 of 99.5%. Error bars indicate the standard deviations of three repeated tests with $p < 0.05$.

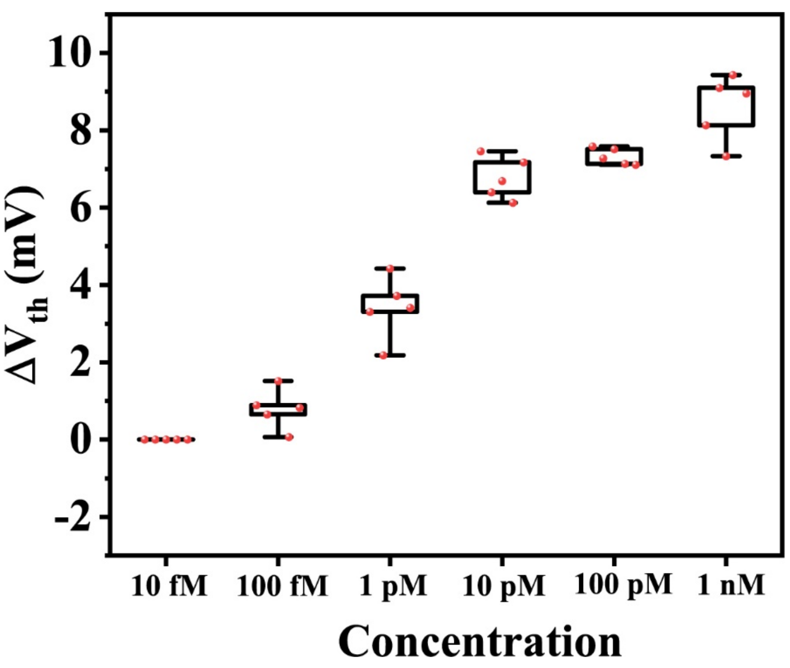

Figure S14. The threshold voltage shift (△V_{th}) as a function of tyrosinase concentration in PBS solution measured by WVFY modified bio-FETs. Related to Figure 4.
Error bars indicate the difference of five repeated tests.

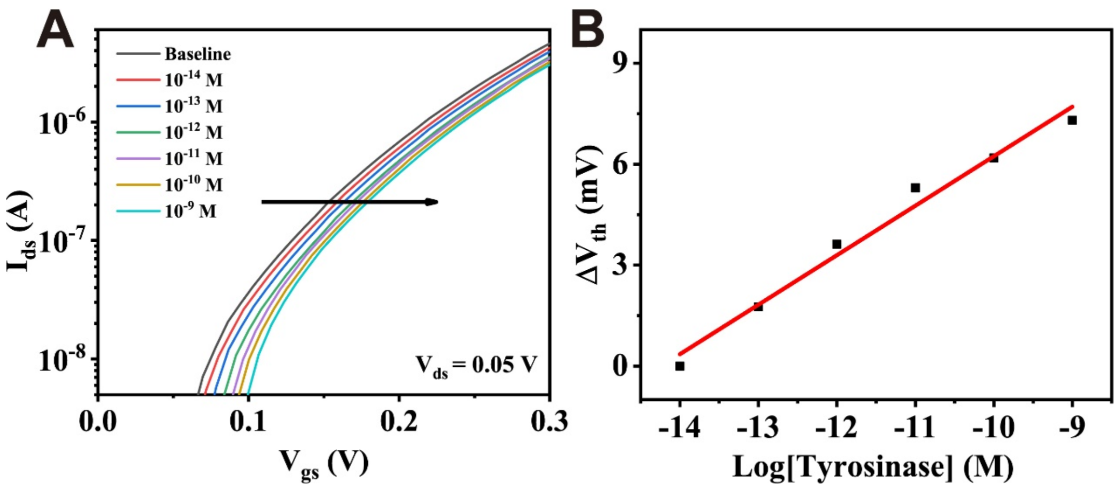

Figure S15. Sensitivity of WVFY peptide modified bio-FETs after one week storage in air. Related to Figure 4.

(A) Transfer characteristics for the WVFY modified device measured in various tyrosinase concentration (C_{TYR}): 0.1× PBS (black), 10^{-14} M (red), 10^{-13} M (deep blue), 10^{-12} M (green), 10^{-11} M (purple), 10^{-10} M (ginger), and 10^{-9} M (sky blue), when the bio-FET stored in air environment for one week. The V_{th} shifted positively with increased C_{TYR} and optimal detection range of 10 fM to 1 nM toward tyrosinase sensing. (B) A corresponding linear relationship between ΔV_{th} as a function of C_{TYR}.

