Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

Self-assembly of sodium and potassium betulinates into hydro- and organo-gels: Entrapment and removal studies of fluorophores and synthesis of gel-gold nanoparticle hybrid materials

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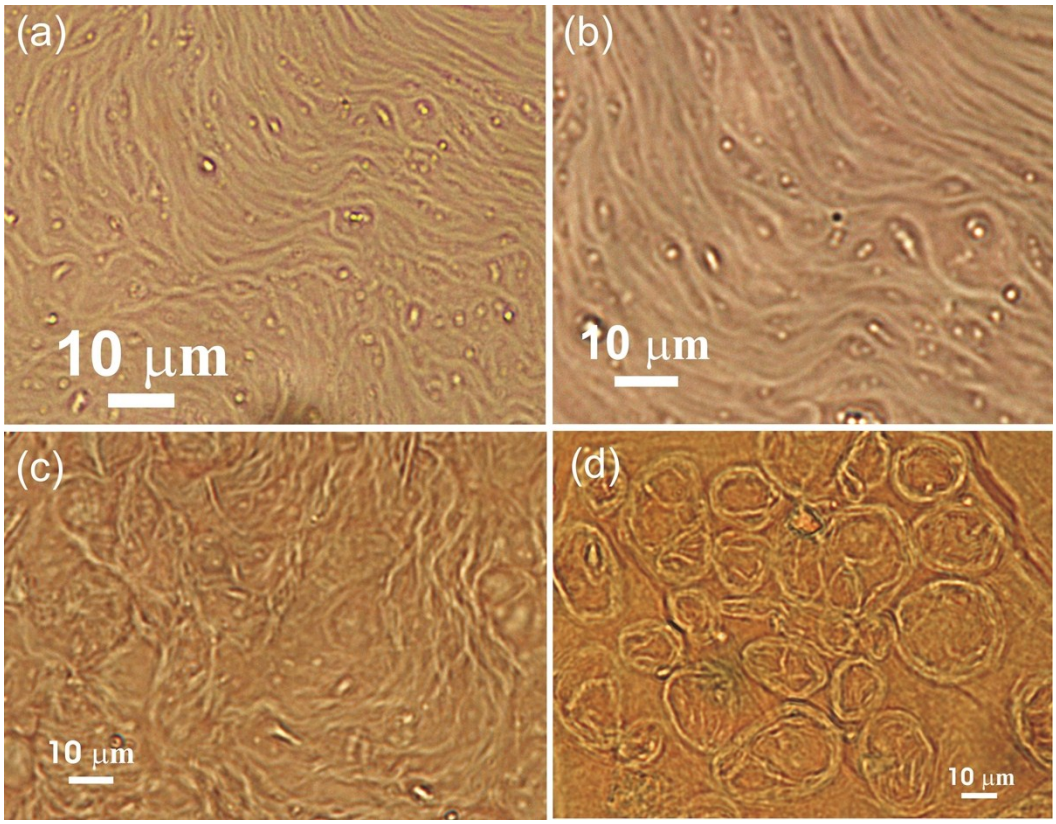


Figure S1: Optical micrographs of potassium betulinate **3:** (a, b) 1.67% w/v in water; (c) 2.8% w/v in EG-water (1:1v/v) and (d) 1.9% w/v in DMF-water (1:1 v/v)

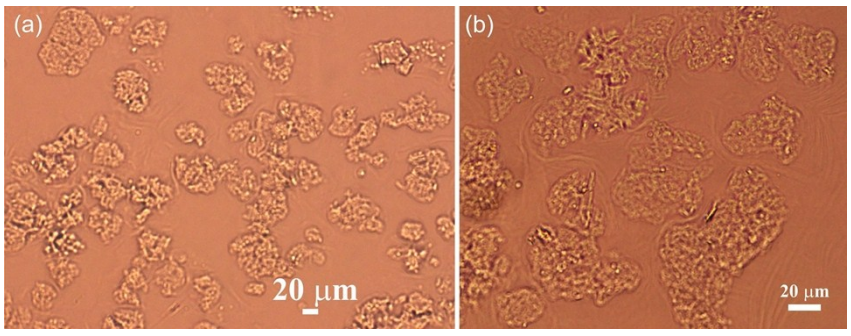


Figure S2: Optical micrographs of sodium betulinate **2** (2.1% w/v) in EG-water (1:1 v/v)

