Supporting Information

Long-Term Stable 2H-MoS_2 Dispersion: Critical Role of Solvent for Simultaneous Phase Restoration and Surface Functionalization of Liquid-Exfoliated MoS_2

Dong Min Sim,‡ Hyeuk Jin Han,‡ Soonmin Yim, Min-Jae Choi, Jaebeom Jeon and Yeon Sik Jung*

Department of Materials Science and Engineering, Korea Advanced Institute of Science and Technology (KAIST) 291 Daehak-ro, Yuseong-gu, Daejeon 34141, Republic of Korea.

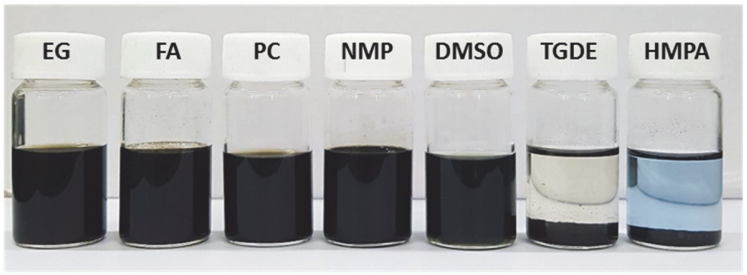
Corresponding Author

* E-mail: ysjung@kaist.ac.kr

‡ D.M.S. and H.J.H. contributed equally.

Figure S1. (a) Photographs showing the dispersion stability of ce-MoS_2 in polar solvents after 1 week. (b) Boiling point and surface tension data of polar solvents used in this study

(a)



(b)

	1T-MoS_2 (water)	**Ethylene glycol (EG)**	**Formamide (FA)**	**Propylene carbonate (PC)**	**NMP**	**DMSO**	**TGDE**	**HMPA**
b.p	100 ℃	197.3 ℃	210 ℃	240 ℃	202 ℃	189 ℃	275 ℃	232.5 ℃
Surface tension [dyne/cm]	72.3	43.4	57.0	40	40.9	42.9	34.3	30.1
Solubility	O	O	O	O	O	O	X	X

Table S1. The molar ratio of 1T-MoS_2, 2H-MoS_2, and MoO_3 calculated from the Mo 3d and S 2p XPS.

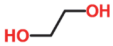
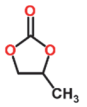
Solvent						
	1T-MoS_2 (water)	Ethylene glycol	Formamide	NMP	Propylene carbonate	DMSO
Mo^{+4} (1T)	62.38 %	17.91 %	19.11 %	17.62 %	16.80 %	19.05 %
Mo^{+4} (2H)	26.27 %	70.61 %	68.04 %	75.18 %	71.98 %	72.02 %
Mo^{+6} (MoO_3)	11.35 %	11.47 %	12.85 %	7.20 %	11.22 %	8.93
2H/(1T+2H) - Mo base	29.63	79.76	78.07	81.01	67.96	69.01
S^{-2} (1T)	78.75 %	14.96 %	14.34 %	12.88 %	15.44 %	13.85 %
S^{-2} (2H)	21.25 %	85.04 %	85.66 %	87.12%	84.56 %	86.15 %

Figure S2. High-resolution O 1s XPS spectra for ce-MoS_2, MoS_2-NMP, MoS_2-PC, MoS_2-FA, MoS_2-EG, and MoS_2-DMSO.

