Electronic Supporting Information

Unveiling of Smartphone mediated ‘ratiometric’ ChromoSensor towards nanomolar level detection of lethal CN⁻: A combined experimental and theoretical validation with proposition of molecular Logic Circuitry

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Table S1. Crystal data of DNMH

| Crystal Data                  |                  |
|-------------------------------|------------------|
| Formula                       | C₁₇ H₁₂ N₄ O₄   |
| Formula Weight                | 336.31           |
| Crystal System                | monoclinic       |
| Space group                   | P2₁/n (No. 14)   |
| a, b, c [Angstrom]            | 6.787(2) / 12.178(5) / 17.714(7) |
| alpha, beta, gamma [deg]      | 90 / 98.997(11) / 90 |
| Volume                        | 1446.1(9) Å³    |
| Z                             | 4                |
| D(calc) [g/cm³]               | 1.545            |
| Mu(MoKa) [/mm]                | 0.114            |
| F(000)                        | 696              |
| Crystal Size [mm]             | 0.12 x 0.14 x 0.18 |

| Data Collection               |                  |
| Temperature (K)               | 120              |
| Radiation [Angstrom]          | MoKa 0.71073     |
| Theta Min-Max [Deg]           | 2.3, 25.0        |
| Dataset                       | -8: 7; -14: 14; -21: 21 |
| Tot., Uniq. Data, R(int)      | 10216, 2539, 0.105 |
| Observed Data [I > 2.0 sigma(I)] | 1613          |

| Refinement                    |                  |
| Nref, Npar                    | 2539, 226        |
| R, wR2, S                     | 0.0945, 0.2092, 1.14 |
| w = √(FO² + (0.0364P)² + 4.8771P) WHERE P = (FO² + 2FC²) / 3 | |
| Max. and Av. Shift/Error      | 0.00, 0.00       |
| Min. and Max. Resd. Dens. [e/Ang³] | -0.34, 0.35 |
**Table S2. Bond Distances (Angstrom) of DNMH**

| Bond Distances (Angstrom) |
|---------------------------|
| O1   -N2       | 1.222(6) | C00F   -C00H       | 1.479(8) |
| O001 -N1   | 1.241(5) | C00G   -C00I       | 1.360(7) |
| O002 -N1   | 1.225(6) | C00J   -C00O       | 1.419(8) |
| O004 -N2   | 1.240(7) | C00J   -C00L       | 1.358(7) |
| N1   -C009   | 1.455(7) | C00K   -C00N       | 1.425(7) |
| N005 -N007 | 1.379(6) | C00K   -C00P       | 1.426(7) |
| N005 -C00A | 1.352(6) | C00M   -C00N       | 1.392(7) |
| N2   -C00D   | 1.443(6) | C00O   -C00P       | 1.353(8) |
| N007 -C00H | 1.273(6) | C00B   -H00B       | 0.9500   |
| C009   -C00A | 1.430(7) | C00C   -H00C       | 0.9500   |
| C009   -C00B | 1.376(7) | C00G   -H00G       | 0.9500   |
| N005 -H005 | 0.8800   | C00H   -H00H       | 0.9500   |
| C00A   -C00I | 1.410(7) | C00I   -H00I       | 0.9500   |
| C00B   -C00D | 1.378(8) | C00J   -H00J       | 0.9500   |
| C00C   -C00M | 1.402(7) | C00L   -H00L       | 0.9500   |
| C00C   -C00F | 1.364(7) | C00M   -H00M       | 0.9500   |
| C00D   -C00G | 1.387(8) | C00N   -H00N       | 0.9500   |
| C00E   -C00F | 1.422(7) | C00O   -H00O       | 0.9500   |
| C00E   -C00L | 1.443(8) | C00P   -H00P       | 0.9500   |
| C00E   -C00K | 1.420(7) |                     |          |

**Table S3. Bond Angles (Degrees) of DNMH**

| Bond Angles (Degrees) |
|-----------------------|
| O001 -N1   -O002   | 123.2(4) | N007 -C00H   -C00F   | 119.1(5) |
| O001 -N1   -C009   | 118.8(4) | C00A   -C00I   -C00G   | 122.7(5) |
| O002 -N1   -C009   | 118.1(4) | C00L   -C00J   -C00O   | 120.4(5) |
| N007 -N005  -C00A   | 117.8(4) | C00E   -C00K   -C00N   | 119.5(5) |
| O1   -N2   -O004   | 122.9(4) | C00N   -C00K   -C00P   | 120.2(5) |
| O1   -N2   -C00D   | 117.8(5) | C00E   -C00K   -C00P   | 120.3(5) |
| O004 -N2   -C00D   | 119.3(4) | C00E   -C00L   -C00J   | 121.8(5) |
| N005 -N007  -C00H   | 115.7(4) | C00C   -C00M   -C00N   | 118.8(5) |
| N1   -C009   -C00A   | 121.9(5) | C00K   -C00N   -C00M   | 120.1(4) |
| N1   -C009   -C00B   | 116.7(4) | C00J   -C00O   -C00P   | 120.2(5) |
| Bond Angles (Degrees) (continued) of DNMH |
|----------------------------------------|
| C00A -C009 -C00B  | 121.4(5) | C00K -C00P -C00O  | 120.8(5) |
| N007 -N005 -H005  | 121.00   | C009 -C00B -H00B  | 120.00   |
| C00A -N005 -H005  | 121.00   | C00D -C00B -H00B  | 120.00   |
| N005 -C00A -C009  | 124.0(5) | C00F -C00C -H00C  | 118.00   |
| N005 -C00A -C00I  | 121.00   | C00D -C00G -H00G  | 120.00   |
| C009 -C00A -C00I  | 115.5(5) | C00D -C00G -H00G  | 120.00   |
| C009 -C00B -C00D  | 120.3(5) | C00I -C00G -H00G  | 120.00   |
| C00F -C00C -C00M  | 123.1(5) | N007 -C00H -H00H  | 120.00   |
| N2 -C00D -C00G    | 120.3(5) | C00F -C00H -H00H  | 120.00   |
| C00B -C00D -C00G  | 120.0(5) | C00A -C00I -H00I  | 119.00   |
| N2 -C00D -C00B    | 119.7(5) | C00G -C00I -H00I  | 119.00   |
| C00K -C00E -C00L  | 116.6(5) | C00L -C00J -H00J  | 120.00   |
| C00F -C00E -C00K  | 119.4(5) | C00O -C00J -H00J  | 120.00   |
| C00F -C00E -C00L  | 123.9(5) | C00E -C00L -H00L  | 119.00   |
| C00E -C00F -C00H  | 120.8(5) | C00J -C00L -H00L  | 119.00   |
| C00C -C00F -C00H  | 120.2(5) | C00C -C00M -H00M  | 121.00   |
| C00C -C00F -C00E  | 119.0(5) | C00N -C00M -H00M  | 121.00   |
| C00D -C00G -C00I  | 120.1(5) | C00K -C00N -H00N  | 120.00   |

