Electronic Supplementary Information

Supramolecular Interactions

The complex shows supramolecular layer structure with inter layer separation (shortest nickel-nickel distance) of 9.5781(7) Å. The H atom, H(10B), attached with O(100) of the lattice water forms bifurcated H-bonding with the methoxy oxygen atom O(9) and phenoxy oxygen atom atom, O(8) of Schiff base ligand. Another H atom, H(10A), attached with O(100) of the lattice water is H-bonded with N(24)$^{\#}$ of a $(PTZ)^{-}$ ligand from a symmetry related ($^{\#}$ = 2-x,1/2+y,5/2-z) molecule. On the other hand, the H atom, H(3), attached with amine nitrogen atom, N(3), is H-bonded with an oxygen atom, O(100)* of a symmetry related (* = 2-x,-y,2-z) water of crystallization. The combination of these three H-bonds resulted a two dimensional H-bonded network as shown in Figure S1. The details of H bonding dimensions are given in Table S3.

The complex also shows significant intra and intermolecular C–H···π interactions. One methylene C(sp^3) hydrogen, H(2A) of N-ethyl group C(2) in Schiff base *HL* shows intramolecular interaction with chelate ring Cg(2) produced by *HPTZ* ligand. An aromatic C(sp^2) hydrogen, H11 of phenyl ring is involved in intermolecular interactions with the pyrazine ring Cg(6) of *HPTZ* forming an 1D array in the crystal packing of **1** (Figure S2). Geometric features of the C–H···π interactions are given in Table S4. Considering the centrosymmetric dimer as a node, each node is connected with two other node by C-H···π interactions and four other node by hydrogen bonding interactions through water molecule. These combined supramolecular force (C-H···π interactions and H-bond) creates a 3^6-**hxl** topological supramolecular network as shown in Figure S3. It is noteworthy that acetonitrile molecule present in the crystal structure is actually

encapsulated as a hydrophobic guest within the 2D supramolecular network as shown in Figure S7.

Table S1: Selected bond angles (º) around nickel(II) in complex **1**.

O(8) – Ni(1) – N(3)	174.15(6)	N(6) – Ni(1) – N(14)	93.57(6)
O(8) – Ni(1) – N(6)	91.53(6)	N(6) – Ni(1) – N(21)	170.82(6)
O(8) – Ni(1) – N(14)	86.76(6)	N(6) – Ni(1) – N(22)*	93.60(6)
O(8) – Ni(1) – N(21)	90.20(6)	N(14) – Ni(1) – N(21)	77.52(6)
O(8) – Ni(1) – N(22)*	92.06(6)	N(14) – Ni(1) – N(22)*	172.76(6)
N(3) – Ni(1) – N(6)	82.88(6)	N(21) – Ni(1) – N(22)*	95.35(6)
N(3) – Ni(1) – N(14)	91.84(6)	Ni(1) – N(21) – N(22)	139.85(12)
N(3) – Ni(1) – N(21)	95.04(6)	Ni(1) – N(22)* – N(21)*	124.60(12)
N(3) – Ni(1) – N(22)*	90.03(6)	-	-

Symmetry element: *= 2-x,-y,2-z

Table S2: The details data of the photoluminescence and time-resolved photoluminescence decays of complex **1**.

Complex	λ_{ex}(nm)	λ_{em} (nm)	T_1(ns)	T_2(ns)	B_1	B_2	χ^2	$<\tau>$
1	385	413	0.542	2.168	0.519	0.481	1.054933	1.322 ns
		436	0.230	0.990	0.101	0.899	1.069045	0.914 ns

Table S3: Hydrogen bond distances (Å) and angles (º) for complex **1**.

D–H···A	D–H	D···A	H···A	∠D–H···A
O(100)–H(10B)···O(8)	0.82(3)	3.017(2)	2.24(3)	159(3)
O(100)–H(10B)···O(9)	0.82(3)	2.911(2)	2.30(2)	132(3)
O(100)–H(10A)···N(24)$^{\#}$	0.77(3)	2.977(2)	2.22(3)	169(3)
N(3)–H(3)···O(100)*	0.91(3)	3.004(2)	2.13(3)	160(2)

Symmetry element * = 2-x,-y,2-z; $^{\#}$ = 2-x,1/2+y,5/2-z D, donor; H, hydrogen; A, acceptor

Table S4: Geometric features (distances in Å and angles in degrees) of the C–H···π interactions obtained for **1**.

