Supporting Information

Supplementary Movie S1

Movie S1 demonstrates the H→DH transition of the gel on application of a small voltage. As the binding energy per water molecule decreases with the increase in the amount of water adsorbed, a small applied voltage of ≈5V can cause enough heat build-up to instigate the transition. The gel was coated on fluorine doped tin oxide (FTO) glass and the voltage was applied using a source meter.

Supplementary Movie S2

Movie S2 demonstrates the IR blocking property of the H gel. Two identical bars of chocolate were kept inside two identical glass tanks. The glass tank on the right has a coating of the H gel on one side. A tungsten halogen bulb was used as a source of IR radiation. The distance of the bulb was adjusted to maintain 1 sun intensity. Due to greenhouse heating, temperatures inside both the glass tanks increase resulting in eventual melting of the chocolate bars. However in the glass tank coated with the gel, the rate of temperature increase was less. The chocolate bar in the glass tank without the gel coating melted at 14:38 minutes whereas the one in the glass tank with the H gel coating melted at 26:55 minutes, thus proving the IR blocking property of the H gel.

Supplementary Movie S3

Movie S3 shows the construction of the electrochemical cell using Zn and Cu as electrodes and the gel as the electrochemical junction between them. An attempt was made to power up a digital meter using the above electrochemical cell. It can be seen that in the DH state of the gel, the power output of the cell is not sufficient to power up the digital meter. When the cell was exposed to humid conditions (around 70% RH maintained by using a moist cotton wool), humidity induced electrochemical changes increase the power output of the cell. This can be seen as the digital meter powers up after exposing the cell to humid conditions for about 10 minutes.

Supplementary Movie S4

Movie S4 demonstrates the application of the gel as a conducting medium in flexible circuits. 3 LEDs were connected in series with the gel in H* state forming the electrical contacts between them. On application of voltage the LEDs light up showing the conducting nature of the gel in H* state.

Supplementary Movie S5

Movie S5 demonstrates the easy washing off of the gel using common reagents like vinegar. This greatly helps in reusability of electrical circuits thus minimising electronic wastes.
Discussion

The DH gel was coated on FTO substrate and X-Ray diffraction measurements were taken. X-ray Diffraction results (Figure S1) of the gel revealed that the gel possessed a short range order of arrangement.

Figure S1: XRD pattern of the gel in DH state
Figure S2: Simulated structures with different Zn:O ratios and their corresponding structures. DOS plot of the corresponding structures (Grey balls: Zn atoms; Red balls: O atoms)
Figure S3: Comparative chart of water absorption capability of the hydrogel when compared to commercial desiccants. A comparison was made with ethanolamine (one of the material used in the hydrogel synthesis) to confirm that the hygroscopicity of the hydrogel is much more than its starting materials.
Figure S4: (a) Variation in energy required for addition of each molecule of water with the number of water molecules introduced into the system. Changes in the DH structure of the gel on introduction of 6 (b), 11 (c), 30 (d) and 43 (e) water molecules. (Green balls: Zn atoms; Red balls: O atoms; White balls: H atoms)
Supplementary Materials and Methods

Materials

All the chemicals were purchased from Sigma Aldrich and were used without any purification. The DI water used was obtained from a milliPore purification system. Fabrication of the electrochemical cell was done on a flexible cellulose acetate sheet (over-head projector sheet). Zinc foil of thickness 0.15-0.25 mm thickness was purchased from Dickson Chemical and commercially available Copper tape were used as the electrodes for the cell.

Methods

Synthesis of the gel:

Zinc Acetate dehydrate (0.7M) and ethanolamine (0.7M) were dissolved in 2-methoxyethanol and was sonicated for about 20 minutes in an ultra-sonicator (Elmasonic S 30H) until all the zinc acetate dissolved to obtain a clear transparent liquid (precursor solution) which when mixed with equal volumes of DI water resulted in the formation of an opaque viscous liquid.
which was then coated on a FTO substrate and heated on a hotplate at 50°C for about 20 minutes, resulting in formation of a thin layer of the DH gel coating on the substrate.

**Simulations:**

Density functional theory (DFT) calculations were done to simulate the possible structure of the experimentally synthesized gel composed of Zn and O atoms by Vienna ab-initio Simulation Package (VASP). The plane-wave basis with the cut-off energy of 400 eV and the generalized gradient approximation (GGA) in the Perdew-Burke-Ernzerh (PBE) format with the projector-augmented wave (PAW) method are employed in all calculations. The structural relaxations were carried out until the Hellmann-Feynman force on each atom is less than 0.01 eV/Å, and the energy convergence criterion was set to 10^{-5} eV. Gamma-point-only is adopted for k sampling of all calculations.

**Characterizations:**

XRD was measured using a Bruker D8 advance at room temperature. Transmittance measurements were carried out using a Shimadzu UV-3600 spectrophotometer. Temperature measurements were carried out using a k-type open thermocouple. IV measurements were recorded on a Keithley K2400 source meter. EIS and CV measurements were done on potentiostat (Autolab PGSTAT128N). Complex viscosity, storage and loss modulus measurements were measured using a MCR302 twindrive rheometer (Anton Paar) with a rotational test. A parallel plate of 2.5cm diameter was employed for the hydrogel sample and a cup type sample holder was used for the sol-gel owing to very low viscosity. Osram classic 1320 lm (77w) halogen lamp was used to simulate 1 sun light intensity conditions.