Table S4. Bond Angles (Degrees) (continued) of DNMH

| Hydrogen bonding in DNMH (Angstrom, Deg) |
|----------------------------------------|
| N005  --  H005  ..  O001  | 0.8800  | 2.0200  | 2.636(5)  | 126.00   |
| C00B  --  H00B  ..  O002  | 0.9500  | 2.3300  | 2.656(6)  | 100.00   |
| C00C  --  H00C  ..  N007  | 0.9500  | 2.4500  | 2.785(7)  | 100.00   |
| C00H  --  H00H  ..  O004  | 0.9500  | 2.4700  | 3.223(7)  | 136.00   |
| C00I  --  H00I  ..  N007  | 0.9500  | 2.3600  | 2.709(6)  | 101.00   |
| C00I  --  H00I  ..  O004  | 0.9500  | 2.5800  | 3.314(7)  | 134.00   |

Table S5. Hydrogen bonding in DNMH (Angstrom, Deg)

Solvatochromic behaviour of DNMH

Solvatochromism is generally induced by changing the environment of the solvent in which the solute undergoes dissolution owing to non-specific interactions including polarity, dielectric constant, dipole moment, polarity, π-conjugation, acid-base chemistry, charge transfer and specific interactions like hydrogen bonding. In this case it is vital to interpret how actually hydrogen bonding, polarity and polarizability of the solvent affect the absorption maxima. Therefore, the electronic spectra of DNMH have been recorded in a series of solvents of varying polarity: Acetonitrile, Dichloromethane, Hexane, Methanol, Tetrahydrofuran, Dimethylsulfoxide, Toluene, Xylene, Benzene, Chloroform, Dioxane, Ethanol, Isopropanol and Water (Fig. S5). DNMH displayed absorption maxima around 390 nm for ACN,
DCM, CHCl$_3$, MeOH and THF whereas bathochromic shift for polar solvents like DMSO by ~20nm and hypsochromic shift for non-polar solvents like Hexane by ~10 nm was observed. Hexane, having the least polarity (0.009), i.e., a non-polar solvent exhibited a blue shift in the absorption spectra. On the contrary although DMSO has comparable polarity with acetonitrile (DMSO: 0.444 and ACN: 0.46) and much lesser than MeOH (0.762), EtOH (0.654), 2-propanol (0.546) yet due to its appreciably high dielectric constant (46.7) in comparison to the highly polar solvents; MeOH (32.7), EtOH (24.55), Isopropanol (19.92) and high hydrogen bond acceptance value altogether causes DNMH to exhibit positive solvatochromism in presence of DMSO solvent. In general, the shifting in polar solvents may be attributed to the alteration in charge density of the solute (DNMH) occurring on optical excitation; whereas in nonpolar solvents devoid of permanent dipole moment, their electron clouds are polarized by the charge density of the solute (DNMH) along with a feeble degree of inductive interaction and dispersive solute-solvent interaction, which results in the spectral shift.

Colorimetric response and optical performance of the chemoreceptor DNMH with CN$^-$ has been investigated in organo-aqueous mixture of varying stoichiometry. This has been purposefully performed to validate the best solvent mixture ratio wherein chromogenic detection of CN$^-$ is the most promising.

In this consequence, the colorimetric response of DNMH has been studied in presence of CN$^-$ in various solvent mixtures; H$_2$O, MeCN: H$_2$O (1:1 v/v), MeCN: H$_2$O (2:1 v/v), MeCN: H$_2$O (3:1 v/v), MeCN: H$_2$O (4:1 v/v) keeping the total volume of (sensor + analyte) constant (Fig. S6†). The outcome of “naked eye” response clearly demonstrated that the chromogenic change of DNMH in existence of CN$^-$ even in purely aqueous medium was unambiguously obvious. Nevertheless, in MeCN: H$_2$O (4:1, v/v), the detection of CN$^-$ is the most promising which is noticeable from the appearance of red coloration due to strong adduct formation. The UV-Vis spectral response further supported the choice of MeCN: H$_2$O=4:1 (v/v) to be the optimal solvent concentration owing to the maximum change in absorbance of DNMH with CN$^-$ in this solvent mixture (Fig. S7†).

**Fig. S5** Spectrophotometric response of DNMH in varying solvents Acetonitrile, Dichloromethane, Hexane, Methanol, Tetrahydrofuran, Dimethylsulfoxide, Toluene, Xylene, Benzene, Chloroform, Dioxane, Ethanol, Isopropanol and Water.

**Fig. S6** Colorimetric changes of DNMH with CN$^-$ in varying proportion of CH$_3$CN and H$_2$O; (a) DNMH (1×10$^{-5}$M, ACN), (b) DNMH + CN$^-$ (Water), (c) DNMH + CN$^-$ (MeCN: Water= 1:1(v/v)), (d) DNMH + CN$^-$ (MeCN : Water= 2:1(v/v)), (e) DNMH + CN$^-$ (MeCN : Water= 3:1(v/v)), (f) DNMH + CN$^-$ (MeCN : Water= 4:1(v/v)).
**Fig. S7** UV-Vis absorption changes of DNMH with gradual addition of CN\(^-\) in varying solvent mixture; H\(_2\)O, MeCN: H\(_2\)O (1:1 v/v), MeCN: H\(_2\)O (2:1 v/v), MeCN: H\(_2\)O (3:1 v/v), MeCN: H\(_2\)O (4:1 v/v).