Complexes	C–H····Cg(Ring)	H····Cg (Å)	C–H····Cg (°)	C····Cg (Å)
1	C(2)–H(2a)···Cg(2)	2.71	3.083(2)	103
	C(11)–H(11)···Cg(6)	2.98	3.872(2)	157

Cg(2) = Centre of gravity of the ring [Ni(1)–N(14)–C(19)–C(20)– N(21)] and Cg(6) = Centre of gravity of the ring [N(14)–C(15)–C(16)–N(17)–C(18)–C(18)]

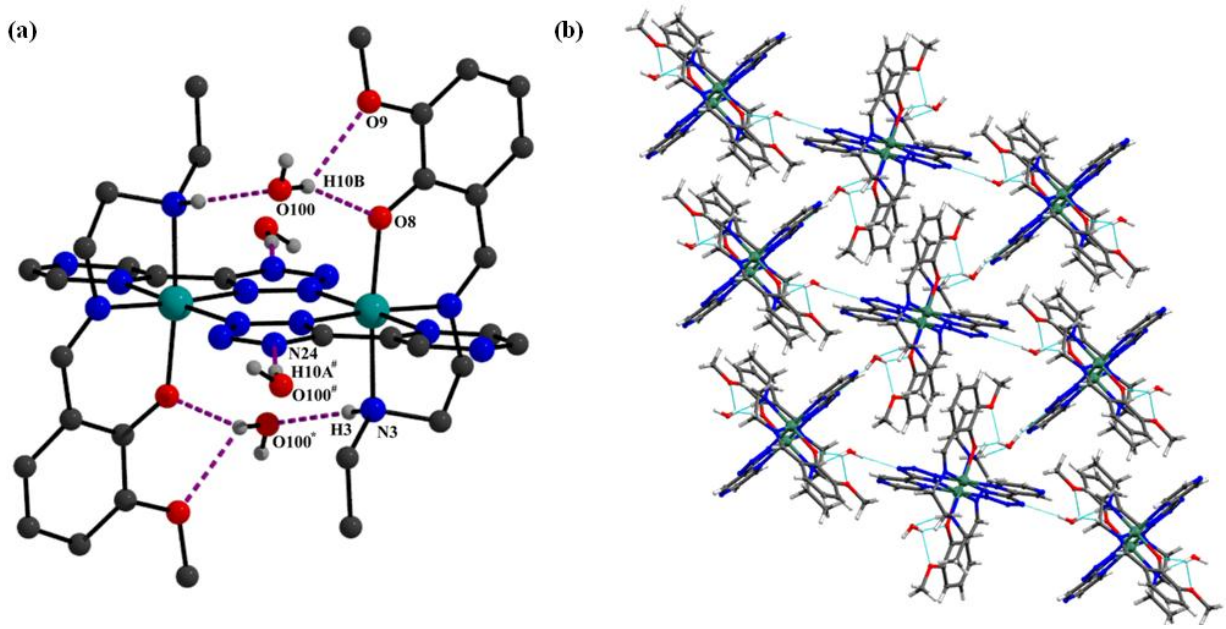

Figure S1: Perspective view of (a) H-bonding interaction and (b) two dimensional H-bonded layer structure of complex **1**. Only the relevant hydrogen atoms are shown for clarity. Symmetry element * = 2-x,-y,2-z; $^{\#}$ = 2-x,1/2+y,5/2-z.

