Supplementary Information
Light Harvesting Bi-component Hydrogel with Riboflavin Acceptor

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Preparation of gel:

Melamine (M), 6, 7-dimethoxy-2, 4[1H, 3H]-quinazolinedione (Q) and riboflavin (R) were purchased from Aldrich, USA and was used as received. The pure MQ hydrogel was prepared by dissolving M and Q in double distilled water in a sealed gel tube followed by addition of few drops of DMSO to increase the solubility of Q. They were heated to make a homogeneous solution which on cooling to 30°C produced hydrogels. The co-assembled hydrogels were prepared by following the same procedure mentioned above with the addition of desired amount of R (mole %).

Microscopy:

Fluorescence micrographs of the pure MQ and co-assembled MQ gel were taken by a fluorescence microscope (Olympus, BX61) by exciting the gel sample with UV radiation ($\lambda = 300$nm) using a FITC filter.

Wide-angle X-ray Diffraction study:

The wide-angle X-ray scattering (WAXS) experiments of the xerogel of MQ and MQ with R were performed using a high resolution X-Ray diffractometer model expert pro PANalytical. Samples were placed on glass slides and were scanned in the range of $2\theta = 4 - 60^0$.

Rheology:

To understand the mechanical property of the MQ gel rheological experiment was performed with an advanced rheometer (AR 2000, TA Instrument, USA) using cone plate geometry on a peltier plate. The diameter of the plate was 40nm and angle $4^0$ with plate gap of 121µm and frequency sweep was performed.

Spectroscopy:
The UV-vis spectra of the samples were recorded with a Hewlett-Packard UV-vis spectrophotometer (model 8453) using a cuvette of 0.1 cm path length. Fluorescence study of MQ and hybrid hydrogel samples prepared in a sealed cuvette were carried out in a Horiba Jobin Yvon Fluoromax 3 instrument. Each gel sample in a quartz cell of 1 cm path length was excited at 297 nm wavelength and emission scans were recorded from 320 to 700 nm using excitation slit width of 2 nm and emission slit is 5 nm with a 1 nm wavelength increment having an integration time of 0.1 s. Fluorescence lifetimes were measured by using a time-correlated single photon counting fluorometer (Fluorecule, Horiba Jobin Yvon). The system is excited with 295 nm nano LED of Horiba Jobin Yvon having $\lambda_{\text{max}}$ at 368 nm with pulse duration <200 pico second. All the samples are prepared for room temperature measurement (30°C) in double distilled water. Average fluorescence lifetimes ($\langle \tau_f \rangle$) for exponential iterative fitting are calculated from the decay times ($\tau_i$) and the relative amplitudes ($a_i$) using the following relation

\[
\langle \tau_f \rangle = a_1 \tau_1 + a_2 \tau_2 + a_3 \tau_3 \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (1)
\]

Where $a_1$, $a_2$, $a_3$ are relative amplitudes and $\tau_1$, $\tau_2$, $\tau_3$ are lifetime respectively.

The percentage of energy transfer $E_{\text{eff}}$ from MQ to R was calculated from the equation:

\[
E_{\text{eff}} = 1 - \frac{\tau_{DA}}{\tau_D} \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (2)
\]

Where $\tau_{DA}$ and $\tau_D$ are fluorescence lifetime of donor (Q) in presence and absence of acceptor (R), respectively. The energy transfer rate constant $K_{ET}$ can be calculated from the Stern-Volmer equation.

\[
\frac{I_0}{I} = 1 + K_{ET} \tau_0 [R] \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (3)
\]
Where $I_0$ and $I$ represent the fluorescence intensity of the donor in the absence and presence of acceptor respectively, $[R]$ is the molar acceptor concentration and $\tau_0$ is the fluorescence lifetime of the donor in the absence of acceptor.