**Fig. S8** UV-Vis spectra of DNMH (2×10\(^{-5}\)M) in acetonitrile.

**Jobs Plot of DNMH with CN\(^-\)**

The stoichiometric ratio of the chemosensor DNMH with CN\(^-\) successive solutions comprising of 10\(^{-4}\) M NBu\(_4\)CN and DNMH were prepared in acetonitrile solvent in such a way that the total concentration of the resulting solution remains constant. The mole fraction of the added analyte CN\(^-\) was varied from 0.1 to 0.9. The absorbance of the chemosensor DNMH at 510 nm has been plotted against the mole fraction of the added analyte. From the Jobs Plot analysis, it clearly affirms a 1:1 type host: guest complexation.

**Fig. S9** Jobs plot of DNMH with CN.
**Benesi-Hildebrand (B-H) Equation and Plot:**

The association constant of adduct formed during interaction between the chemosensor (host) and incoming targeted analyte (guest) can be determined by the following complex equilibrium.

\[
L + mX^\text{n-} \rightleftharpoons (X_mL)^{mn-}
\]

Hence the association constant can be determined as,

\[
K_a = \frac{[\{(X_mL)^{mn-}\}]}{[L][X^{n-}]^m}
\]

Where \([X^\text{n-}],[L]\) and \([(X_mL)^{mn-}]\) are the concentration of the guest analyte, chemosensor and host-guest adduct respectively.

For 1:1 stoichiometry \((m=1)\), linear Benesi-Hildebrand equation can be expressed in terms of optical density \((A)\) as follows

\[
A = \frac{A_0 + A_1 K[X^{n-}]}{1 + K[X^{n-}]} - \frac{1}{A_0}
\]

\[
\frac{1}{A - A_0} = \frac{1}{A_{\text{max}} - A_0} \frac{[\text{guest}]}{K_a} - \frac{1}{A_{\text{max}} - A_0}
\]

Where \(A_0\) and \(A\) indicate the optical density or absorbance at a particular wavelength of DNMH in the absence of the guest analyte \((CN^-)\) and presence of guest analyte in each successive step respectively and \(A_{\text{max}}\) represents the maximum absorbance, *i.e.* the saturated absorbance of the chemosensor molecule in presence of excess analyte; \([\text{guest}]\) is the concentration of guest ion added and \(K_a\) is the complex association constant. Where \([X^{n-}],[L]\) and \([(X_mL)^{mn-}]\) are the concentration of the added targeted analyte, chemosensor and the complexation between the analyte and the chemosensor respectively. \(A_0\), \(A\) and \(A_1\) prior to the addition of the analyte, absorbance after adding the analyte at every successive step and finally excess amount of the added analyte, respectively. The binding constant or association constant \(K_a\) \((\text{M}^{-1})\) is determined from the ratio of slope and intercept of Benesi-Hildebrand plot of optical density. As depicted in the following In the Benesi-Hildebrand (B-H) plot of \(1/[A-A_0]\) vs \(1/[CN^-]\) for the titration of the chemosensor DNMH and CN\(^-\), provides a straight line (best fitted), indicating a 1:1 type complex formation. The association constant \(K_a\) for CN\(^-\) is \(4.48 \times 10^6 \text{ M}^{-1}\)

![Benesi-Hildebrand plot for absorbance of CN\(^-\) with DNMH for the association constant determination.](image-url)
Limit of detection

The detection limit (DL) has been calculated following UV-Vis titration. The absorbance spectrum of DNMH was repeated for 10 times, and its standard deviation is measured. The limit of detection (LOD) is calculated from the following formula: $DL = 3\sigma/k$. $\sigma$ is the standard deviation of the blank solution of DNMH. Gradual emergence of new absorbance values during colorimetric titration with targeted analytes is plotted against its concentration. The (k) is derived from the slope of these plots.

Fig. S11 Change of absorbance at 510 nm as the linear function of the concentration of CN$^-$ for the calculation of LOD of DNMH towards CN$^-$.  

Fig. S12 Pseudo second order reaction kinetics of DNMH with CN$^-$.  

Time evolution and Kinetic study of DNMH in existence of CN$^-$. In order to determine the practical applicability of DNMH towards target specific analyte detection, photo stability of the chemosensor in presence of guest analyte (CN$^-$ herein) is significant. The time dependant host: guest interaction has been recorded by taking the UV-Vis spectroscopic data upon addition of 5 equivalents of CN$^-$ in ACN: H$_2$O (4:1, v/v) into DNMH (2×10$^{-5}$M, ACN) solution. A very feeble change in the absorption spectra was observed with increasing time which implicates high photostability of the resultant adduct. From the kinetics profile it was observed that the faster reaction rate of DNMH towards CN$^-$ displays a pivotal role underlying the practical applicability of the chemosensor.
Fig. S13
Time
dependant spectral response of DNMH \((2\times10^{-5}\text{M}, \text{ACN})\) in existence of 5 equivalent of CN\(^{-}\) (ACN: water, 4:1 v/v) \((a)\) plot of Absorbance vs wavelength; \((b)\) plot of absorbance vs time (h).

Electrochemical properties of DNMH with CN\(^{-}\) have been investigated by using cyclic voltammetry. During this study TBAPF\(_6\) as supporting electrolyte, glassy carbon as working electrode, Ag/AgCl electrode as reference electrode and platinum wire as the counter electrode has been used.

