Supporting Information

Melamine derived N rich C entrapped Au nanoparticles for sensitive and selective monitoring of dopamine from blood samples

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S-1 Instruments

The electrochemical measurements were performed with a Gamry electrochemical workstation (Inreface-1010). A conventional three-electrode cell was used at room temperature (24℃), graphitic lead pencil electrode (GPE) as a working electrode, platinum wire as a counter electrode, and Ag/AgCl electrode as reference electrode, respectively. Electrochemical impedance spectroscopy (EIS) was performed in 5mM $[Fe(CN)_6]^{4-/3-}$ as a redox probe. The morphology of the modified electrode was examined using a transmission electron microscopy (TEM) system (F200×, The Thermo Scientific, Waltham, MA USA). Raman spectroscopy was performed using a EnSpectr R532 Raman spectrometer (Enhanced Spectrometry, Inc., Torrance, CA, USA).

S-2 Synthesis of benzimidazolium-1-acatate ionic liquid (IL)

Benzimidazolium-1-acetate ionic liquid (IL) was synthesized by conventional neutralization of benzimidazole with acetic acid as reported in literature [1]. Briefly, 15 mM of benzimidazole was dissolved into 10 mL of methanol in a round bottom flask under classical conditions and the solution was continuously stirred for 2h at 65°C. Next, 0.95 mL of acetic acid solution was added dropwise into the above prepared solution and kept under reflux overnight. Finally, the resulting light-yellow mixture was heated at 100°C in a rotary evaporator for 3h and upon cooling at room temperature, the IL product was made and used for analytical purposes.

S-3 NMR analyses of benzimidazolium-1-acetate ionic liquid (IL)

The purities of BAIL were verified by NMR.

^{1}H NMR: δ = 1.97 (s, 3H), 5.11 (s, 1H), 7.23 –7.25 (m, 2H), 7.58 –7.6 (m, 2H), 8.23 (s, 1H) [2](Figure S1).

^{13}C NMR: 21.5 (methyl peak), 115.7, 122.1, 138.4, 142.3, 172.5 (carbonyl carbon of acetate ion)[3] (Figure S2).