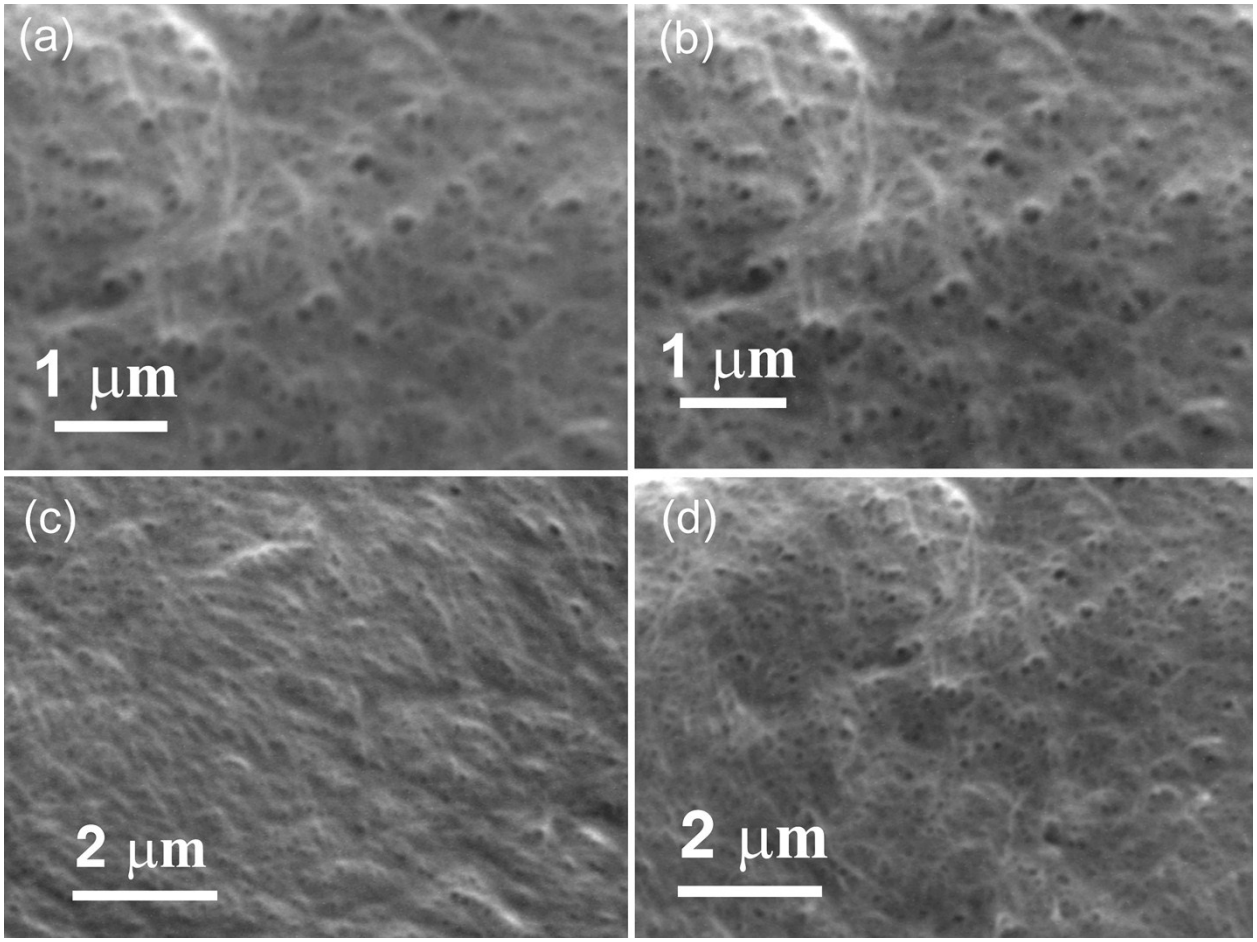


Figure S3: SEM images of dried self-assemblies prepared from a hydrogel of sodium betulinate **2** (0.71 % w/v).

Calculation of thermodynamic Parameters:

The thermoreversible melting of a gel can be expressed as:

Gel $\rightleftharpoons$ liquid

The equilibrium constant can be expressed as:

$$K = [Gelator]/ [Gel]$$

Assuming unit activity of the gel, the equilibrium constant can be expressed as:

$$K = [Gelator]$$

The Gibbs free energy change during gel melting can be expressed as:

$\Delta G° = - RT \ln K = \Delta H° – T\Delta S°$, Hence, $\ln K = -\Delta H°/R. (1/T) + \Delta S°/R$

Table S1: Thermodynamic parameters (ΔH^o, ΔS^o, ΔG^o) at 298 ºK of the sodium betulinate gel melting in different liquids.