![Cyclic voltammogram of DNMH in acetonitrile.](image1)

**Fig. S14** Cyclic voltammogram of DNMH in acetonitrile.

![Electrochemical response of DNMH in ACN with CN\(^{-}\) \((10^{-3}\text{M}, \text{ACN})\) in existence of TBAPF\(_6\) as supporting electrolyte; scan rate 0.05 Vs\(^{-1}\).](image2)

**Fig. S15** Electrochemical response of DNMH in ACN with CN\(^{-}\) \((10^{-3}\text{M}, \text{ACN})\) in existence of TBAPF\(_6\) as supporting electrolyte; scan rate 0.05 Vs\(^{-1}\).

**Table S6.** Redox potential of DNMH with CN\(^{-}\)
The [DNMH•••CN⁻] ensemble was utilized to investigate its reversibility in presence of any cation. In this context, the sensing experiment was performed by adding 200μL solutions of different cations (10⁻⁶M) to 500μL solution of the [DNMH•••CN⁻] adduct. It was observed that the reddish orange color of the ensemble changed to light yellow in the presence of only Cd²⁺, whereas for the other cations, the color of the adduct solution remained unperturbed (Fig. S17†). This indicates the profound selective nature of the ensemble towards Cd²⁺ only and therefore, this divalent ion was chosen to establish the reversibility process of DNMH+CN⁻ ensemble.

**Fig. S17** Colorimetric response of DNMH•••CN⁻ ensemble in presence of varying cations.

**Fig. S16** Isothermal calorimetric titration of DNMH with CN⁻ in ACN at 298 K.

The [DNMH•••CN⁻] ensemble was utilized to investigate its reversibility in presence of any cation. In this context, the sensing experiment was performed by adding 200μL solutions of different cations (10⁻⁶M) to 500μL solution of the [DNMH•••CN⁻] adduct. It was observed that the reddish orange color of the ensemble changed to light yellow in the presence of only Cd²⁺, whereas for the other cations, the color of the adduct solution remained unperturbed (Fig. S17†). This indicates the profound selective nature of the ensemble towards Cd²⁺ only and therefore, this divalent ion was chosen to establish the reversibility process of DNMH+CN⁻ ensemble.

**Fig. S17** Colorimetric response of DNMH•••CN⁻ ensemble in presence of varying cations.

**Proposition of electronic circuit based on different logic gate:**
**Fig. S18** Proposition of logic gate by changing the inputs AND-NOT-XNOR-NAND-OR logic functions.

**Table S7.** Truth table for molecular logic operation.

| In1 (S2) | In2 (CN) | In3 (Cd²⁺) | OUT Y1 (390 nm) | OUT Y2 (510 nm) |
|----------|----------|------------|----------------|-----------------|
| 0        | 0        | 0          | 0 (Low)        | 0 (Low)         |
| 0        | 1        | 0          | 0 (Low)        | 0 (Low)         |
| 0        | 0        | 1          | 0 (Low)        | 0 (Low)         |
| 1        | 0        | 0          | 1 (High)       | 0 (Low)         |
| 1        | 0        | 1          | 1 (High)       | 0 (Low)         |
| 1        | 1        | 0          | 0 (Low)        | 1 (High)        |
| 0        | 1        | 1          | 0 (Low)        | 0 (Low)         |

**Dynamic Light Scattering (DLS) Analysis**
DNMH in ACN medium acquires a partial aggregated form due to weak intermolecular hydrogen bonding between the 2° amine centre and imine centre of DNMH with the solvent molecules which causes the average particle size to be 75 nm as observed from DLS analysis. However, after CN\(^-\) addition, the self-aggregation of DNMH molecule is disrupted by minimizing the solvation effect. Consequently, the population density of DNMH in presence of CN\(^-\) exhibited two peaks with reduced size distribution <10 nm (Fig. S16\textsuperscript{†}) due to non-covalent weak hydrogen bonding interactions between the antenna centres (-NH-, -CH=N-) of DNMH with the incoming CN\(^-\).\textsuperscript{3}

![Fig. S19 DLS analysis of a) DNMH (1×10\(^{-4}\)M, CH\(_3\)CN), b) DNMH + CN\(^-\) in CH\(_3\)CN: H\(_2\)O (4:1).](image)

**FT-IR study**

The FT-IR spectroscopy of DNMH has been carried out by using the KBr disks. The 3400 cm\(^{-1}\) and 1618 cm\(^{-1}\) peak suggests the presence of secondary amine and aldimine bond in DNMH. Upon addition of CN\(^-\) the electron density on the -NH bond drifted towards -N atom of -CN owing to the intermolecular H-bonding interaction (\(-\text{NH}•••\text{NC}\)) and furthermore the lone pairs on the N-atom undergo delocalisation. Therefore, the NH bond becomes weak and broadened in nature. Additionally, the peak at 1618 cm\(^{-1}\) also swung to the lower frequency region (1600 cm\(^{-1}\)) due to H-bonding (\(-\text{N=CH}•••\text{NC}\)). The peaks within the range 1600 cm\(^{-1}\) to 500 cm\(^{-1}\) also become flattened due to enhancement of electronic distribution all over the molecular scaffold (Fig. S17\textsuperscript{†}).

![Fig. S20 FT-IR Spectroscopy of DNMH and DNMH\(•••\)CN\(^-\) complex.](image)

**Antibacterial Assay**

A quantification of cellular growth and inhibition can be well understood from the cytotoxicity study. For the purpose of preliminarily counting and culturing of the cells, 24-well plates and centrifuge tubes have been utilized. At the initial stage the cells have been incubated at ambient conditions and thereby observed at regular time interval. The in vitro cytotoxicity assay study has been performed in the microtiter well plates during their logarithmic phase after 12-16 h of incubation. The Bt and E. Coli cells are then centrifuged to form pellets and subsequently washed with saline water. A wide range of varying concentration of the cytotoxic agent (DNMH), 10\(^{-2}\)M-10\(^{-7}\)M has been added to the previously formed cell pellets in separate tubes. The cells are then suspended via mild vortexing followed by incubation at 37°C for 2 hrs. The treated cells are centrifuged again and the pellets are washed with normal saline and suspended in sterile broth medium followed by its incubation for 2-4 hrs at 37°C. After incubation the MTT reagent (3-(4,5-dimethylthiazol-2-yl)-2,5-diphenyl tetrazolium bromide) is added in individual tubes which is further subjected to 2-4 hrs incubation at 37°C until formation of purple coloured formazan crystals. A comparative study has been performed taking into consideration
one positive control (contains no cytotoxic agent), one negative control (contains only the broth and the MTT reagent) and one blank (contains only the broth). For terminating the reaction as well as dissolving the so formed formazan crystals acidic isopropanol is added following the process of incubation. After addition of the solubilising agent all the contents were thoroughly mixed and further subjected to 1hr incubation at 37˚C for complete dissolution of the obtained formazan crystals. The spectrophotometric reading is taken at 570 nm wherein the absorbance measurement of the formazan is taken separately. The entire spectrophotometric study has been performed for individual set that has been taken in triplicate.