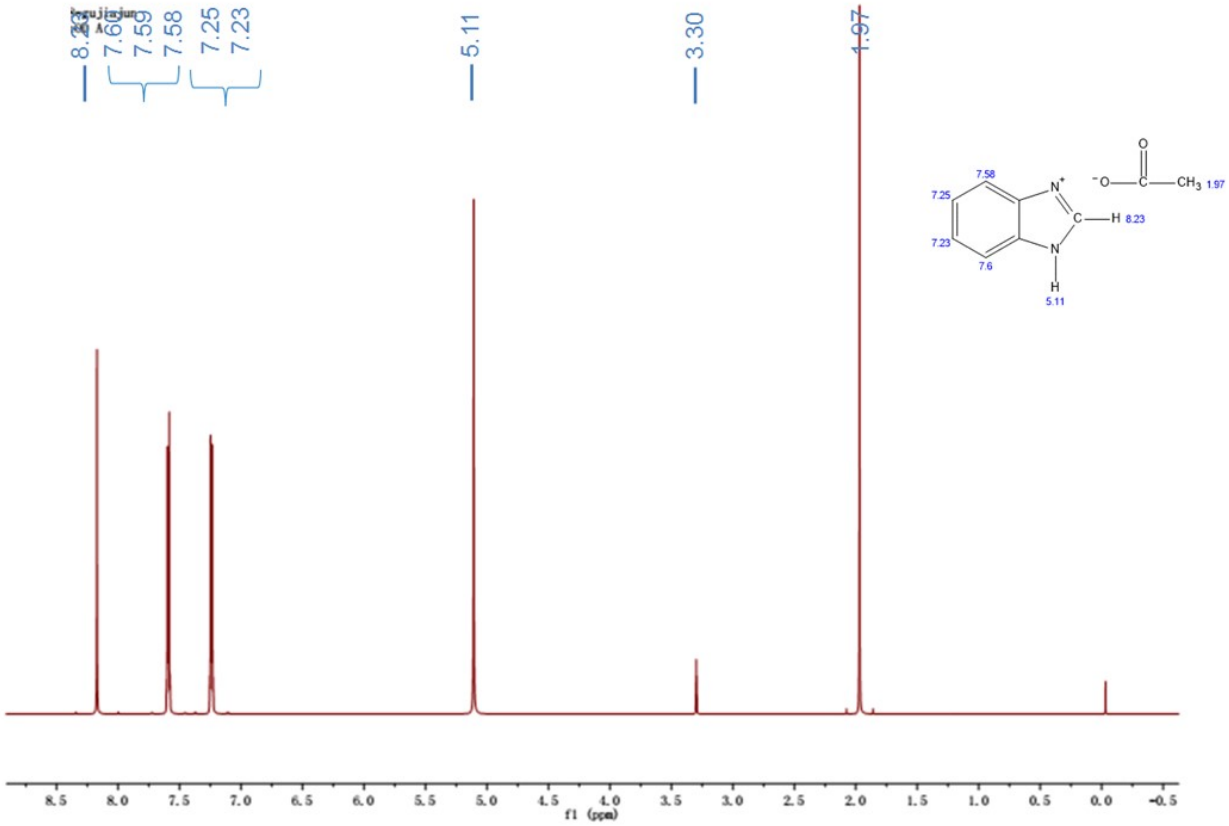


Figure S1. ^{1}H NMR of IL

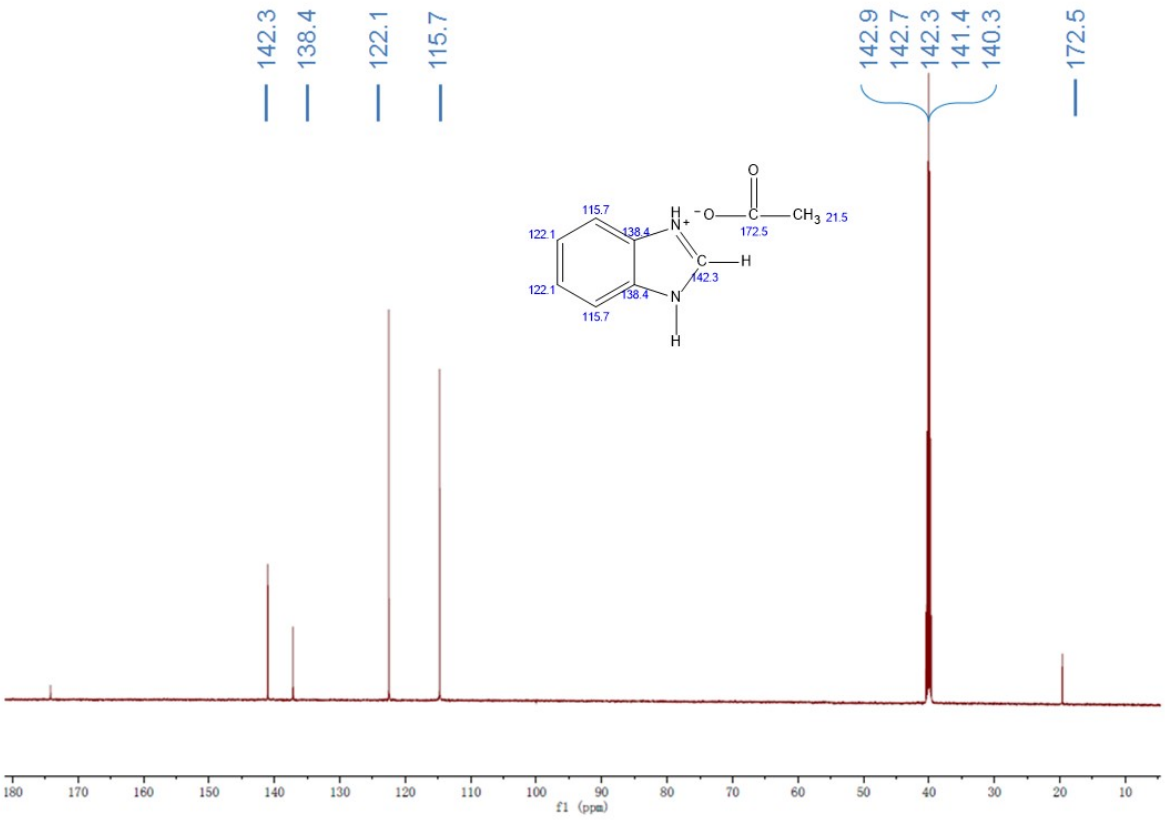


Figure S2. ^{13}C NMR of IL

S-4 Fourier transforms infrared (FTIR) spectroscopy analyses of IL

FTIR spectroscopy has been employed to analyze the chemical structure of as-synthesized IL. The band at 2981 cm $^{-1}$ is assigned to the alkyl group. The peak around 3112 cm^{-1} corresponds to alkenyl C—H stretching. The band at 1701 and 1619 cm^{-1} are due to the occurrence of C=O bonds and C=N, respectively. Also, the absence of stretching vibrations around 3000 cm^{-1} (O—H) verifies the presence of acetate ion.

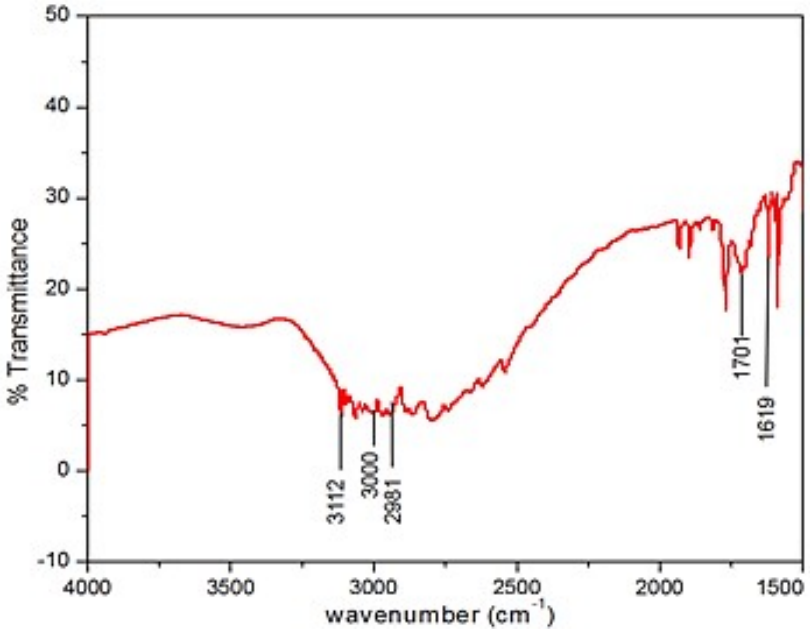


Figure S3 FTIR analysis of benzimidazolium-1-acetate ionic liquid (IL)

Scherer equation used to calculate the crystalline size

$$d = k.\lambda / \beta.Cos\theta$$

Where,

d = Crystallite size (nm)

β (the peak width of the diffraction peak or full width at the half maximum: FWHM)

Cosθ = (θ is the half of diffraction angle).

Scherer constant k = 0.9

λ = 0.15406 nm X-ray wavelength

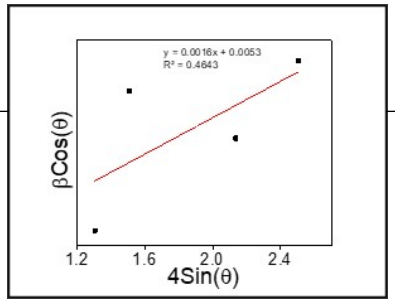
Williamson-Hull equation to calculate the strain

$$\beta\cos(\theta) = (k\lambda/d) + (\eta\backslash\sin(\theta))$$

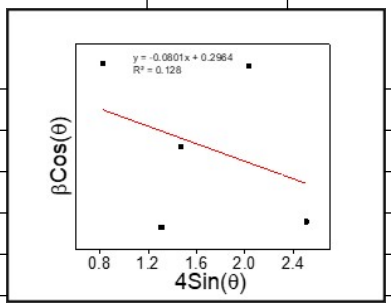
η= Strain factor

Table S1 Table showing the calculation for average crystalline size and strain.