Liquids	$\Delta H°$ kJ/mol	$\Delta S°$ J/mol/°K	$\Delta G°$ kJ/mol
DMSO-water	76.01	202.36	15.71
DMF-water	58.71	154.39	12.7

The gel melting temperature (T_{gel}) increases with increasing concentration of the "solutes". A plot of ln K vs 1/T allowed us to calculate the thermodynamic parameters. Representative plots for gel in DMSO-water (1:1v/v) and DMF-water (1:1v/v) for both **2** and **3** were given in the figure below:

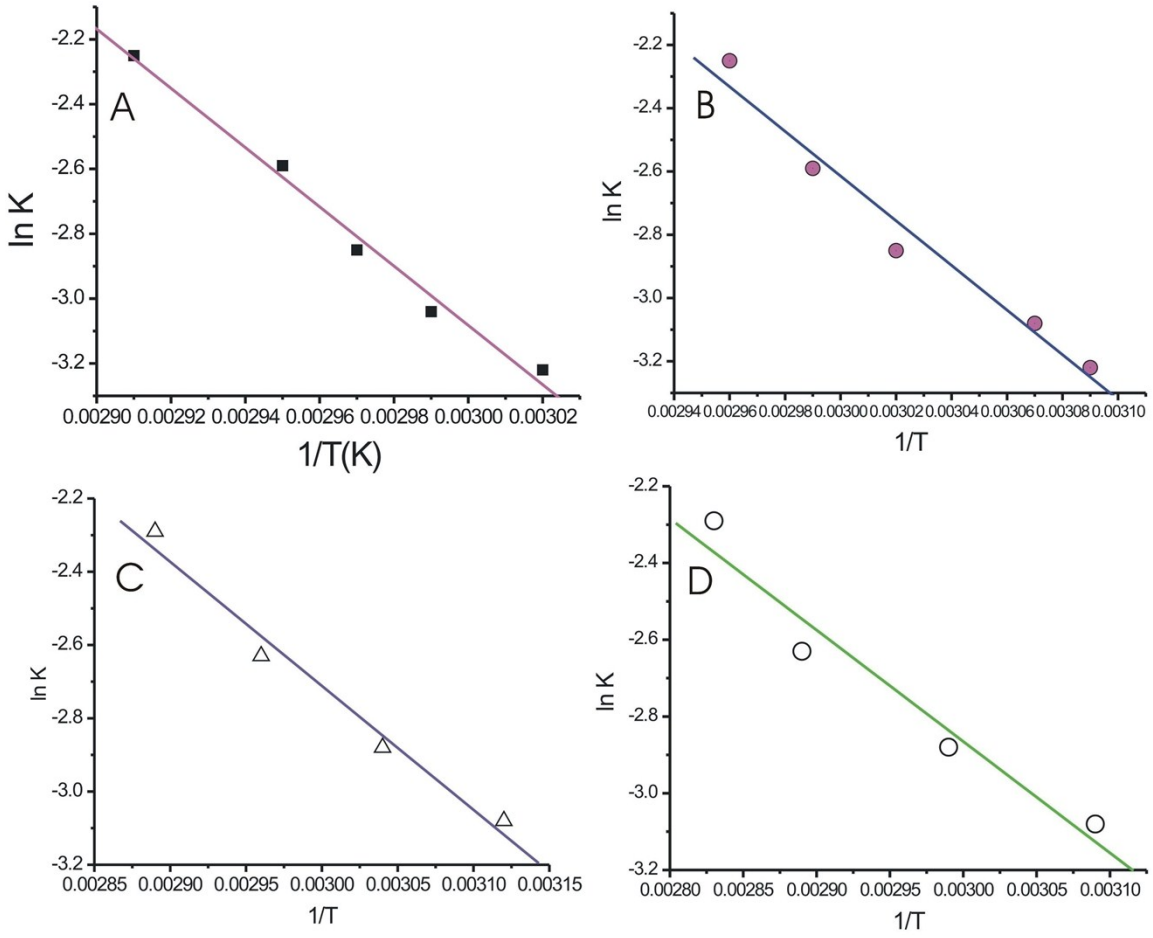


Figure S4: ln K vs 1/T (K) plot of (A) **2** in DMSO-water; (B) **2** in DMF-water; (C) **3** in DMSO-water and (D) **3** in DMF-water.