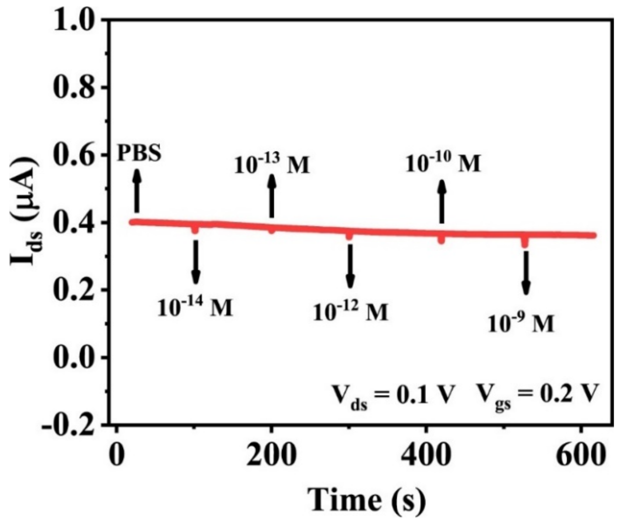

Figure S16. Real-time sensing response of Al_2O_3/In_2O_3 bio-FET without WVFY to tyrosinase in 0.1× PBS solution, showing no significant current fluctuations. Related to Figure 4.

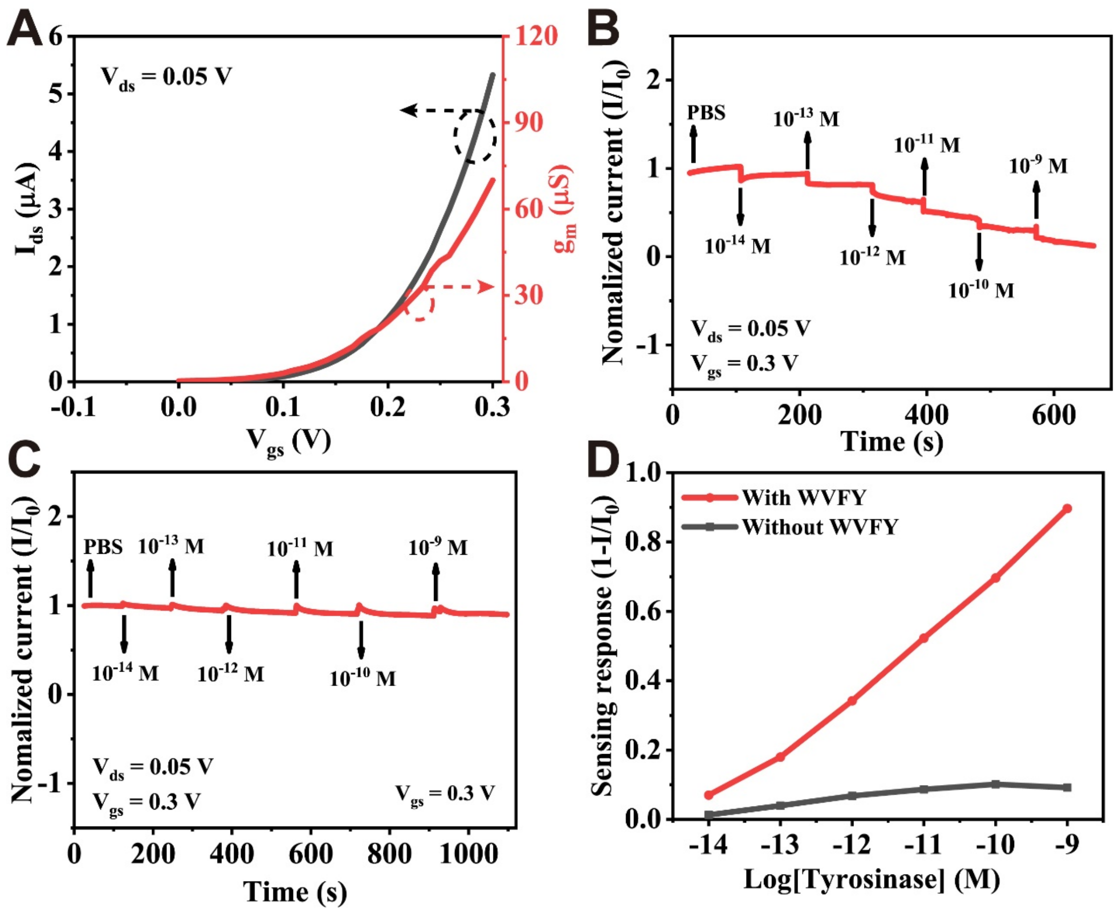

Figure S17. Detection of tyrosinase using nanostructured WVFY peptide modified bio-FETs. Related to Figure 4.

(A) Transfer curve and transconductance characteristics in linear scale when the V_{ds} was fixed at 0.05 V. (b-c) Real-time sensing response of Al_2O_3/In_2O_3 bio-FET with (B) and without(C) WVFY modification when exposed to various concentration of tyrosinase. The drain and gate voltage (V_{ds} and V_{gs}) were 0.05 and 0.3 V, respectively. (D) Sensing response (1 - I/I_0) as a function of tyrosinase concentration in logarithm scale.