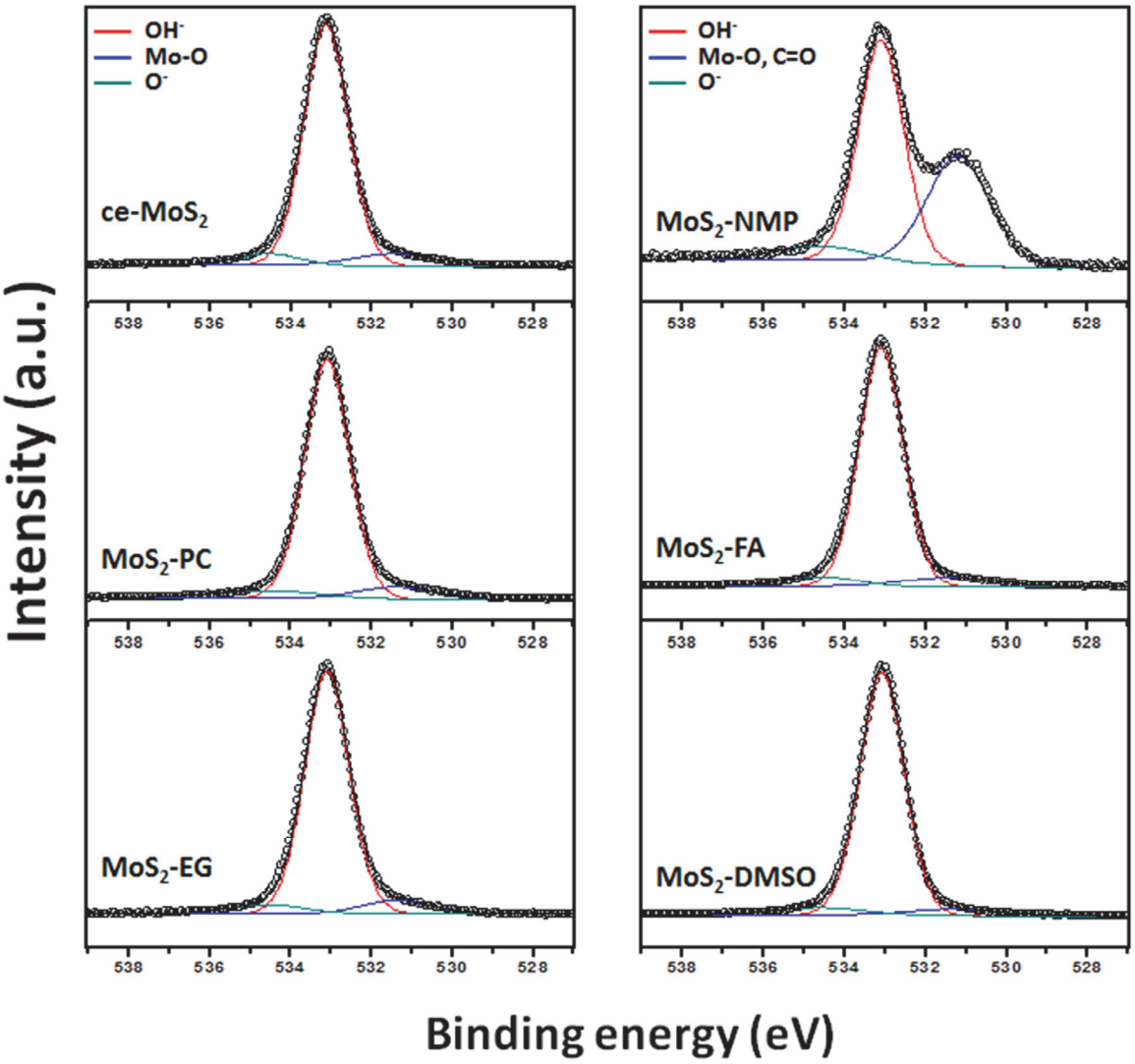


Figure S3. (a) S 2p XPS spectra of as-prepared ce-MoS_2 and after solvent thermal treatment in polar solvents at 180℃ for 5h. Red and blue lines represent the 2H-MoS_2 and 1T-MoS_2, respectively. (b) Calculated fraction of 2H phase after solvent thermal treatment in each polar solvent from the S 2p XPS analysis. The calculated phase transition yield is about 85%.