**Time Dependant Density Functional Theoretical (TDDFT) calculation.** To congregate detailed information regarding the experimentally obtained absorption bands of DNMH and DNMH•••CN⁻, TDDFT calculations have been performed. B3LYP hybrid functional and defSV(P)/defTZVP were used in Turbomole (V7.0) software TmoleX interface, 4.1.1. The experimental along with simulated spectra (applying COSMO/acetonitrile) of DNMH, DNMH•••CN⁻ are shown in Fig. S18†. In case of DNMH, the simulated peak at 390 nm is in good agreement with the experimentally obtained $\lambda_{\text{max}}$ at 425 nm. For DNMH•••CN⁻ adduct, the theoretical peak at 510 nm appeared close to its experimentally resolved $\lambda_{\text{max}}$ at 550 nm. The major electronic excitation calculations and the coordinates for TDDFT of DNMH and DNMH•••CN⁻ have been presented in Table S8-S10†.

**Fig. S21**
Experimental and simulated UV-Vis spectrum of (a) DNMH and (b) DNMH + CN⁻ (Simulation
done at TDDFT/B3LYP/defSV(P)/dEFTZVP level of theory).

**Table S8.** Major electronic excitation calculated by TDDFT in Turbomole (V7.0) software TMoleX interface, 4.1.1.

| Chemosensor | $\lambda_{\text{max}}$ (Experimental) | $\lambda_{\text{max}}$ (Theoretical) | Energy (eV) | Oscillator Strength |
|-------------|-------------------------------------|-------------------------------------|-------------|--------------------|
| DNMH        | 390                                 | 425                                 | 2.97        | 0.7887             |
| DNMH + CN$^-$ | 510                                | 550                                 | 1.72        | 0.4921             |

**Fig. S22** The plausible mechanistic course of interaction of DNMH with CN$^-$.

**Table S9.** Coordinates for TDDFT of DNMH

| Frequency | Oscillator strength |
|-----------|---------------------|
| 234.76840115 | 0.000000063 |
| 237.13851714 | 0.000000115 |
| 239.50863313 | 0.00000207  |
| 241.87874912 | 0.00000366  |
| 244.24886511 | 0.0000064   |
| 246.6189811  | 0.00001103   |
| 248.98909708 | 0.00001877   |
| 251.35921307 | 0.00003149   |
| 253.72932906 | 0.00005214   |
| 256.09944505 | 0.00008515   |
| 258.46956104 | 0.0001372    |
| 260.83967703 | 0.00021806   |
| 263.20979302 | 0.00034188   |
| 265.579909   | 0.00052876   |
| 267.95002499 | 0.00080673   |
| 270.32014098 | 0.00121414   |
| 272.69025697 | 0.00180255   |
| 275.0637296  | 0.00263989   |
| 277.43048895 | 0.00381383   |
| 279.80060493 | 0.00543523   |
| 282.17072092 | 0.00764117   |
| 284.54083691 | 0.01059719   |
| 286.9109529  | 0.01449827   |
| 289.28106889 | 0.01956787   |
| 291.65118488 | 0.02605443   |
| 294.02130087 | 0.03422472   |
| 296.39141685 | 0.04435378   |
Table S10. Coordinates for TDDFT of DNMH•••CN-

| Frequency | Oscillator strength |
|-----------|---------------------|
| 321.35222227 | 0.00000072          |
| 324.51339905 | 0.00000125          |
| 327.67457583 | 0.00000213          |
| 330.8357526  | 0.00000358          |
| 333.99692938 | 0.00000594          |
| 337.15810616 | 0.00000971          |
| 340.31928294 | 0.00001568          |
| 343.48045971 | 0.00002498          |
| 346.64163649 | 0.00003925          |
| 349.80281327 | 0.00006085          |
| 352.96399005 | 0.00009307          |
| 356.12516682 | 0.00014044          |
| 359.2863436  | 0.00020909          |
| 362.44752038 | 0.00030711          |
| 365.60869715 | 0.00044505          |
| 368.76987393 | 0.00063628          |
| 371.93105071 | 0.00089749          |
| 375.09222749 | 0.00124897          |
| 378.25340426 | 0.00171479          |
| 381.41458104 | 0.00232278          |
| 384.57575782 | 0.00310415          |
| 387.7369346  | 0.00409276          |
| 390.89811137 | 0.00532389          |
| 394.05928815 | 0.0068325           |
| 397.22046493 | 0.00865105          |
| 400.3816417  | 0.0108068           |
| 403.54281848 | 0.01331878          |
| 406.70399526 | 0.01619464          |
| 409.86517204 | 0.01942758          |
| 413.02634881 | 0.02299362          |
| 416.18752559 | 0.02684967          |
| 419.34870237 | 0.03093259          |
| 422.50987915 | 0.03515952          |
| 425.67105592 | 0.03942983          |
### Table S11. Geometry optimized coordinates of DNMH