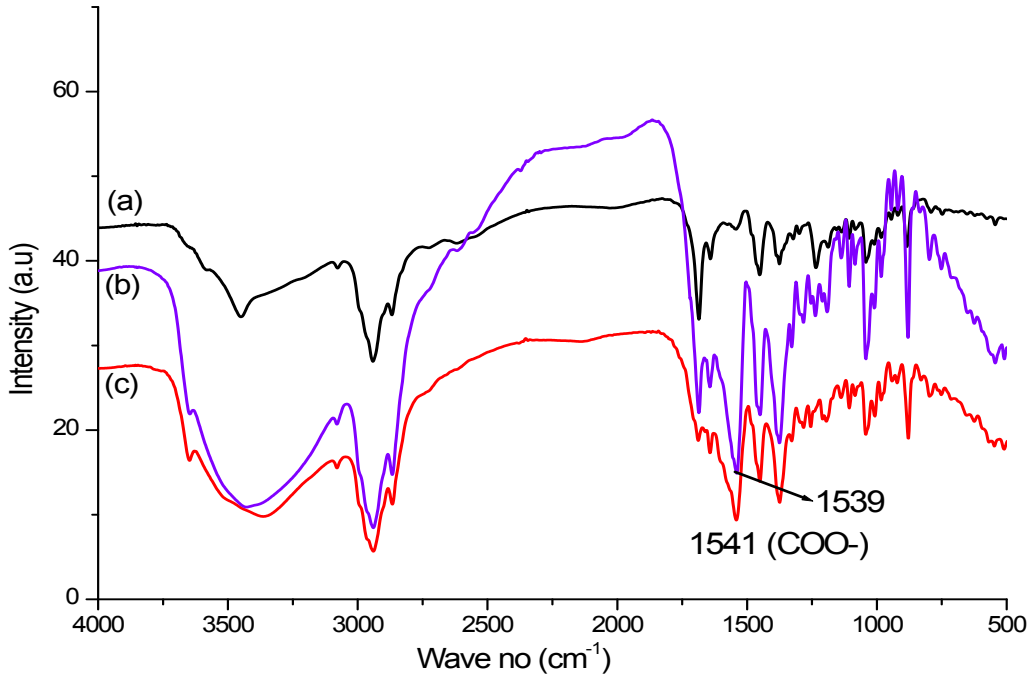


Figure S5: FTIR spectra (a) powder sample of betulinic acid **1**; (b) dried self-assemblies prepared from a hydrogel of **2** and (c) powder sample of **2**.

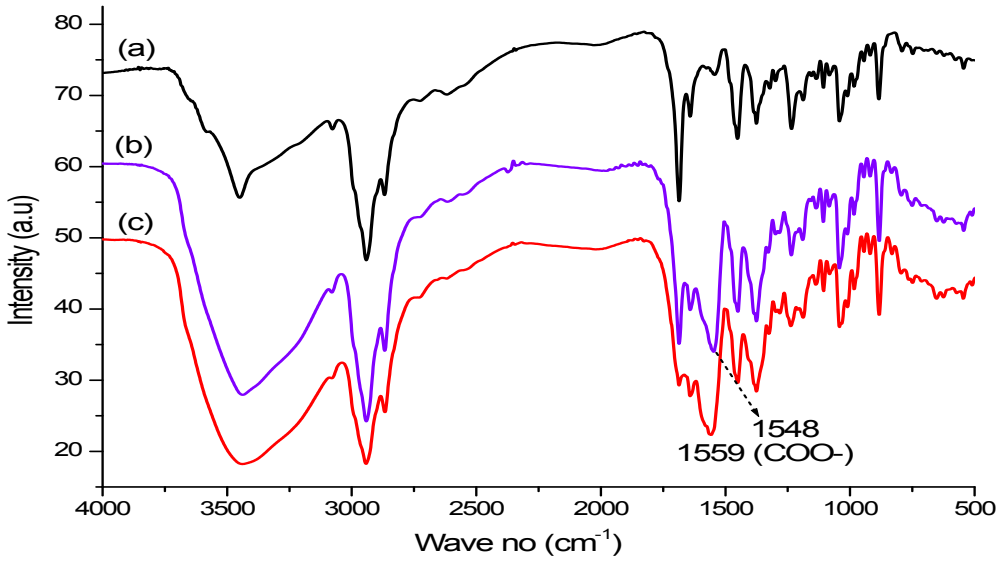


Figure S6: FTIR spectra (a) powder sample of betulinic acid **1**; (b) dried self-assemblies prepared from a hydrogel of **3** and (c) powder sample of **3**.