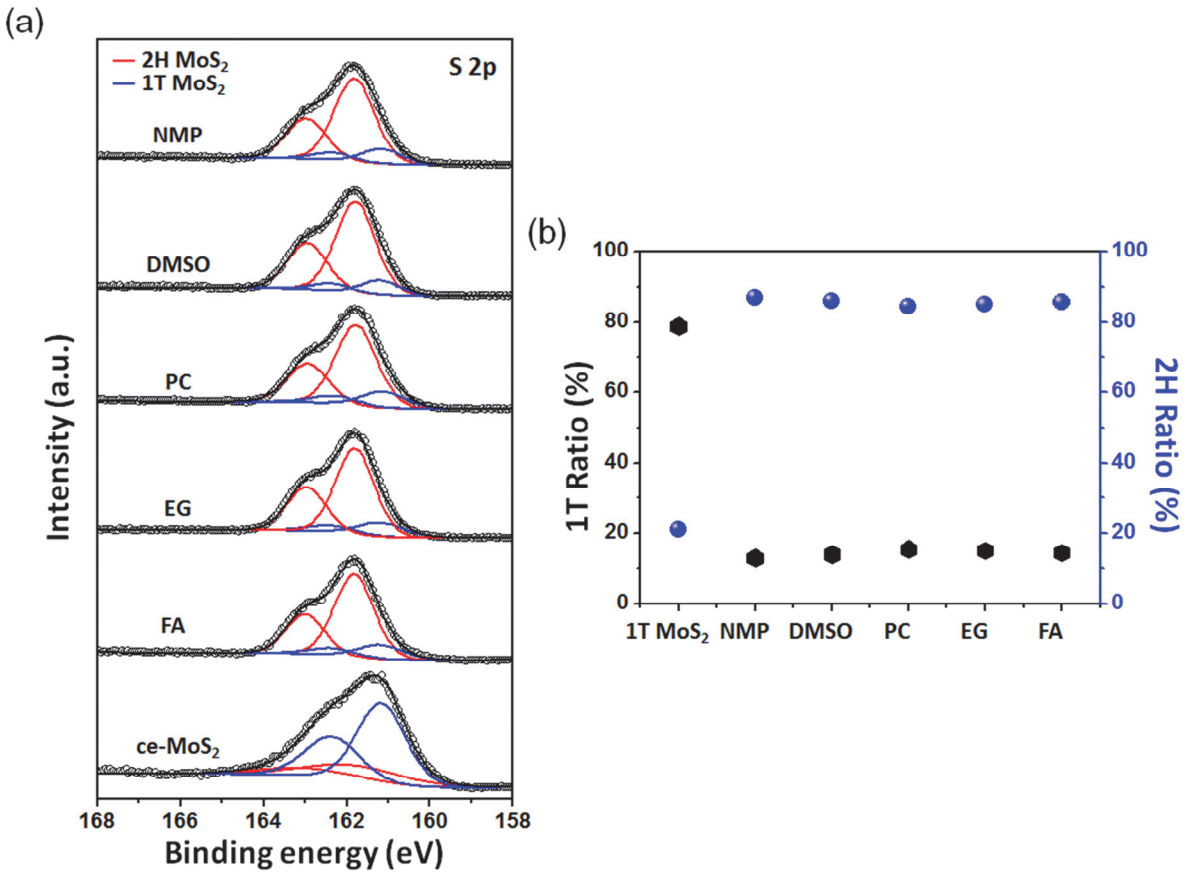


Figure S4. Raman spectra for (a) ce-MoS_2 and (b) MoS_2-NMP on Si substrate.