Parameter		Peak position	FWHM		Sherer	X-axis	Y-axis	Intercepts	Slope	Micro-strain
K	Lambda (A)	2θ	β	Crystalline size (nm)	Average size (nm)	4 Sinθ	βCosθ	C=K A/D	m	*10³
Au-NPs										
0.9	0.154	38.1167	0.3945	21.29714	17.19782	1.306124	0.006508	0.0053	0.002	1.6
		44.3051	0.55006	15.58759		1.508296	0.008892			
		64.5425	0.54746	17.15554		2.135712	0.008079			
		77.5342	0.69046	14.75099		2.504625	0.009396			



NC@Au-NPs										
0.9	0.154	23.8106	18.4801	0.439163	3.202313	0.825178	0.3156	0.2964	-0.08	-80.1
		38.185	0.9401	8.938894		1.308376	0.015505			
		43.0662	10.0202	0.851982		1.468155	0.162679			
		60.9684	20.6313	0.44665		2.029203	0.31031			
		77.6111	1.91016	5.334874		2.506718	0.02598			



S-6

Tauc and Davis–Mott relationship

$$(\alpha h\nu)^{n} = A\,(h\nu - E_g)$$

where

α= absorption coefficient

hν =energy

Eg = band gap energy

A = constant

Absorption edge was observed for the transitions for n = 2

Table S2 Table showing the calculation for band gap.

Wavelength (nm)	Absorbance (a.u.)	Absorbance (a.u.)	Absorption coefficient (α) cm^{-1}	Absorption coefficient (α) cm^{-1}	Energy (eV)	$(\alpha h\nu)^2$	$(\alpha h\nu)^2$
698	0.029	0.025	0.066787	0.057575	1.776504	0.118647	0.102282
696	0.029	0.026	0.066787	0.059878	1.781609	0.118988	0.106679
694	0.029	0.026	0.066787	0.059878	1.786744	0.119331	0.106987
692	0.029	0.027	0.066787	0.062181	1.791908	0.119676	0.111423
690	0.029	0.028	0.066787	0.064484	1.797101	0.120023	0.115884
688	0.03	0.028	0.06909	0.064484	1.802326	0.124523	0.116221
686	0.03	0.029	0.06909	0.066787	1.80758	0.124886	0.120723
684	0.03	0.029	0.06909	0.066787	1.812865	0.125251	0.121076
682	0.03	0.03	0.06909	0.06909	1.818182	0.125618	0.125618
680	0.03	0.03	0.06909	0.06909	1.823529	0.125988	0.125988
678	0.031	0.031	0.071393	0.071393	1.828909	0.130571	0.130571
676	0.031	0.031	0.071393	0.071393	1.83432	0.130958	0.130958
674	0.031	0.032	0.071393	0.073696	1.839763	0.131346	0.135583
672	0.031	0.033	0.071393	0.075999	1.845238	0.131737	0.140236
670	0.032	0.034	0.073696	0.078302	1.850746	0.136393	0.144917
668	0.032	0.034	0.073696	0.078302	1.856287	0.136801	0.145351
666	0.032	0.035	0.073696	0.080605	1.861862	0.137212	0.150075

664	0.033	0.036	0.075999	0.082908	1.86747	0.141926	0.154828
662	0.033	0.037	0.075999	0.085211	1.873112	0.142355	0.15961
660	0.033	0.038	0.075999	0.087514	1.878788	0.142786	0.16442
658	0.034	0.039	0.078302	0.089817	1.884498	0.14756	0.16926
656	0.034	0.041	0.078302	0.094423	1.890244	0.14801	0.178483
654	0.035	0.042	0.080605	0.096726	1.896024	0.152829	0.183395
652	0.035	0.043	0.080605	0.099029	1.90184	0.153298	0.188337
650	0.036	0.045	0.082908	0.103635	1.907692	0.158163	0.197704
648	0.037	0.046	0.085211	0.105938	1.91358	0.163058	0.202721
646	0.037	0.048	0.085211	0.110544	1.919505	0.163563	0.21219
644	0.038	0.05	0.087514	0.11515	1.925466	0.168505	0.221717
642	0.038	0.051	0.087514	0.117453	1.931464	0.16903	0.226856
640	0.039	0.053	0.089817	0.122059	1.9375	0.17402	0.236489
638	0.04	0.055	0.09212	0.126665	1.943574	0.179042	0.246183
636	0.041	0.058	0.094423	0.133574	1.949686	0.184095	0.260427
634	0.042	0.06	0.096726	0.13818	1.955836	0.18918	0.270257
632	0.043	0.063	0.099029	0.145089	1.962025	0.194297	0.284668
630	0.044	0.066	0.101332	0.151998	1.968254	0.199447	0.299171
628	0.045	0.069	0.103635	0.158907	1.974522	0.20463	0.313765
626	0.046	0.072	0.105938	0.165816	1.980831	0.209845	0.328453
624	0.047	0.075	0.108241	0.172725	1.987179	0.215094	0.343236
622	0.048	0.078	0.110544	0.179634	1.993569	0.220377	0.358113
620	0.049	0.081	0.112847	0.186543	2	0.225694	0.373086
618	0.05	0.085	0.11515	0.195755	2.0064	0.2310	0.3927