| Atom | X   | Y   | Z   | E   |
|------|-----|-----|-----|-----|
| O    | 0.5641191 | 2.2992252 | 8.3952957 |
| O    | 0.6128636 | 2.7667700 | 9.0087350 |
| O    | 0.4591261 | 3.7426301 | 15.2552727 |
| O    | 0.5951047 | 1.7092019 | 14.5338963 |
| N    | 0.3938059 | 4.8465100 | 9.0518469 |
| N    | 0.5651332 | 1.9428624 | 9.5780545 |
| N    | 0.3044936 | 6.1701642 | 8.8000230 |
| N    | 0.5148491 | 2.9186343 | 14.3491066 |
| C    | 0.5078360 | 2.9595683 | 10.6145321 |
| C    | 0.4238536 | 4.3622801 | 10.3188551 |
| C    | 0.5368223 | 2.5007317 | 11.9344138 |
| C    | 0.1441757 | 8.9284558 | 8.1104692 |
| C    | 0.4833197 | 3.4084436 | 12.9720234 |
| C    | 0.1176309 | 8.2808276 | 5.7464407 |
| C    | 0.1775574 | 7.9221244 | 7.1452498 |
| C    | 0.3983811 | 4.7943432 | 12.7292314 |
| C    | 0.2713031 | 6.5195407 | 7.5581616 |
| C    | 0.3696208 | 5.2540089 | 11.431617 |
| C    | 0.0840575 | 7.7341164 | 3.3595428 |
| C    | 0.0233385 | 9.6760197 | 5.3986373 |
| C    | 0.1454045 | 7.3328603 | 4.6807729 |
| C    | 0.0525478 | 10.2899586 | 7.7576637 |
| C    | -0.0071789 | 10.6583228 | 6.4269999 |
| C    | -0.0093590 | 9.1085190 | 3.0237338 |
| C    | -0.0385303 | 10.0556632 | 4.0257452 |
| H    | 0.4366378 | 4.1648636 | 8.2873752 |
| H    | 0.6010713 | 1.4323050 | 12.1274015 |
| H    | 0.1914415 | 8.6462664 | 9.1649681 |
| H    | 0.3566111 | 5.4935016 | 13.562192 |
| H    | 0.3138653 | 5.7438182 | 6.7799233 |
| H    | 0.3042940 | 6.3214450 | 11.2258177 |
| H    | 0.1078718 | 6.9819463 | 2.5649270 |
| H    | 0.2163152 | 6.2644849 | 4.8938122 |
| H    | 0.0294275 | 11.0507964 | 8.5435470 |
| H    | -0.0787939 | 11.7137670 | 6.1453421 |
| H    | -0.0578378 | 9.4116616 | 1.9734244 |
| H    | -0.1101795 | 11.1203618 | 3.7816881 |

### Table S12. Geometry optimized structure of DNMH•••CN⁻

| Atom | X   | Y   | Z   | E   |
|------|-----|-----|-----|-----|
| O    | 1.1132480 | 2.4269030 | 8.4665878 |
| O  | 1.1983456 | 0.8320113 | 9.9533358 |
|----|------------|------------|------------|
| O  | 0.6267295  | 3.6803476  | 15.3780434 |
| O  | 0.9315731  | 1.6563751  | 14.6056856 |
| N  | 0.5553439  | 4.9345257  | 9.1672719  |
| N  | 1.0599879  | 2.0221321  | 9.6424910  |
| N  | 0.4293000  | 6.2768868  | 9.0057819  |
| N  | 0.7645059  | 2.8745356  | 14.4451017 |
| C  | 0.8399690  | 3.0042190  | 10.6976103 |
| C  | 0.6339123  | 4.4066764  | 10.4243023 |
| C  | 0.8755315  | 2.5173816  | 12.0120672 |
| C  | 0.4089474  | 9.0855176  | 8.4566444  |
| C  | 0.7262485  | 3.3959897  | 13.0831473 |
| C  | -0.1527359 | 8.5489064  | 6.1181548  |
| C  | 0.1777158  | 8.1191104  | 7.4637563  |
| C  | 0.5442434  | 4.7832577  | 12.8598467 |
| C  | 0.2804117  | 6.6935933  | 7.773268   |
| C  | 0.5018295  | 5.2678510  | 11.564404  |
| C  | -0.7392092 | 8.1116146  | 3.7726408  |
| C  | -0.2166438 | 9.700084   | 5.8415576  |
| C  | -0.4297727 | 7.6432212  | 5.0456717  |
| C  | 0.3360829  | 10.4688889 | 8.1759374  |
| C  | 0.0328257  | 10.9059062 | 6.889359   |
| C  | -0.7904715 | 9.5070787  | 3.5023453  |
| C  | -0.5354762 | 10.4152776 | 4.5205974  |
| H  | 0.5509960  | 4.2963590  | 8.339722   |
| H  | 1.0309044  | 1.4449193  | 12.1808299 |
| H  | 0.6640240  | 8.7443254  | 9.4722573  |
| H  | 0.4323916  | 5.4616920  | 13.717594  |
| H  | 0.2200838  | 5.9448776  | 6.9616640  |
| H  | 0.3514573  | 6.3382788  | 11.3747184 |
| H  | -0.9510689 | 7.3928719  | 2.9637781  |
| H  | -0.4130171 | 6.5549304  | 5.2181303  |
| H  | 0.5280560  | 11.1985931 | 8.9795085  |
| H  | -0.0189077 | 11.9839511 | 6.6627453  |
| H  | -1.0363679 | 9.8623776  | 2.4881078  |
| H  | -0.5779461 | 11.5002609 | 4.3264926  |
| N  | -0.2850853 | 3.9629664  | 6.2187202  |
| C  | -0.9279644 | 3.3421980  | 5.4375832  |
| N  | -0.306354  | 2.0132642  | 1.977340   |
| C  | 0.7369353  | 3.0412606  | 2.3462243  |
| C  | -1.6693838 | 2.6434203  | 2.0061807  |
| C  | -0.2309228 | 0.8677886  | 2.9567133  |
| C  | -0.0486613 | 1.5114390  | 0.5820209  |
| H  | 0.6716479  | 3.8846455  | 1.6317659  |
| H  | 0.5315589  | 3.3832453  | 3.3812034  |
| H  | 1.7397304  | 2.5700479  | 2.2830419  |
| H  | -0.4400091 | 1.2765980  | 3.9674078  |
| H  | -0.9919200 | 0.1151132  | 2.6727455  |
| H  | 0.7823131  | 0.4236149  | 2.9098194  |
| H  | 0.9569145  | 1.0493115  | 0.5456857  |
| H  | -0.8229789 | 0.7617505  | 0.3284115  |
| H  | -0.1016086 | 2.3670061  | -0.1188661 |
| H  | -1.8127023 | 2.9927167  | 3.1054569  |
| H  | -2.4249741 | 1.8794813  | 1.7919569  |
Fig. S23 Geometry optimized structures (a) HOMO of DNMH, (b) LUMO of DNMH, (c) HOMO of DNMH in presence of CN⁻, (d) LUMO of DNMH in presence of CN⁻. [Colour codes: White (H); Magenta (C); Blue (N); Red (O)].