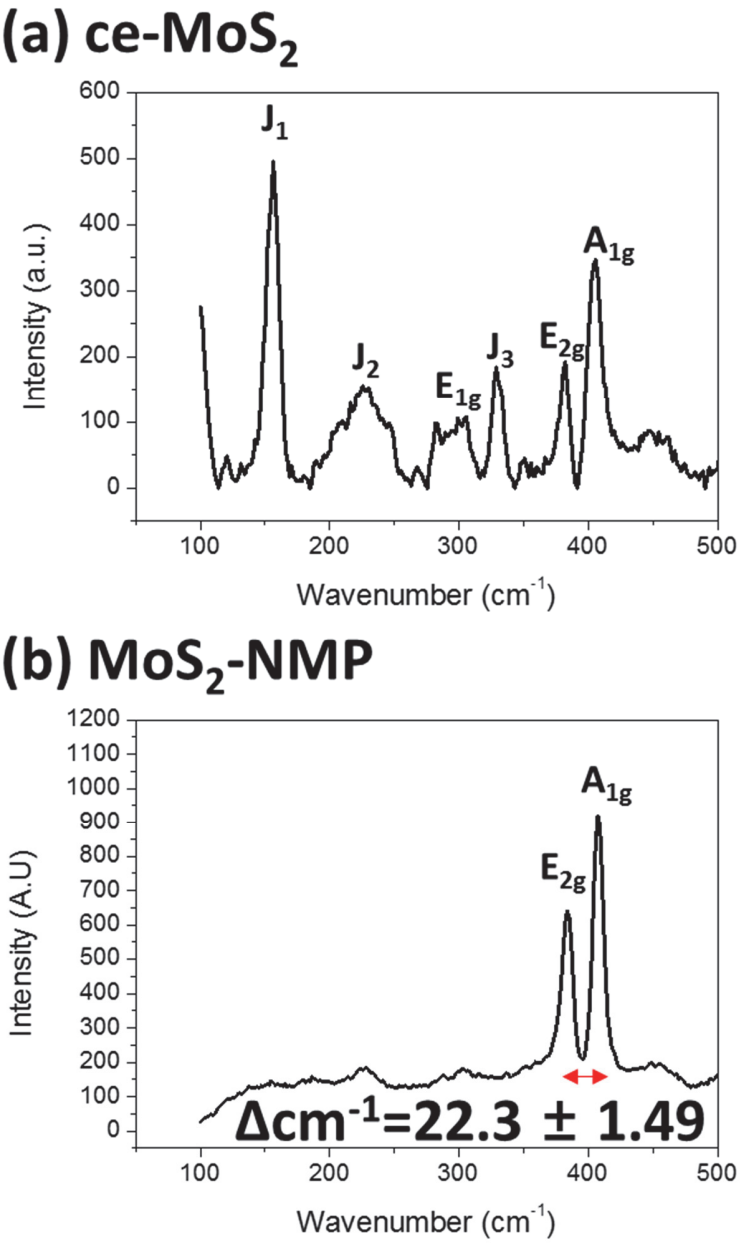


Table S2. Positions of A and B exciton peaks in the optical absorption spectra of 2H-MoS_2 after solvent thermal treatment. (In monolayer MoS2, A exciton – 1.887 eV (657.01nm), B exciton – 2.042 eV (607.77nm)

Solvent	A exciton [nm]	B exciton [nm]	Δ A exciton (based on NMP)
1T	-	-	-
EG	660.2	611.6	+3.2
FA	659	611.2	+2
PC	658	609.4	+1
DMSO	659.8	610.6	+2.8
NMP	657	607.4	-

Figure S5. (a) AFM height topography of MoS_2-NMP flakes on Si substrate. (b) Height profile along the black and green lines in Figure S5a. (c) Height histogram and (d) diameter histogram of 50 different MoS_2-NMP flakes.