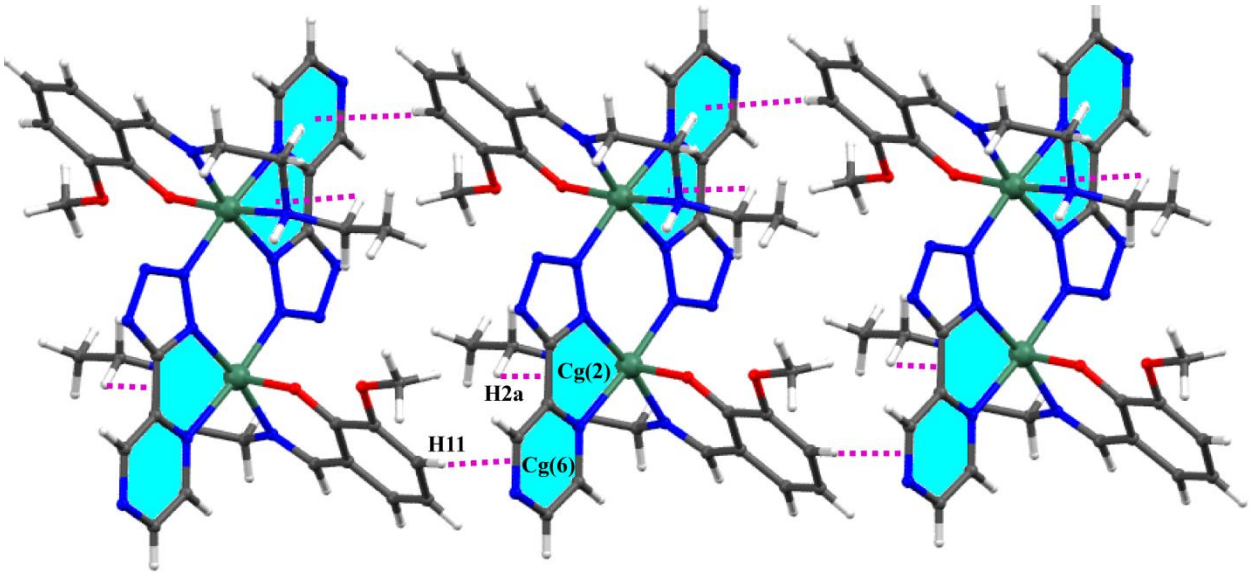

Figure S2: Perspective view of one dimensional array in crystal packing via C–H···π interactions.

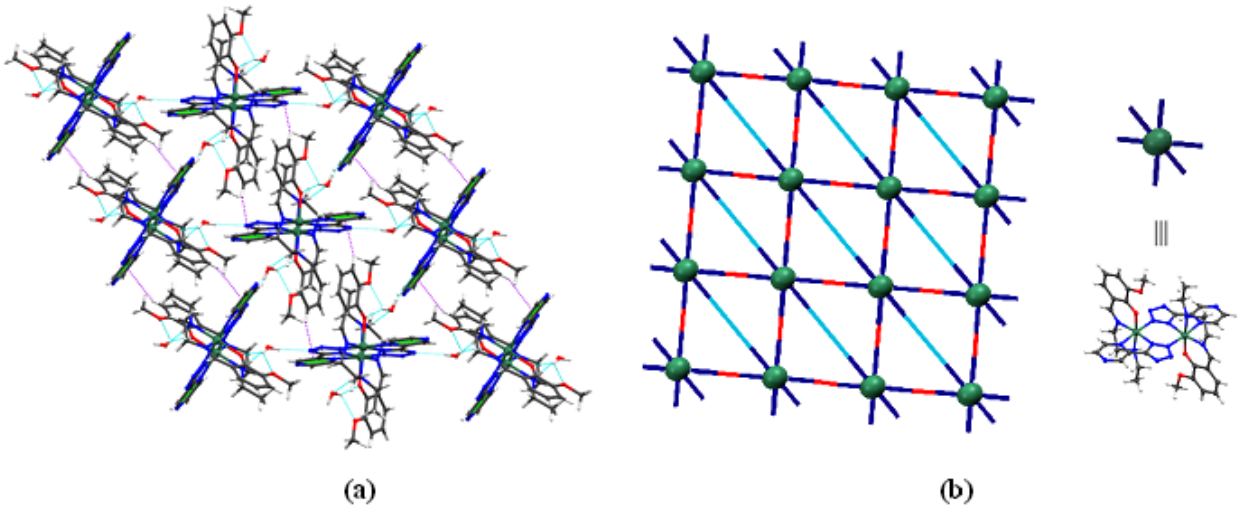

Figure S3: (a) Perspective view of two-dimensional supramolecular layer structure highlighting H-bonding and intermolecular C-H···π interactions (b) A view of 3^6-**hxl** topological supramolecular network. The acetonitrile molecules have been omitted for clarity. Red and aquamarine lines represent water mediated hydrogen bonding interactions and intermolecular C-H···π interactions respectively.