**Determination of rate constant by Avrami equation:**

The Avrami equation\textsuperscript{13} is usually expressed as

$$1 - V(t) = \exp(-kt^n)$$

Where $V(t)$ is the fraction transformed, $k$ is the rate constant, $t$ is the time of transformation, and $n$ is a constant whose value depends on the nature of the nucleation and growth process. The fraction transformed can be obtained from the fluorescence data SI fig. X. If the intensity at $t = 0$ is $I_0$ and at $t = \infty$ is $I_\infty$, $V(t)$ is equal to $I_\infty - I_t / I_\infty - I_0$. Putting these values into equation 1, we obtain

$$I_\infty - I_t / I_\infty - I_0 = \exp(-kt^n)$$

Taking logarithms on both sides, the equation transforms to

$$\ln(-\ln I_\infty - I_t / I_\infty - I_0) = \ln k + n \ln t$$

Thus, by plotting the left-hand side of eq 2 with $\ln t$, straight lines are expected, and from the slope the Avrami exponent $n$ can be obtained. The rate constant $k$ of the process can be obtained from the intercept of the plot and for MQ gel $k$ has been found $8.4 \times 10^{-4}$ min$^{-1}$.

**Reference:**

Table S1: Fluorescence life time and percent energy transfer of MQ hydrogel with different mole % of R at 30° C.

<table>
<thead>
<tr>
<th>Hydrogel</th>
<th>Mole % of R</th>
<th>Av. life time (ns)</th>
<th>% of energy transfer</th>
</tr>
</thead>
<tbody>
<tr>
<td>MQ (0.5% (w/v))</td>
<td>0</td>
<td>16.01</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>0.25</td>
<td>5.25</td>
<td>67.2</td>
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<tr>
<td></td>
<td>0.50</td>
<td>4.99</td>
<td>68.9</td>
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<tr>
<td></td>
<td>0.71</td>
<td>3.77</td>
<td>76.6</td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>3.61</td>
<td>77.5</td>
</tr>
<tr>
<td></td>
<td>1.25</td>
<td>2.76</td>
<td>82.8</td>
</tr>
</tbody>
</table>
Fig.S1 Uv-vis spectra of pure Q and MQ hydrogel at 0.01% (v/w)
Fig. S2  Fluorescence spectrum of R on excitation at 373 nm.
Fig.S3 (a) pure MQ hydrogel, (b) under UV light ($\lambda = 300\text{nm}$) showing blue fluorescence emission, fig.3e Fluorescence micrograph of MQ gel and fig. 3(d) MQ gel with 1 mole % of R, fig. 3(e) under uv light ($\lambda_{\text{max}} = 300\text{nm}$) showing green fluorescence emission, fig. 3(f) Fluorescence micrograph of MQ gel with 1 mole % of R (scale bar 210 um) fig (g) and (h) are MQ xerogels in abscence and present of 1 mole % of R, fig (I) and (j) after irradiation of uv light ($\lambda_{\text{max}} = 300\text{nm}$).
Fig. S4 Solid state fluorescence spectra of intimate mixture of M and Q, mixed MQ with different mole % R dried from DCM solution excited at 297nm.
Fig. S5 WAXS patterns of xerogel MQ and MQ with 1 mole % R.
Fig. S6 (a) Time dependent fluorescence spectra of MQ hydrogel at 30 °C (Ex = 297nm). (b) Plot of fluorescence intensity vs time at 25 °C (c) Avrami plot of the fluorescence intensity data of Figure b of MQ gel.
Fig S7 The fluorescence intensity of donor MQ (10^{-3} M) decreases while that of R increases at solution phase with addition of acceptor R (10^{-3} M).
Fig. S8a Temperature dependent fluorescence spectra of MQ gel in presence of 1 mole % R.
Fig. S8b Variation of fluorescence intensity of donor and acceptor (1 mole % R) with temperature.
Fig. S8c Variation of fluorescence wavelength maxima of donor and acceptor (1 mole % R) with temperature.
Fig. S9 (a) Time dependent fluorescence spectra of MQ gel with 1 mole \% R excited at 297nm. (b) intensity vs time plot (sigmoidal fit).
Fig.S10  pH dependent fluorescence spectra of MQ gel in presence of 1 mole % R.
Fig.S11 Storage modulus vs different mole % of R plot. Inset A represents a plot of modulus (G', G'') vs. angular frequency plot of MQ gel in presence of 1 mole % of R.