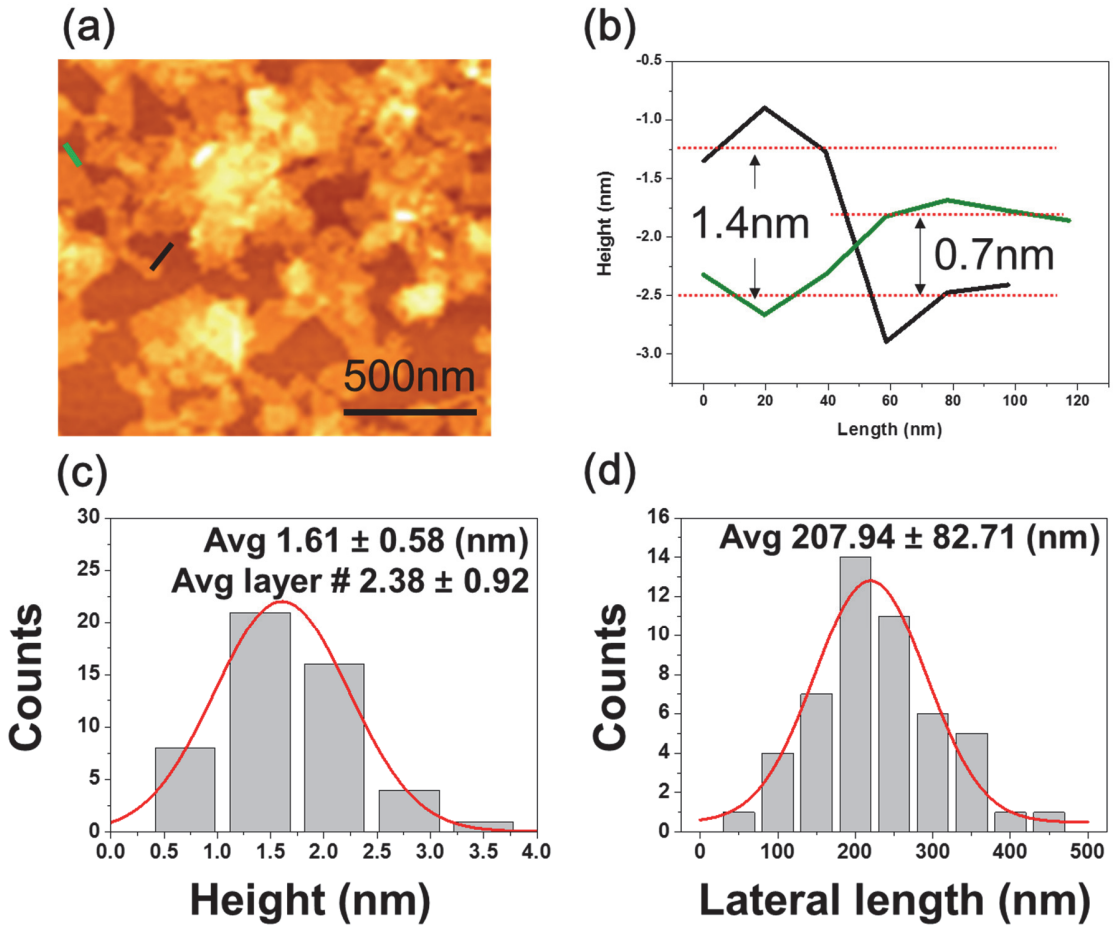


Figure S6. (a) AFM height topography of MoS_2-NMP flakes on Si substrate, which were stored for 10 months. (b) Diameter histogram of 50 different MoS_2-NMP flakes stored for 10 months.

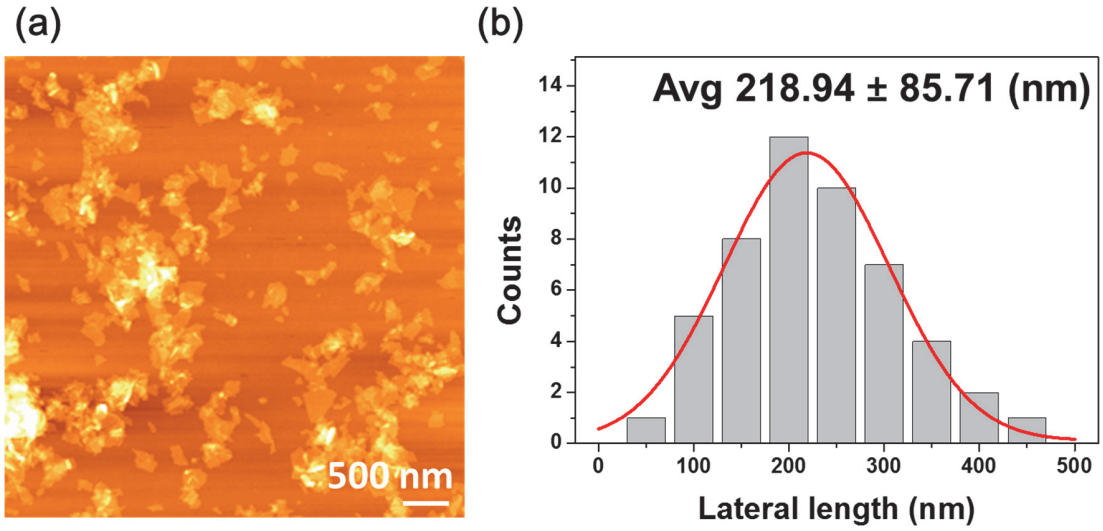


Figure S7. Photographs of pristine NMP and solvent-thermal-treated NMP at 180℃ for 5h.