Epifluorescence Microscopy: The epifluorescence microscopy images revealed the entrapment of rhodamine B on the self-assemblies of potassium betulinate **3** in water.

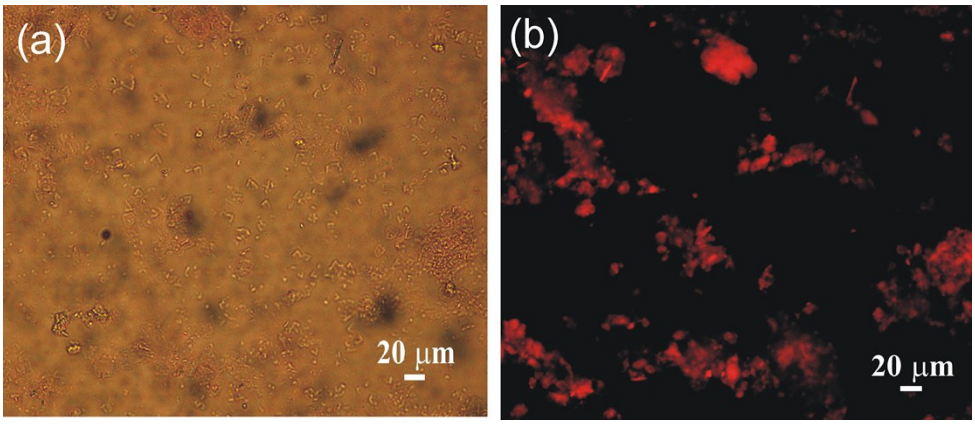


Figure S7: (a-b) Self-assemblies of potassium betulinate **3** loaded with rhodamine B in water: (a) under normal light; (b) under fluorescence light.

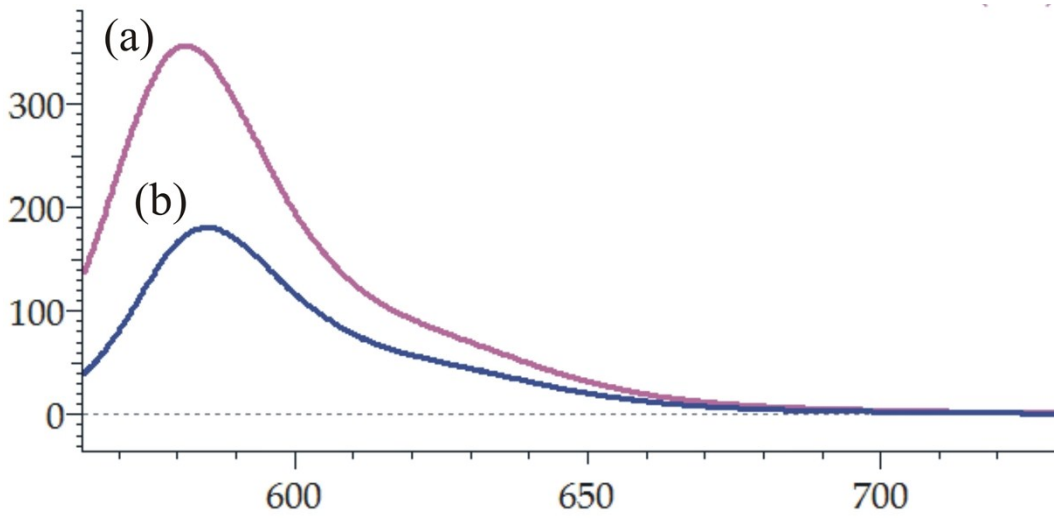


Figure S8: Fluorescence Emission Spectra: (a) rhodamine-B (c = 0.03 mM) in phosphate buffer (0.01 M, pH = 7.2); (b) in the presence of a hydrogel obtained from potassium betulinate **3** after 4 hours. Upon excitation at 554 nm (rhodamine B), emission spectra appeared at 581.4 nm. Decrease of fluorescence intensity with time indicated the removal of rhodamine-B dye from the aqueous buffer solution.