Table S13. Geometry optimized coordinates of Fukui Indices for Nucleophilic Attack (Fukui (+)): DNMH

| atom | Mulliken | Hirshfeld |
|------|----------|-----------|
| O (1) | 0.131    | 0.125     |
| O (2) | 0.139    | 0.131     |
| O (3) | 0.081    | 0.079     |
| O (4) | 0.077    | 0.074     |
| N (5) | 0.019    | 0.025     |
| N (6) | 0.080    | 0.093     |
| N (7) | -0.001   | 0.008     |
| N (8) | 0.043    | 0.050     |
| C (9) | 0.007    | 0.027     |
| C (10)| 0.045    | 0.034     |
| C (11)| 0.087    | 0.067     |
| C (12)| 0.012    | 0.010     |
| C (13)| 0.008    | 0.028     |
| C (14)| 0.006    | 0.004     |
| C (15)| -0.004   | 0.003     |
| C (16)| 0.043    | 0.043     |
| C (17)| 0.029    | 0.026     |
| atom   | Mulliken | Hirshfeld |
|--------|----------|-----------|
| O ( 1) | 0.015    | 0.017     |
| O ( 2) | 0.025    | 0.025     |
| O ( 3) | 0.019    | 0.019     |
| O ( 4) | 0.020    | 0.020     |
| N ( 5) | 0.038    | 0.043     |
| N ( 6) | 0.008    | 0.011     |
| N ( 7) | 0.046    | 0.050     |
| N ( 8) | 0.006    | 0.009     |
| C ( 9) | 0.024    | 0.019     |
| C (10) | 0.002    | 0.011     |
| C (11) | 0.008    | 0.011     |
| C (12) | 0.047    | 0.052     |
| C (13) | 0.023    | 0.022     |
| C (14) | 0.013    | 0.017     |
| C (15) | 0.050    | 0.058     |
| C (16) | 0.012    | 0.016     |
| C (17) | 0.027    | 0.030     |
| C (18) | 0.024    | 0.022     |
| C (19) | 0.036    | 0.045     |
| C (20) | 0.003    | 0.018     |
| C (21) | 0.049    | 0.052     |
| C (22) | 0.032    | 0.046     |
| C (23) | 0.072    | 0.075     |
| C (24) | 0.031    | 0.041     |
| C (25) | 0.050    | 0.054     |
| H (26) | 0.019    | 0.015     |
| H (27) | 0.012    | 0.007     |
| H (28) | 0.036    | 0.023     |

Table S14. Geometry optimized coordinates of Fukui Indices for Electrophilic Attack (Fukui (-): DNMH
Table S15. Geometry optimized coordinates of Fukui Indices for Nucleophilic Attack (Fukui(+)): DNMH\text{•••}\text{CN}^-

| atom | Mulliken | Hirshfeld |
|------|----------|-----------|
| O ( 1) | 0.107    | 0.101     |
| O ( 2) | 0.112    | 0.106     |
| O ( 3) | 0.108    | 0.103     |
| O ( 4) | 0.105    | 0.099     |
| N ( 5) | 0.019    | 0.023     |
| N ( 6) | 0.063    | 0.073     |
| N ( 7) | 0.000    | 0.008     |
| N ( 8) | 0.058    | 0.068     |
| C ( 9) | 0.008    | 0.026     |
| C (10) | 0.040    | 0.033     |
| C (11) | 0.085    | 0.068     |
| C (12) | 0.013    | 0.012     |
| C (13) | 0.004    | 0.027     |
| C (14) | 0.006    | 0.004     |
| C (15) | -0.004   | 0.003     |
| C (16) | 0.039    | 0.038     |
| C (17) | 0.031    | 0.027     |
| C (18) | 0.022    | 0.029     |
| C (19) | 0.005    | 0.005     |
| C (20) | 0.000    | 0.003     |
| C (21) | 0.001    | 0.002     |
| C (22) | 0.001    | 0.005     |
| C (23) | 0.014    | 0.013     |
| C (24) | 0.003    | 0.004     |
| C (25) | 0.004    | 0.004     |
| N (26) | -0.009   | -0.001    |
| C (27) | 0.013    | 0.008     |
| N (28) | 0.000    | 0.000     |
| C (29) | 0.000    | 0.000     |
Table S16. Geometry optimized coordinates of Fukui Indices for Electrophilic Attack (Fukui (-)): DNMH•••CN⁻

| atom | Mulliken | Hirshfeld |
|------|----------|-----------|
| O (1) | 0.019 | 0.019 |
| O (2) | 0.030 | 0.029 |
| O (3) | 0.025 | 0.024 |
| O (4) | 0.026 | 0.025 |
| N (5) | 0.047 | 0.051 |
| N (6) | 0.008 | 0.012 |
| N (7) | 0.053 | 0.055 |
| N (8) | 0.006 | 0.012 |
| C (9) | 0.028 | 0.022 |
| C (10) | 0.004 | 0.013 |
| C (11) | 0.009 | 0.013 |
| C (12) | 0.048 | 0.051 |
| C (13) | 0.027 | 0.027 |
| C (14) | 0.012 | 0.016 |
| C (15) | 0.041 | 0.052 |
| C (16) | 0.014 | 0.019 |
| C (17) | 0.035 | 0.036 |
| C (18) | 0.027 | 0.026 |
| C (19) | 0.028 | 0.036 |
| C (20) | 0.002 | 0.016 |
| C (21) | 0.038 | 0.040 |
| C (22) | 0.024 | 0.039 |
| C (23) | 0.068 | 0.070 |
| C (24) | 0.028 | 0.035 |
Fukui Indices (Fls)