					72	45	77
616	0.051	0.088	0.117453	0.202664	2.012987	0.236431	0.40796
614	0.052	0.092	0.119756	0.211876	2.019544	0.241853	0.427893
612	0.054	0.096	0.124362	0.221088	2.026144	0.251975	0.447956
610	0.055	0.1	0.126665	0.2303	2.032787	0.257483	0.468151
608	0.057	0.104	0.131271	0.239512	2.039474	0.267724	0.488478
606	0.058	0.109	0.133574	0.251027	2.046205	0.27332	0.513653
604	0.06	0.113	0.13818	0.260239	2.05298	0.283681	0.534265
602	0.061	0.118	0.140483	0.271754	2.059801	0.289367	0.559759
600	0.063	0.123	0.145089	0.283269	2.066667	0.299851	0.585423
598	0.065	0.129	0.149695	0.297087	2.073579	0.310404	0.616033
596	0.067	0.134	0.154301	0.308602	2.080537	0.321029	0.642058
594	0.069	0.14	0.158907	0.32242	2.087542	0.331725	0.673065
592	0.072	0.145	0.165816	0.333935	2.094595	0.347317	0.699458
590	0.074	0.151	0.170422	0.347753	2.101695	0.358175	0.730871
588	0.077	0.157	0.177331	0.361571	2.108844	0.373963	0.762497
586	0.08	0.163	0.18424	0.375389	2.116041	0.389859	0.794338
584	0.082	0.17	0.188846	0.39151	2.123288	0.400974	0.831288
582	0.085	0.176	0.195755	0.405328	2.130584	0.417073	0.863585
580	0.089	0.183	0.204967	0.421449	2.137931	0.438205	0.901029
578	0.092	0.189	0.211876	0.435267	2.145329	0.454544	0.933791
576	0.095	0.197	0.218785	0.453691	2.152778	0.470995	0.976696
574	0.099	0.204	0.227997	0.469812	2.160279	0.492537	1.014925
572	0.103	0.211	0.237209	0.485933	2.167832	0.514229	1.053421

570	0.107	0.219	0.246421	0.504357	2.175439	0.536074	1.097198
568	0.111	0.227	0.255633	0.522781	2.183099	0.558072	1.141282
566	0.116	0.234	0.267148	0.538902	2.190813	0.585271	1.180633
564	0.12	0.242	0.27636	0.557326	2.198582	0.6076	1.225327
562	0.125	0.25	0.287875	0.57575	2.206406	0.635169	1.270338
560	0.13	0.257	0.29939	0.591871	2.214286	0.662935	1.310572
558	0.135	0.265	0.310905	0.610295	2.222222	0.6909	1.356211
556	0.14	0.272	0.32242	0.626416	2.230216	0.719066	1.397043
554	0.146	0.28	0.336238	0.64484	2.238267	0.75259	1.443324
552	0.152	0.287	0.350056	0.660961	2.246377	0.786358	1.484767
550	0.158	0.294	0.363874	0.677082	2.254545	0.82037	1.526512
548	0.164	0.301	0.377692	0.693203	2.262774	0.854632	1.568562
546	0.17	0.307	0.39151	0.707021	2.271062	0.889144	1.605689
544	0.176	0.313	0.405328	0.720839	2.279412	0.923909	1.643089
542	0.182	0.318	0.419146	0.732354	2.287823	0.958932	1.675496
540	0.187	0.322	0.430661	0.741566	2.296296	0.988925	1.702855
538	0.192	0.326	0.442176	0.750778	2.304833	1.019142	1.730418
536	0.197	0.33	0.453691	0.75999	2.313433	1.049584	1.758186
534	0.201	0.332	0.462903	0.764596	2.322097	1.074906	1.775466
532	0.205	0.334	0.472115	0.769202	2.330827	1.100418	1.792877
530	0.208	0.334	0.479024	0.769202	2.339623	1.120735	1.799642
528	0.209	0.334	0.481327	0.769202	2.348485	1.130389	1.806459
526	0.211	0.333	0.485933	0.766899	2.357414	1.145545	1.807899
524	0.211	0.33	0.485933	0.75999	2.3664	1.1499	1.7984

					12	18	5
522	0.211	0.327	0.485933	0.753081	2.375479	1.154324	1.788928
520	0.209	0.323	0.481327	0.743869	2.384615	1.14778	1.773841
518	0.208	0.318	0.479024	0.732354	2.393822	1.146698	1.753125
516	0.205	0.313	0.472115	0.720839	2.403101	1.13454	1.732249
514	0.202	0.307	0.465206	0.707021	2.412451	1.122287	1.705654
512	0.199	0.3	0.458297	0.6909	2.421875	1.109938	1.673273
510	0.195	0.293	0.449085	0.674779	2.431373	1.091893	1.640639
508	0.191	0.286	0.439873	0.658658	2.440945	1.073706	1.607748
506	0.187	0.278	0.430661	0.640234	2.450593	1.055375	1.568953
504	0.182	0.27	0.419146	0.62181	2.460317	1.031232	1.52985
502	0.178	0.262	0.409934	0.603386	2.47012	1.012586	1.490436
500	0.173	0.255	0.398419	0.587265	2.48	0.988079	1.456417
498	0.169	0.247	0.389207	0.568841	2.48996	0.96911	1.416391
496	0.165	0.24	0.379995	0.55272	2.5	0.949988	1.3818
494	0.161	0.234	0.370783	0.538902	2.510121	0.93071	1.352709
492	0.158	0.228	0.363874	0.525084	2.520325	0.917081	1.323382
490	0.154	0.222	0.354662	0.511266	2.530612	0.897512	1.293816
488	0.151	0.217	0.347753	0.499751	2.540984	0.883635	1.269859
486	0.148	0.213	0.340844	0.490539	2.55144	0.869643	1.251581
484	0.146	0.209	0.336238	0.481327	2.561983	0.861436	1.233152
482	0.144	0.205	0.331632	0.472115	2.572614	0.853161	1.21457
480	0.142	0.202	0.327026	0.465206	2.583333	0.844817	1.201782
478	0.14	0.199	0.32242	0.458297	2.594142	0.836403	1.188888