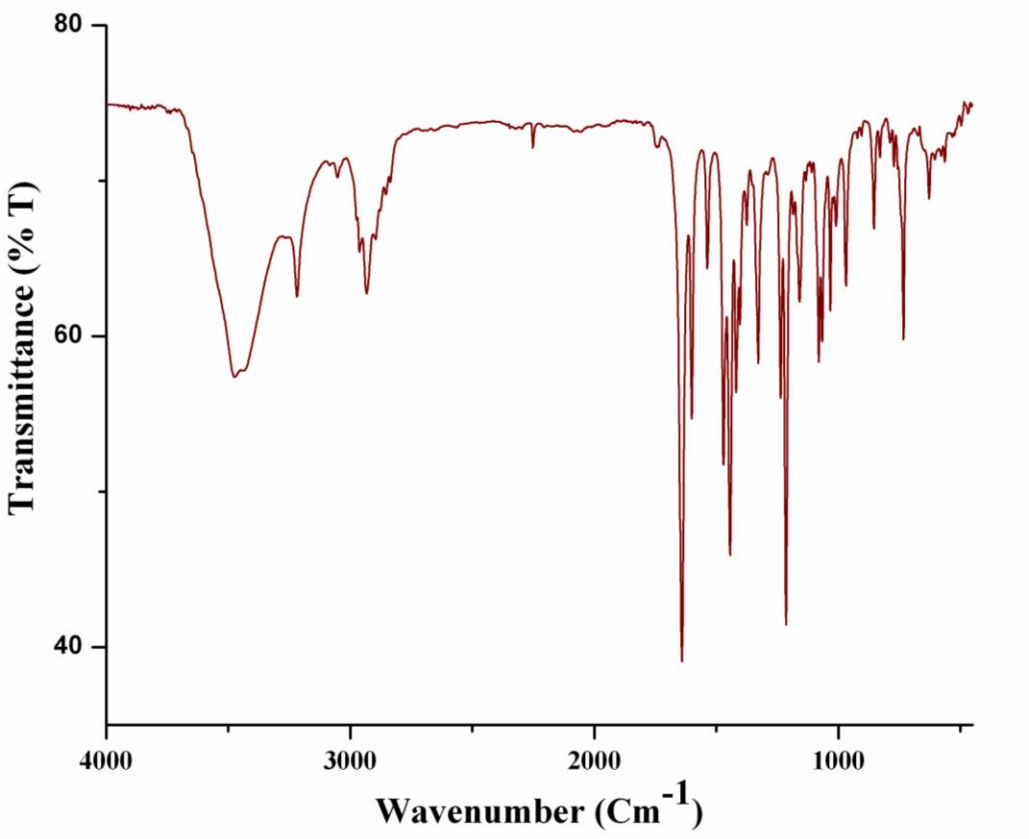

Figure S4: IR spectrum (450-4000 cm^{-1}) of complex **1**.