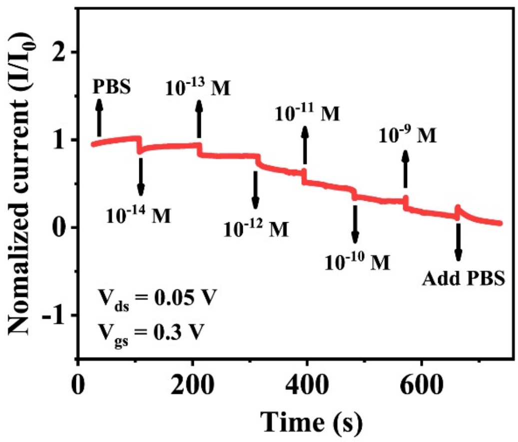

Figure S18. Real-time sensing response of WVFY modified bio-FET with tyrosinase. Related to Figure 4.

PBS solution was injected (at ~660 s) to dilute the tyrosinase concentration from 1×10^{-9} to 1×10^{-10} M. The figure is extended from Figure S17B.

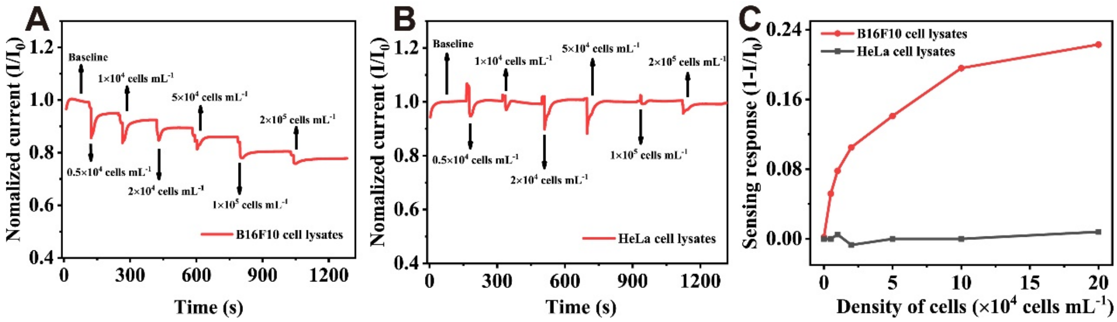

Figure S19. Tyrosinase screening in cell lysates using WVFY modified bio-FETs. Related to Figure 4.

Real-time sensing response curves obtained at bio-FET with (A) B16F10 cell lysates and (B) HeLa cell lysates. (C) Relationships between the sensing response (1 - I/I_0) and density of cells.

Table S1. Comparation of electrical characteristics of flexible bio-FETs before and after being transferred onto soft PDMS substrates. Related to Figure 1.

	V_{th} (V)	$I_{on/off}$	μ ($cm^2\ V^{-1}\ s^{-1}$)	SS ($mV\ dec^{-1}$)
Before transfer	0.362	9.5×10^1	0.865	70.6
On PDMS	0.365	8.8×10^1	0.839	73.1

Table S2. Comparation of performance between the presented bio-FET and other previous method on tyrosinase sensing. Related to Figure 4.

Method	Materials	Linear range	LOD	Ref.
Fluorescence	Resorufin/ *m*-tolylboronic acid pinacol ester	1 - 100 U mL^{-1}	0.5 U mL^{-1}	(Li et al., 2018)
Microneedle electrochemical sensor	Catechol/agarose	0 - 0.5 mg mL^{-1}	-	(Ciui et al., 2018)
SERS	ITO/AuNPs/p-TC	0.1 - 100 U mL^{-1}	0.07 U mL^{-1}	(Wang et al., 2019)
Colorimetry	KA/AgNPs	0.5 - 4 U mL^{-1}	0.117 U mL^{-1}	(Liu et al., 2017)
Electrocatalysis	$[(PSS/PPy)(P_2Mo_{18}/PPy)_5]$	3.66 - 26.87 U mL^{-1}	0.0021 U mL^{-1}	(Ding et al., 2021)
Fluorescence	Morpholine/4-aminophenol	0.5 - 60 U mL^{-1}	0.007 U mL^{-1}	(Zhou et al., 2016)
Spectrophotometry	Boronic acid/AuNPs	10^{-10} - 10^{-8} U mL^{-1}	10^{-10} U mL^{-1}	(Li et al., 2012)
Molecularly imprinted polymer-based sensor	Scopoletin/o-phenylenediamine	-	3.97 nM	(Yarman et al., 2018)
Photofuel cell-based self-powered sensor	$g\text{-}C_3N_4\text{-}Bi_2S_3$/ hemin-graphene	0.01 - 5 U mL^{-1}	0.005 U mL^{-1}	(Yan et al., 2019)
Field-Effect Transistors	$WVFY/Al_2O_3/In_2O_3$	10 fM - 1 nM (1.3 × 10^{-6} - 0.13 U mL^{-1})	1.9 fM (2.5 × 10^{-7} U mL^{-1})	This work