Herein the electronic charges were calculated from the condensed Fukui functions, defined as the first order derivative of electron density of $ρ(\vec{r})$, with respect to ‘$N$’ number of electrons at steady external potential of $ν(\vec{r})$ as follows:

$$f_k = \left(\frac{\partial ρ(\vec{r})}{\partial N}\right)_{ν(\vec{r})}$$

(1)

For the susceptible nucleophilic attack ($\square ^{-}$) and electrophilic attack ($\square ^{+}$) the corresponding Fls are denoted separately by the following equations [37]:

$$f_k^+ = q_k(N+1) - q_k(N)$$

(2)

$$f_k^- = q_k(N) - q_k(N-1)$$

(3)

Herein, $q_k(N)$ is associated with the gross charge of $k^{th}$ atom, whilst $q_k(N)$, $q_k(N+1)$ and $q_k(N-1)$ represents the neutral, negatively charged and positively charged entities Hirshfeld and Mulliken population analysis-based Fukui functions that have been derived by the finite difference approximation.

Application in Smartphone based colorimetric sensory prototype

Components of the prototype. The overall set up of the sensory prototype comprises of the indigenously fabricated light weight black acrylic make prototype box which consists of a compartment on one side wherein the Smartphone readily slides in. The other end consists of a circular sample holder with black lid for placing the vial containing the sample solution and covering the same to avoid false positive signals owing to penetration of any external light. 1 shield is basically one type of Arduino shield which is being used as a user interface to establish communication between the Smartphone and microcontroller. Turning on of any of the three LEDs (yellow, green or red) that are externally connected
with the 1 sheeld would provide the resulting output of the chemical response. Additionally, a power bank has been combined to provide the requisite power supply. The overview and segment wise visualization of the fabricated Smartphone assisted sensory prototype along with its AutoCAD diagram (SOLIDWORKS Software) has been provided in Fig. 9 and Fig. S24-S29†.

**Fig. S24** An overview of the primary components of the Smartphone based prototype: smartphone, 1 sheeld and power bank.

**Fig. S25** Sensory prototype displaying the Smartphone, black acrylic prototype box along with its lid separately.
Fig. S26 AutoCAD diagram displaying the front view of the Smartphone based prototype.

Fig. S27 AutoCAD diagram displaying the output response as ‘Yellow LED’ turned on when only DNMH solution (yellow colour) is present in the sample vial.
Fig. S28 AutoCAD diagram displaying the output response as “Green LED” turned on when DNMH•••CN⁻ (low conc) solution (orangish red colour) is present in the sample vial.

Fig. S29 AutoCAD diagram displaying the output response as “Red LED” turned on when DNMH•••CN⁻ (high conc) solution (red colour) is present in the sample vial.

Table S17. Comparative literature survey of DNMH with the reported chemosensor for CN⁻ detection

| Sl. no | Structure of the molecular probes | Detection Limit (M) | Binding Constant | Logic gate | Electrochemical | Smartphone based | Ref. |
|-------|----------------------------------|--------------------|------------------|-----------|----------------|-----------------|-----|


|   | Structure | $(M^{-1})$      | Sensing | Detection |
|---|-----------|-----------------|---------|-----------|
| 1. | ![Structure 1](image1) | $8 \times 10^{-7}$ | No      | No        | No        | [4a]     |
| 2. | ![Structure 2](image2) | $2.31 \times 10^{-8}$ | 6.692 $\times 10^4$ | No      | No        | No        | [4b]     |
| 3. | ![Structure 3](image3) | $1.1 \times 10^{-6}$ | 3.25 $\times 10^5$ | No      | No        | No        | [4c]     |
| 4. | ![Structure 4](image4) | $1.47 \times 10^{-6}$ | --      | No        | No        | No        | [4d]     |
| 5. | ![Structure 5](image5) | $9.8 \times 10^{-9}$ | --      | No        | No        | No        | [4e]     |
| 6. | ![Structure 6](image6) | $7.5 \times 10^{-7}$ | --      | No        | No        | No        | [4f]     |
| 7. | ![Structure 7](image7) | $1.26 \times 10^{-6}$ | --      | No        | No        | No        | [4g]     |
| 8. | ![Structure 8](image8) | $1.47 \times 10^{-6}$ | --      | No        | No        | No        | [4h]     |
| No. | Structure | $S_1$ ($\times 10^{-6}$) | $S_2$ ($\times 10^3$) | $P_{on}$ | $P_{off}$ | $P_{on/off}$ | Reference |
|-----|-----------|--------------------------|----------------------|---------|---------|-------------|-----------|
| 9.  | ![Structure](image1) | $1.94 \times 10^{-6}$ | $2.2 \times 10^3$ | No      | No      | No          | [4i]      |
| 10. | ![Structure](image2) | $6.8 \times 10^{-7}$ | --                   | No      | No      | No          | [4j]      |
| 11. | ![Structure](image3) | $9 \times 10^{-8}$    | --                   | No      | No      | No          | [4k]      |
| 12. | ![Structure](image4) | $2 \times 10^{-7}$    | --                   | No      | No      | No          | [4l]      |
| 13. | ![Structure](image5) | $1.26 \times 10^{-7}$ | $1.4 \times 10^{-7}$ | No      | No      | No          | [4m]      |
| 14. | ![Structure](image6) | $1 - 10 \times 10^6$  | --                   | Yes     | Yes     | No          | [4n]      |
| No. | Compound | IC_{50} | Affinity | Cytotoxicity | Inhibitory | This work |
|-----|----------|---------|----------|-------------|------------|-----------|
| 15. | ![DNMH](image) | 298 nM | 4.48×10^6 M^{-1} | Yes | Yes | Yes | This work |

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