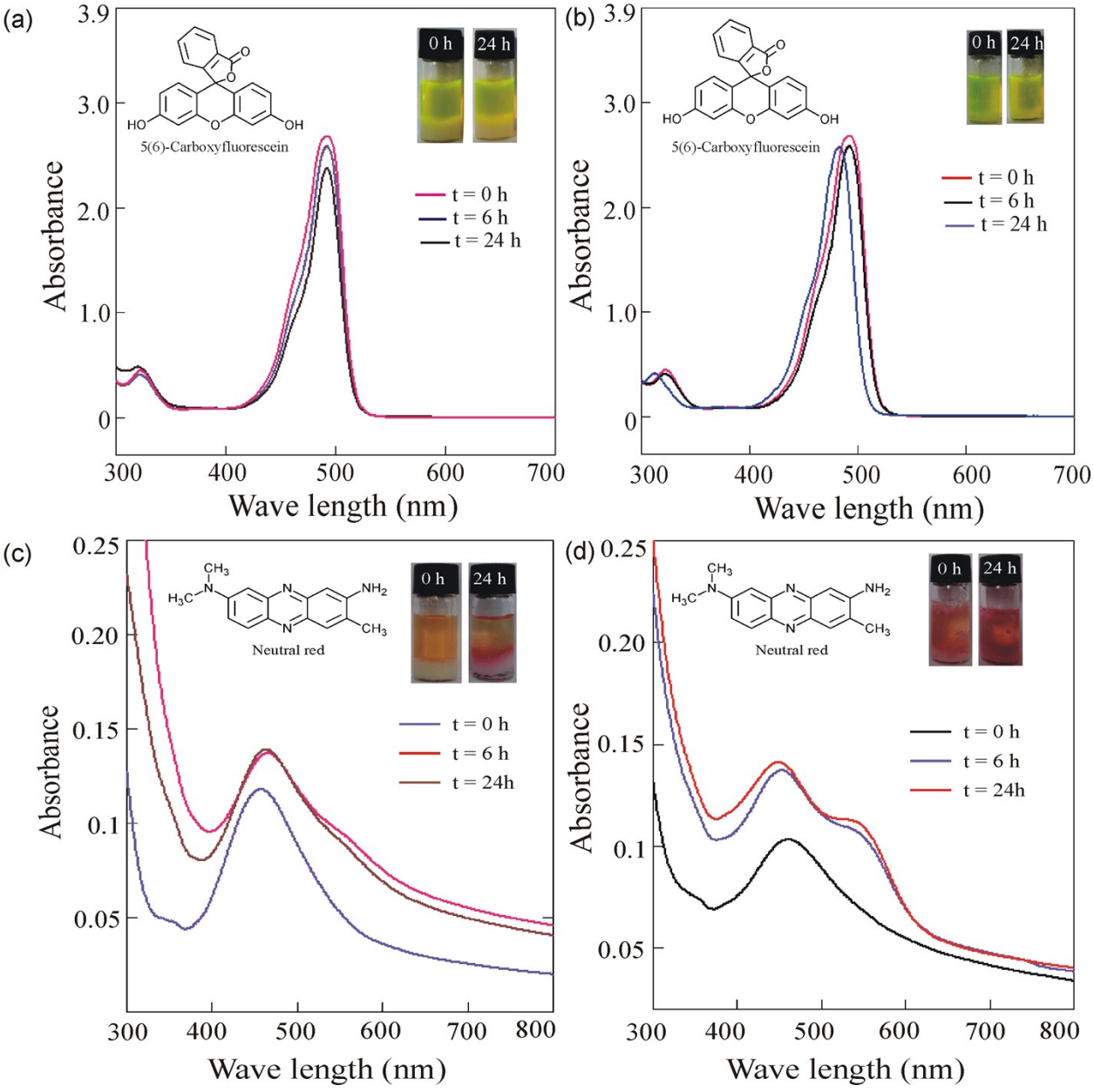
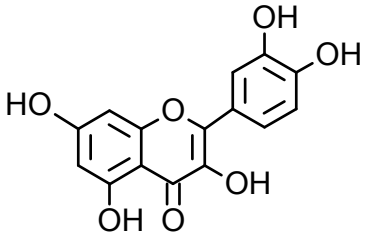
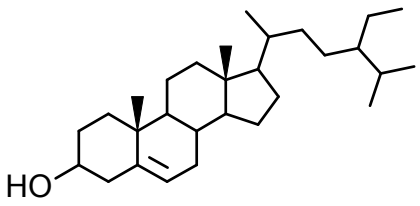