476	0.139	0.197	0.320117	0.453691	2.605042	0.833918	1.181884
474	0.137	0.195	0.315511	0.449085	2.616034	0.825387	1.174822
472	0.136	0.193	0.313208	0.444479	2.627119	0.822835	1.167699
470	0.135	0.191	0.310905	0.439873	2.638298	0.82026	1.160516
468	0.134	0.19	0.308602	0.43757	2.649573	0.817663	1.159374
466	0.133	0.189	0.306299	0.435267	2.660944	0.815045	1.158221
464	0.133	0.188	0.306299	0.432964	2.672414	0.818558	1.157059
462	0.132	0.187	0.303996	0.430661	2.683983	0.8159 2	1.155887
460	0.132	0.186	0.303996	0.428358	2.695652	0.819467	1.154704
458	0.131	0.185	0.301693	0.426055	2.707424	0.816811	1.153511
456	0.131	0.185	0.301693	0.426055	2.719298	0.820393	1.158571
454	0.13	0.185	0.29939	0.426055	2.731278	0.817717	1.163674
452	0.13	0.184	0.29939	0.423752	2.743363	0.821335	1.162505
450	0.13	0.184	0.29939	0.423752	2.755556	0.824986	1.167672
448	0.13	0.184	0.29939	0.423752	2.767857	0.828669	1.172885
446	0.129	0.184	0.297087	0.423752	2.780269	0.825982	1.178145
444	0.129	0.184	0.297087	0.423752	2.792793	0.829702	1.183452
442	0.129	0.184	0.297087	0.423752	2.80543	0.833457	1.188807
440	0.129	0.184	0.297087	0.423752	2.818182	0.837245	1.19421
438	0.13	0.185	0.29939	0.426055	2.83105	0.847588	1.206183
436	0.13	0.185	0.29939	0.426055	2.844037	0.851476	1.211716
434	0.13	0.185	0.29939	0.426055	2.857143	0.8554	1.2173
432	0.13	0.185	0.29939	0.426055	2.87037	0.8593 6	1.222936
430	0.13	0.186	0.29939	0.428358	2.8837	0.8633	1.2352

					21	57	65
428	0.131	0.186	0.301693	0.428358	2.897196	0.874064	1.241037
426	0.131	0.187	0.301693	0.430661	2.910798	0.878167	1.253567
424	0.131	0.187	0.301693	0.430661	2.924528	0.88231	1.25948
422	0.132	0.188	0.303996	0.432964	2.938389	0.893258	1.272216
420	0.132	0.188	0.303996	0.432964	2.952381	0.897512	1.278275
418	0.132	0.189	0.303996	0.435267	2.966507	0.901806	1.291223
416	0.133	0.19	0.306299	0.43757	2.980769	0.913007	1.304295
414	0.133	0.19	0.306299	0.43757	2.995169	0.917417	1.310596
412	0.134	0.191	0.308602	0.439873	3.009709	0.928802	1.32389
410	0.134	0.192	0.308602	0.442176	3.02439	0.933333	1.337313
408	0.135	0.193	0.310905	0.444479	3.039216	0.944907	1.350868
406	0.135	0.193	0.310905	0.444479	3.054187	0.949562	1.357522
404	0.136	0.194	0.313208	0.446782	3.069307	0.961331	1.371311
402	0.137	0.195	0.315511	0.449085	3.084577	0.973218	1.385237
400	0.137	0.196	0.315511	0.451388	3.1	0.978084	1.399303

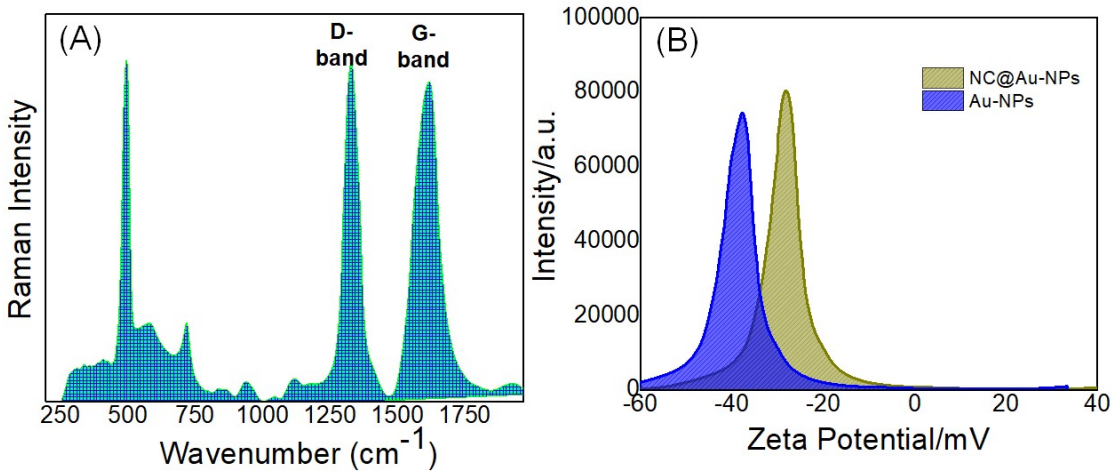


Figure S4 (A) Raman spectra of NC@Au-NPs. (B) Zeta potential of NC@Au-NPs and Au-NPs.

