ESI 1.



Wavelength / nm

ESI 1. Absorption spectra of the R6G-PVP-SA films; R6G amounts are 0.1% of CEC of SA and the weight ratio of the added PVP amounts to SA are 0 (a), 0.25 (b), 1 (c), 1.5 (d) and 2.5 (e).

ESI 2.



Wavelength / nm

ESI 2. Absorption spectra of the R6G-PVP-SA films; R6G amounts are 0.3% of CEC of SA and the weight ratio of the added PVP amounts to SA are 0 (a), 0.25 (b), 1 (c), 1.5 (d)- and 2.5 (e)-.

ESI 3.



Wavelength / nm

ESI 3. Absorption spectra of the R6G-PVP-SA films; R6G amounts are 0.6% of CEC of SA and the weight ratio of the added PVP amounts to SA are 0 (a), 0.25 (b), 1 (c), 1.5 (d) and 2.5(e)-.

ESI 4.



ESI 4. Photoluminescence spectra of the R6G-PVP-SA films; R6G amounts are 0.1% of CEC of SA and the weight ratio of the added PVP amounts to SA are 0 (a), 0.25 (b), 1 (c), 1.5 (d) and 2.5 (e).

ESI 5.



ESI 5. Photoluminescence spectra of the R6G-PVP-SA films; R6G amounts are 0.3% of CEC of SA and the weight ratio of the added PVP amounts to SA are 0 (a), 0.25 (b), 1 (c), 1.5 (d) and 2.5 (e).

ESI 6.



ESI 6. Photoluminescence spectra of the R6G-PVP-SA films; R6G amounts are 0.6% of CEC of SA and the weight ratio of the added PVP amounts to SA are 0 (a), 0.25 (b), 1 (c), 1.5 (d) and 2.5 (e).

## ESI 7.

Followings are possible analyses and explanations for the changes of absorption and photoluminescence spectra.

a) The ratios of the absorbance at 540 nm to that at 500 nm are 0.3 - 0.4 for all the R6G-PVP-SA films but the sample with no PVP (0.5-0.6). This suggests that R6G in the R6G-PVP-SA films (weight ratio of PVP to SA > 0) were in the similar condition. In addition, when the PVP: SA ratio are high (> 1), the emission band at around 560 nm is dominant as shown in Fig. 7 in the revised manuscript. When the ratios are low (0 or 0.25), on the other hand, the emission band centered at around 580 nm is dominant. These differences of absorption and emission spectra as a function of the PVP contents can be attributed to the dye aggregation; the dye tends to aggregate at low PVP contents (0 or 0.25).

b) All the absorption spectra of the R6G-PVP-SA films were separated into four absorption bands with the absorption maxima at 555±3, 538±3, 505±3 and 480±5 nm, irrespective of the R6G amount and the PVP amount (see below figure), by the deconvolutions of absorption spectra carried out by applying the nonlinear least mean squares method (Levenberg-Marquardt algorithm). With the increase of the concentration of R6G in the interlayer nanospace, which is determined by the amount of PVP and R6G, the absorption maxima shifted to a lower wavelength region (Fig. 6 inset in the revised manuscript). This suggests that the absorption band at ca. 555 nm is ascribable to the aggregation of R6G. The other absorption bands (at 540, 505 and 480 nm) are ascribable to the R6G monomer. The excitation wavelength of 490 nm is appropriate for the discussion because the contribution of the R6G aggregate is less probable. When the weight ratio of PVP to SA ratio are low (0 or 0.25), the emission band centered at around 580 nm is dominant, while when the weight ratio of PVP to SA ratio are high (> 1), the emission band at around 560 nm is dominant as shown in Fig. 7 in the revised manuscript. At low PVP loadings (PVP: SA ratio = 0 or 0.25), the distance between adjacent R6G is close to affect the state each other. In addition, the interactions of R6G with silicate laver play a role for the difference in the emission band.



Representative absorption spectrum with the separated absorption bands of R6G-PVP-SA film; R6G amount is 1% of CEC of SA and the weight ratio of the added PVP amount to SA is 0. The red line indicates an original spectrum.

ESI 8.



ESI 8. Absorption spectra of R6G-PVP film under artificial sunlight irradiation; 0 h (a), 0.5 h (b), 1 h (c), 1.5 h (d), 2 h (e) and 3 h (f). The R6G content is 0.142  $\mu$ mol (1.42  $\mu$ mol (g of PVP)<sup>-1</sup>) and the weight ratio of the added PVP amounts to is 5.

ESI 9.



ESI 9. Absorption spectra of R6G-SA film under artificial sunlight irradiation; 0 h (a), 0.5 h (b), 1 h (c), 1.5 h (d), 2 h (e) and 3 h (f). The R6G content is 0.142  $\mu$ mol (1% of CEC of SA).

ESI 10.



ESI 10. Absorption spectra of R6G-PVP-SA film under artificial sunlight irradiation; 0 h (a), 0.5 h (b), 1 h (c), 1.5 h (d), 2 h (e) and 3 h (f). The R6G content is 1.42  $\mu$ mol (g of PVP)<sup>-1</sup> (1% of CEC of SA) and the weight ratio of the added PVP amounts to SA = 5.