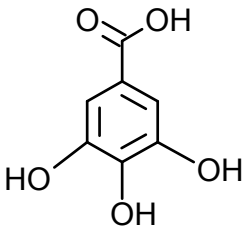
Figure S9: (a, b) UV−visible spectroscopy of CF solution kept in contact with the hydrogels of sodium betulinate **2** (2.5% w/v) and potassium betulinate **3** (2.5% w/v) respectively; (c, d) UV−visible spectroscopy of NR solution kept in contact with the hydrogels of sodium betulinate **2** (2.5% w/v) and potassium betulinate **3** (2.5% w/v) respectively, Inset: *left:* structure of the dye molecules, *right*: photograph of vials at different times.



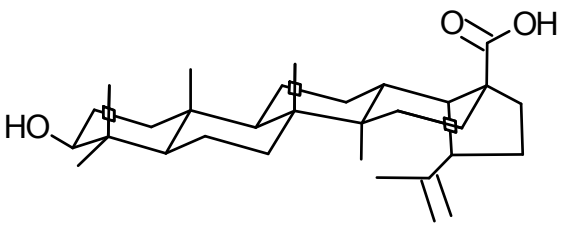
Quercetin



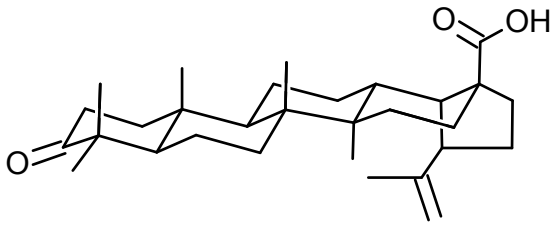