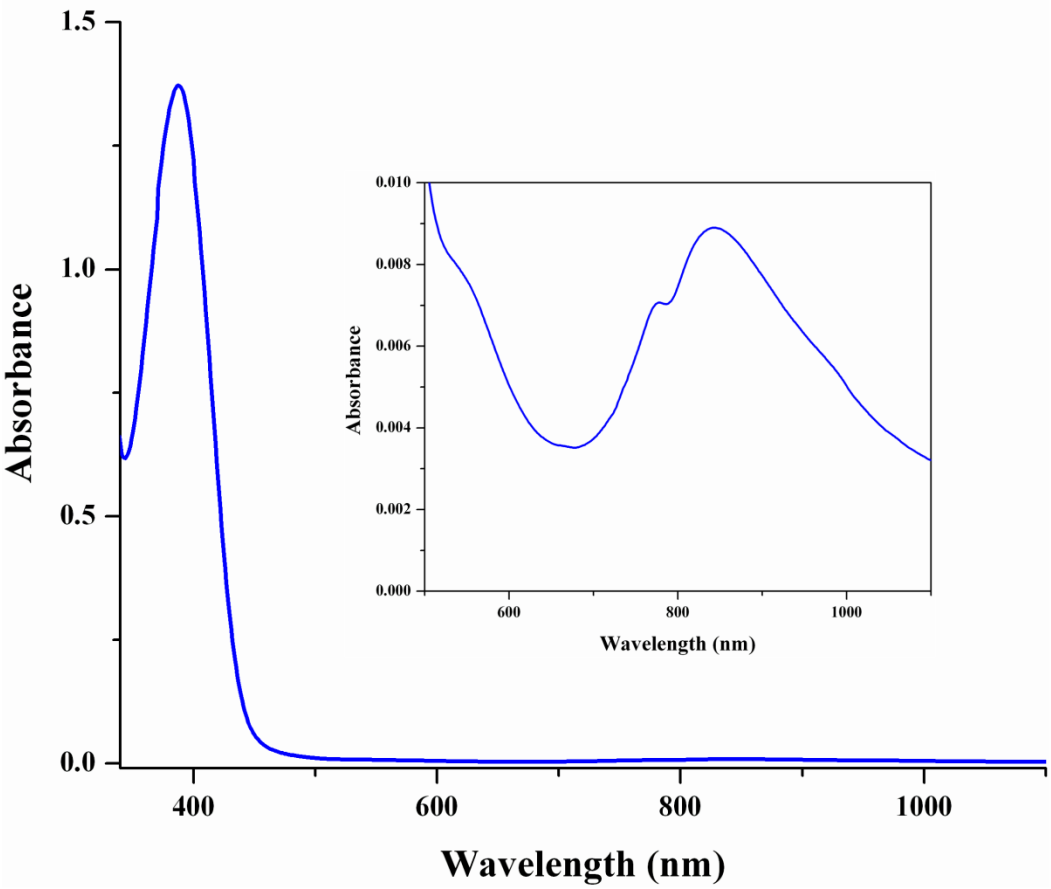

Figure S5: The absorption spectrum of complex **1** in DMSO medium.