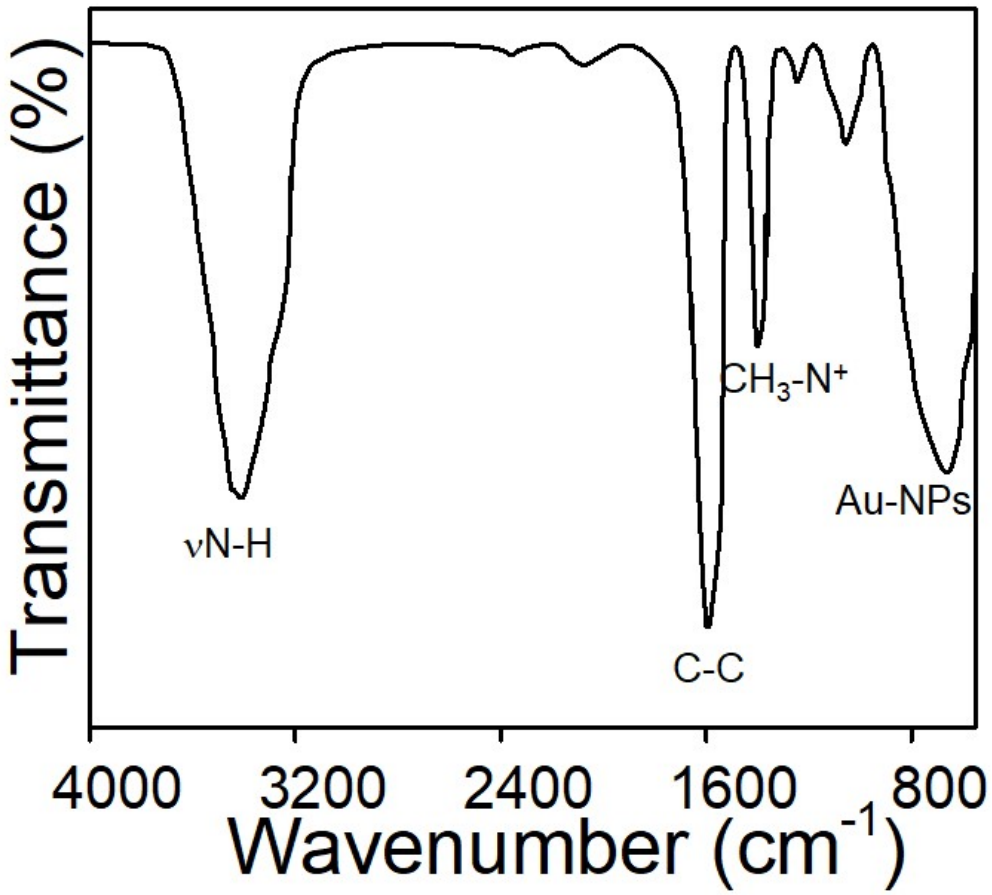


Figure S5 FTIR spectra of IL modified NC@Au-NPs.

Table S3 Full width at Half Maxima (FWHM) of (A) NC@Au-NPs and (B)Au-NPs.

(A) NC@Au-NPs

		Value	Standard Error
Book1_C	y0	0.10659	0.00641
	xc	528.60563	1.94352
	w	67.65688	4.75435
	A	18.95404	1.48874
	sigma	33.82844	2.37717
	FWHM	79.65989	5.59782
	Height	0.22353	0.01201

(B) Au-NPs

		Value	Standard Error
Book1_B	y0	0.07522	0.00414
	xc	520.5291	2.27891
	w	63.87246	5.45418
	A	9.96513	0.93388
	sigma	31.93623	2.72709
	FWHM	75.20407	6.42181
	Height	0.12448	0.00823

$$FWHM=0.5346wL+sqrt\ (0.2166*wL^2+wG^2)$$

whereas,

wL and wG (widths of the Lorentzian and Gaussian contributions) are given by the Origin software after the fitting.

Table S4 Table showing the peak current and peak potential difference of each electrode calculated from figure S2.

Electrode	***Anodic current***	***Ep***	***Ec***	***Peak Potential difference***
Au-NPS	0.096270791	0.445678228	0.026025712	0.419652517
NC@Au-NPs	0.2329252	0.430804696	0.022319052	0.408485644
Au-NPs/PDA	0.102208367	0.446608261	0.012255572	0.458863833

NC@Au-NPs/PDA	0.26041408	0.348232281	0.067018129	0.281214152
Au-NPs/PDA-IL	0.124572978	0.359583405	0.085441434	0.274141971
<u>NC@Au-NPs/PDA-IL</u>	0.32852887	0.310595934	0.102511091	0.208084843