Beta-sitosterol



Gallic acid



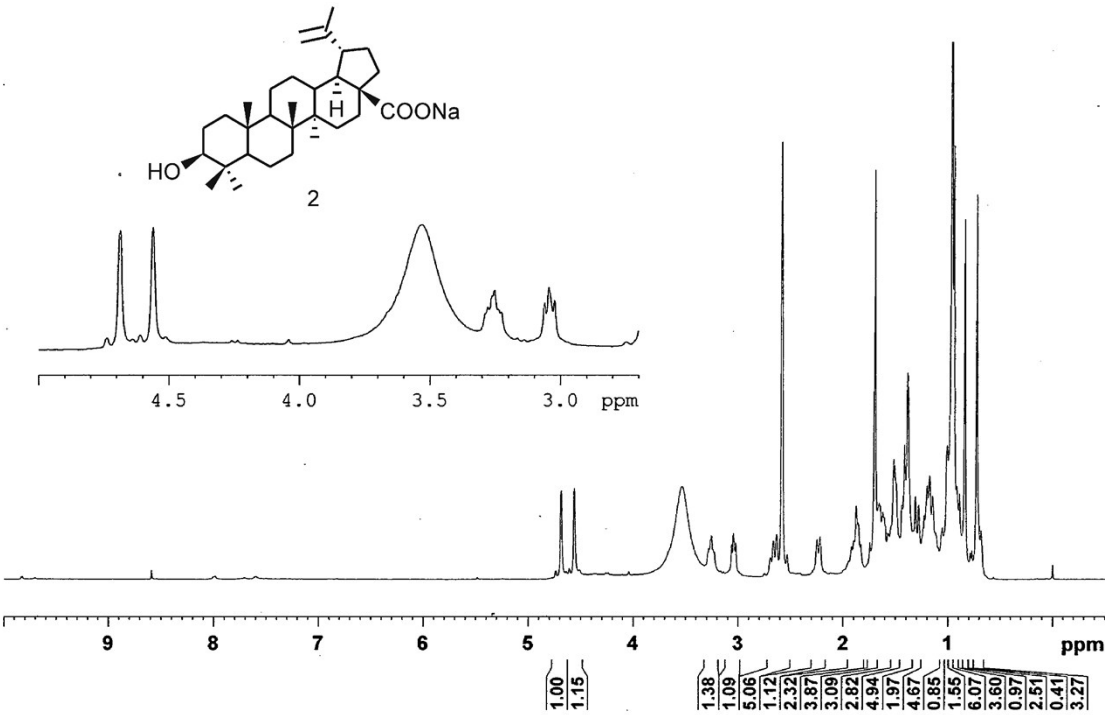
Betulinic acid



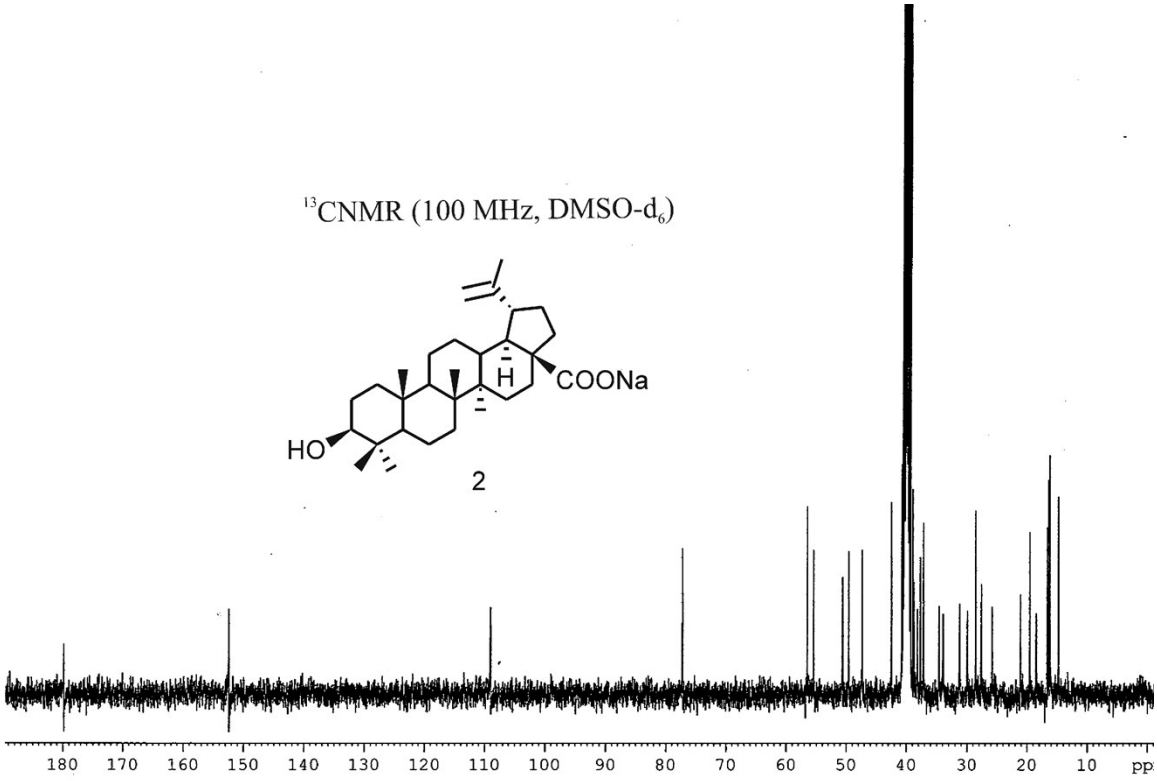
Betulonic acid

Figure S10: Probable structures of compounds present in the bark extract of *Ziziphus jujube,* as evident from the mass spectral analysis.

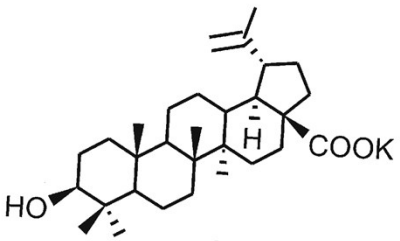
^{1}H-NMR (400 MHz, DMSO-d_6)



^{13}CNMR (100 MHz, DMSO-d_6)



^{1}H-NMR (400 MHz, DMSO-d_6)



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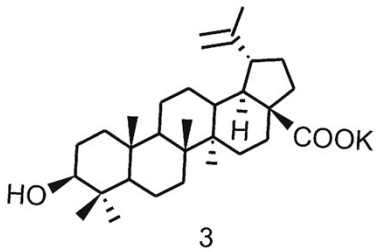
^{13}C-NMR (100 MHz, DMSO-d_6)



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