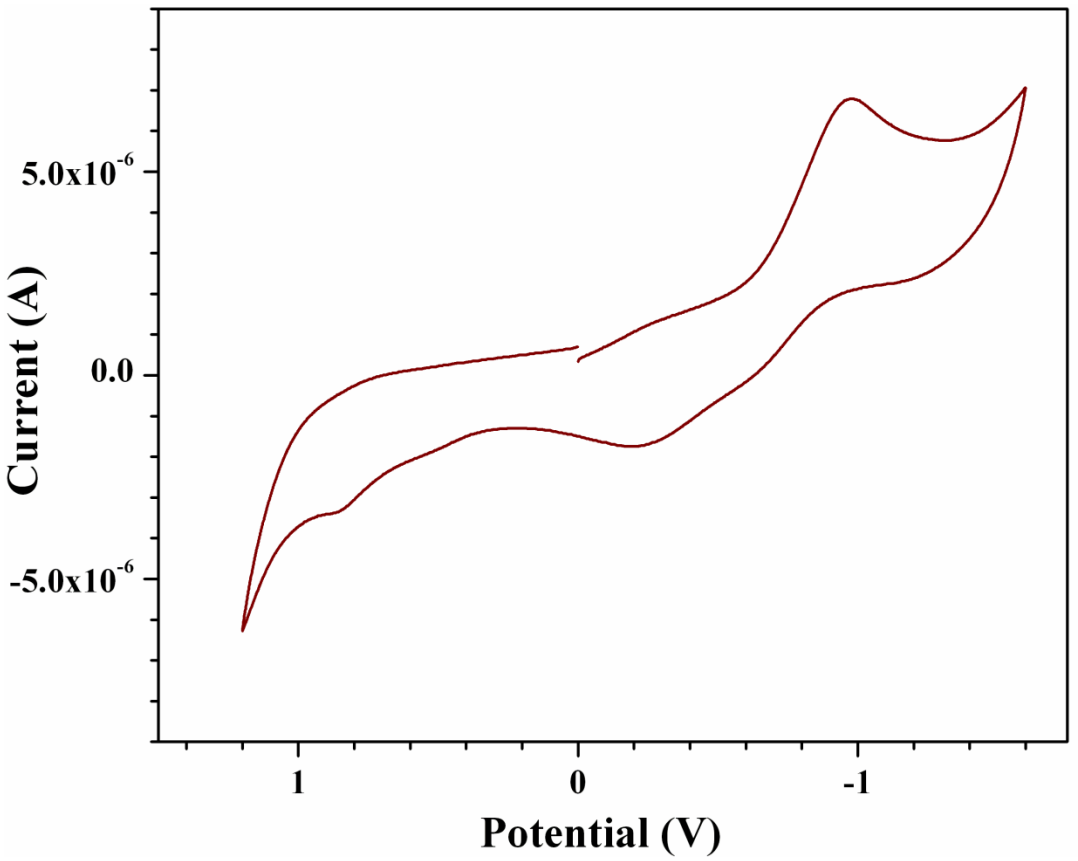

Figure S6: Cyclic voltammogram of complex **1** in DMSO at room temperature.

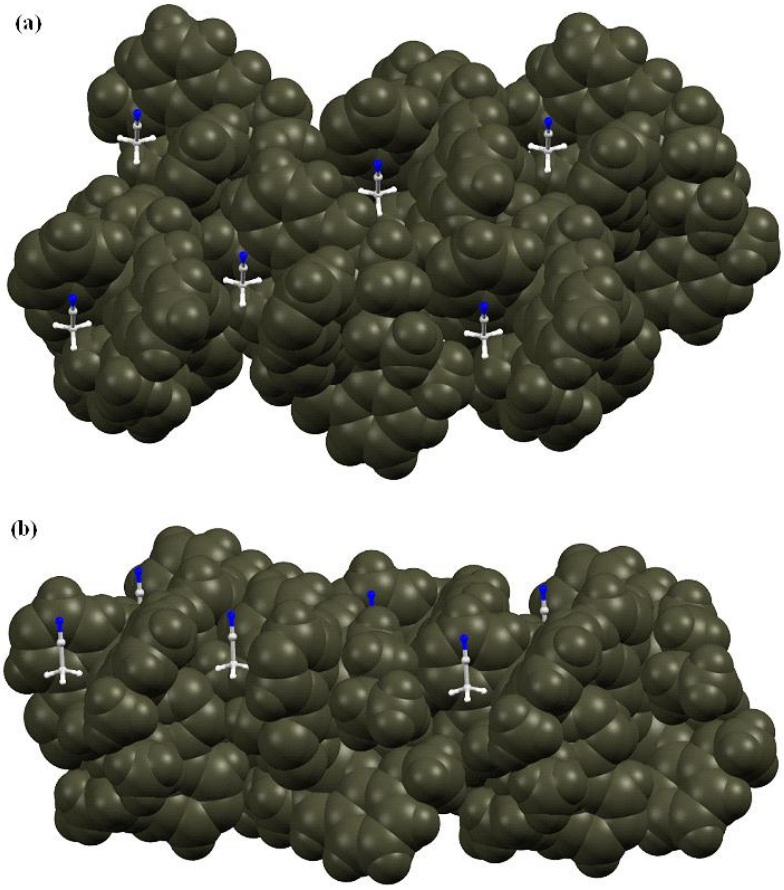

Figure S7: Top view **(a)** and side view **(b)** of encapsulated acetonitrile (hydrophobic) molecule in the 2D supramolecular self-assembly of complex **1**. Dinuclear Schiff base complex and water of crystallization forming supramolecular self-assembly has been shown in space-fill model.