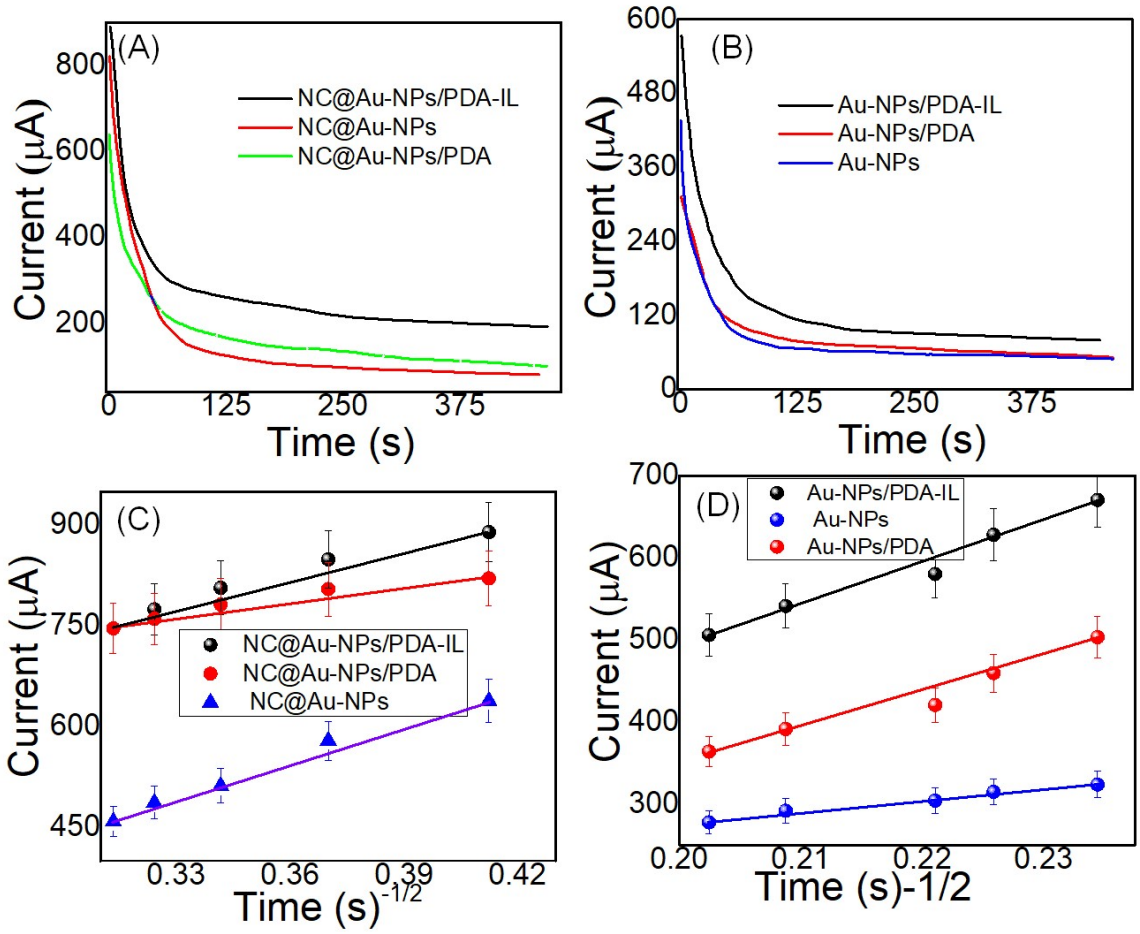


Figure S6. Chronoamperometric curve of (A) NC@Au-NPs, NC@Au-NPs/PDA, NC@Au-NPs/PDA-IL electrodes and (B) Au-NPs, Au-NPs/PDA, Au-NPs/PDA-IL in PBS against 1 mM of $[Fe(CN)_6]^{3-}$. Electrochemical impedance spectroscopy (EIS) of NC@Au-NPs (C) and Au (D) based electrodes by using 2mM of $[Fe(CN)_6]^{3-}$ as a redox probe. (C & D) Dependence of IC/IL on the $(time)^{1/2}$ derived from their respective chronoamperogram data.

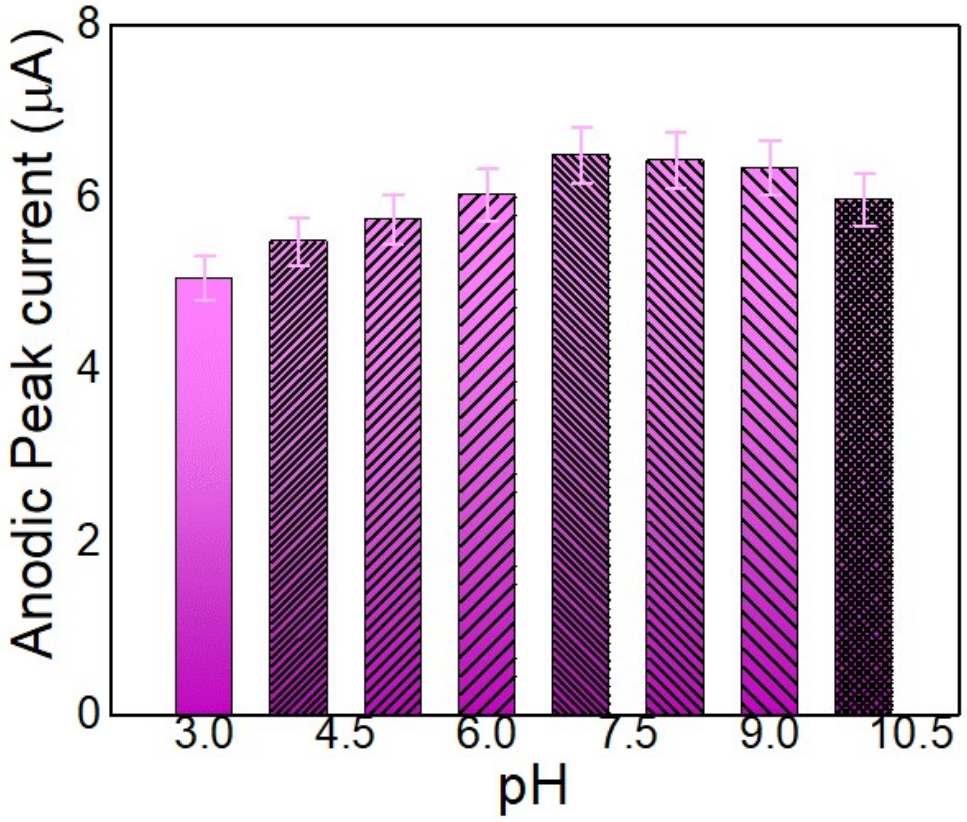


Figure S7. Optimization parameters pH at which the sensitive behavior of NC@Au-NPs/PDA/IL electrode was determined.

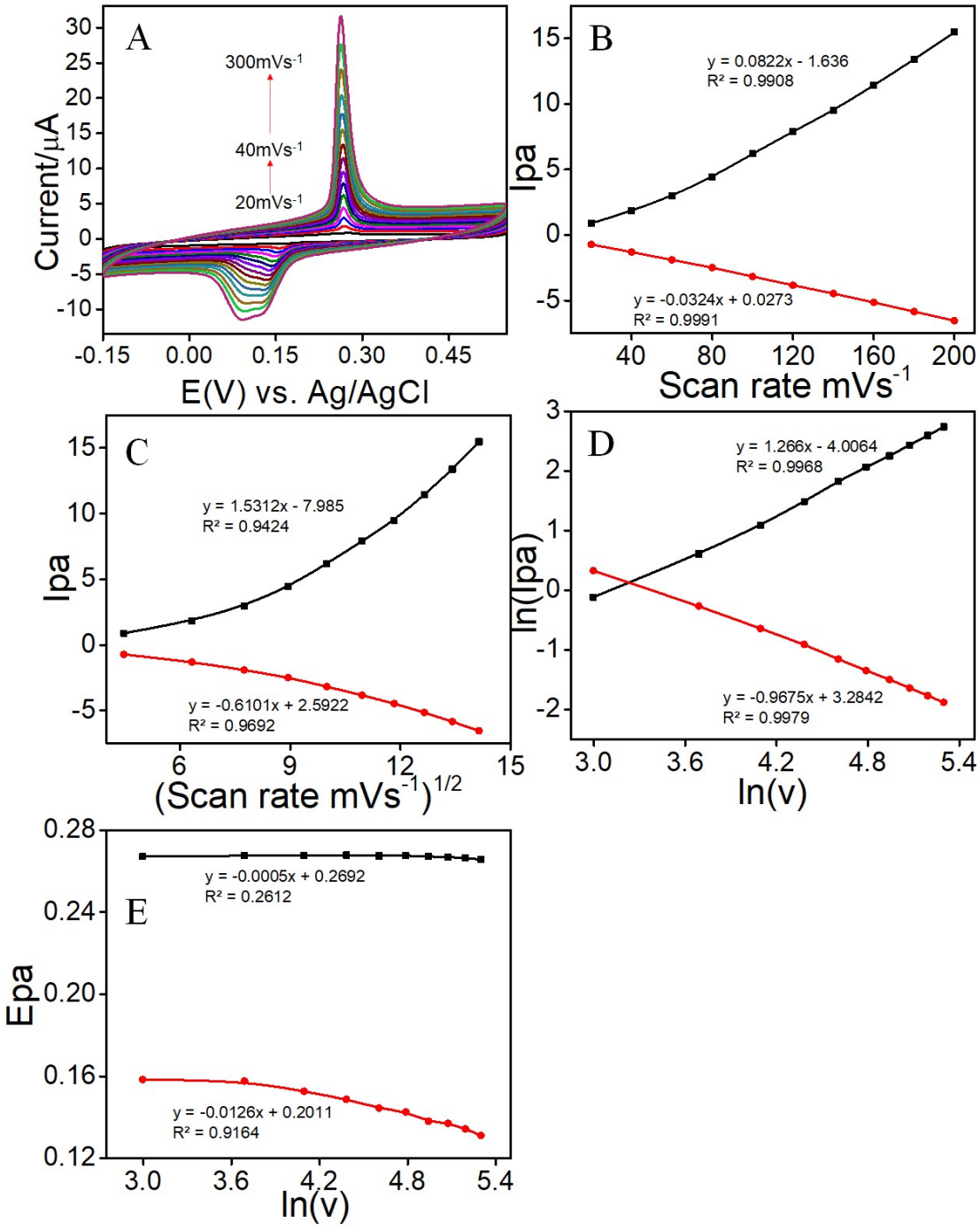


Figure S8. (A) cyclic voltammetry of Au-NPs/PDA-IL electrode at the scan rate ranging from 5 to 200 mV/s in the presence of 5 µM DA in phosphate buffer (pH 7.0) and its corresponding calibration plots (B-C) scan rate (mV/s) vs Current(µA) and square root scan rate (mV/s) vs Current(µA), respectively and (D-

E) log of scan rate (mV/s) vs oxidizing potential(mV) and log of scan rate (mV/s) vs log of Current(μA), respectively.

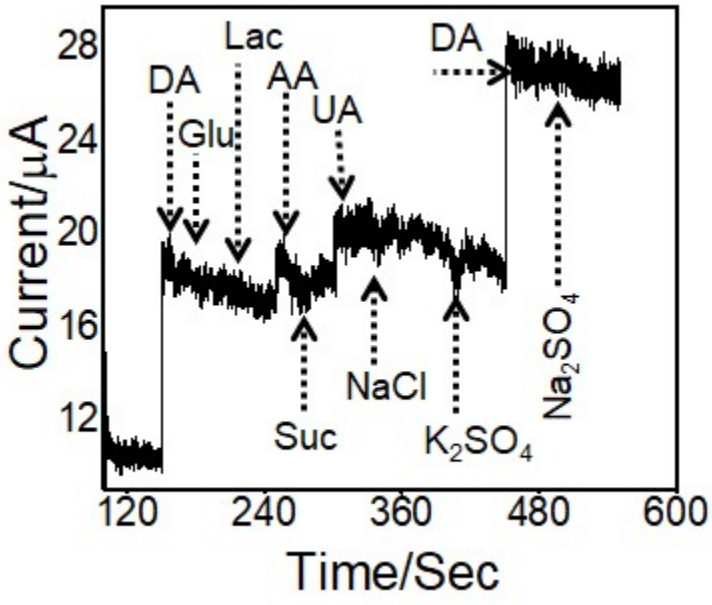


Figure S9: Selective amperometric response of the NC@Au-NPs/PDA/IL electrode exposed towards simultaneous monitoring of DA and co-existing electroactive species such as organic (DA, AA, UA, & Lac) and inorganic (KCl, NaCl, Na_2SO_4, & K_2SO_4) species in 0.1M PBS (pH;7) electrolyte.

Table S5 comparison of the electrochemical determination of NC@Au-NPs/PDA/IL modified electrode towards DA detection with the reported data.

Sr. #	Modified Electrode	Applied Potential (V)	Detection Limit (μM)	Linear Range (μM)	Ref.
1	RGO/Au nanoplate	-0.2–6	1.4	6.8–41	S[4]
2	RGO-AuNPs-CSHMs	-0.2–8	0.3	1–200	S[5]
3	AgNPs/rGO	-1–1	5.4	10–800	S[6]
4	RGO/AuNPs	-1.5–1	0.137	0.14–700	S[7]
5	PANI-AuNPs	-0.4–1.4	16	0.270–0.361	S[8]
6	Au NPs/N-doped CN	-0.3–0.4	0.007	0.02–700	S[9]
7	NC@Au-NPs/PDA/IL	-0.6-0.6	0.002	0.05-1	This work

Table S6. Determination of DA in DA hydrochloride injection using NC@Au-NPs/PDA/IL electrode.

DA sample	Added (μM)	Found [a] (μM)	Recovery (%)	RSD [b] (%)
1	0	1.78	-	-
2	2	3.75	87.7	3.7
3	2	4.7	92.8	3.65

1, 2 and 3 diluted DA hydrochloride injection sample, [a] Standard deviation, [b] Relative standard deviation (n = 3).

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