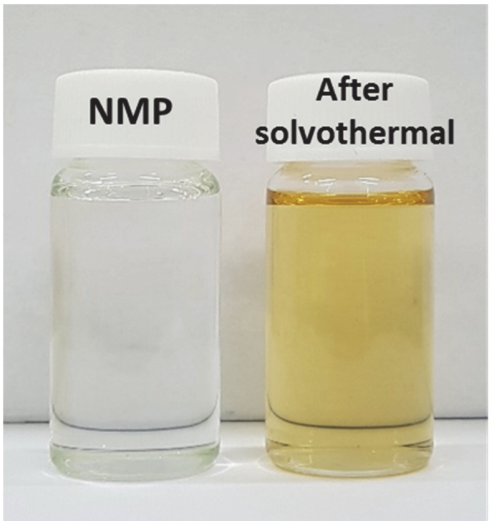


Figure S8. High-resolution C 1s XPS spectra for ce-MoS_2, MoS_2-NMP, MoS_2-PC, MoS_2-FA, MoS_2-EG, and MoS_2-DMSO.

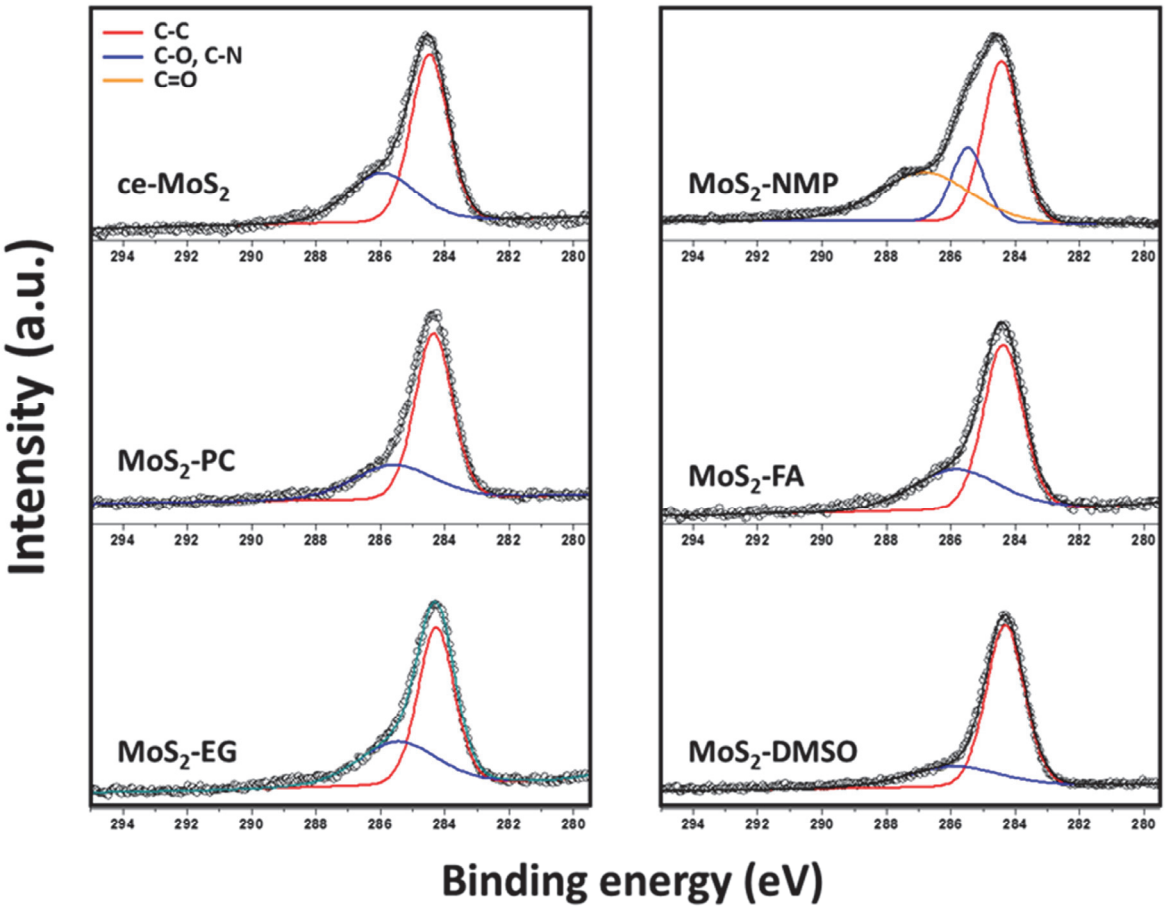


Figure S9. FT-IR spectra of the ce-MoS_2, MoS_2-NMP, and pure acetamide.

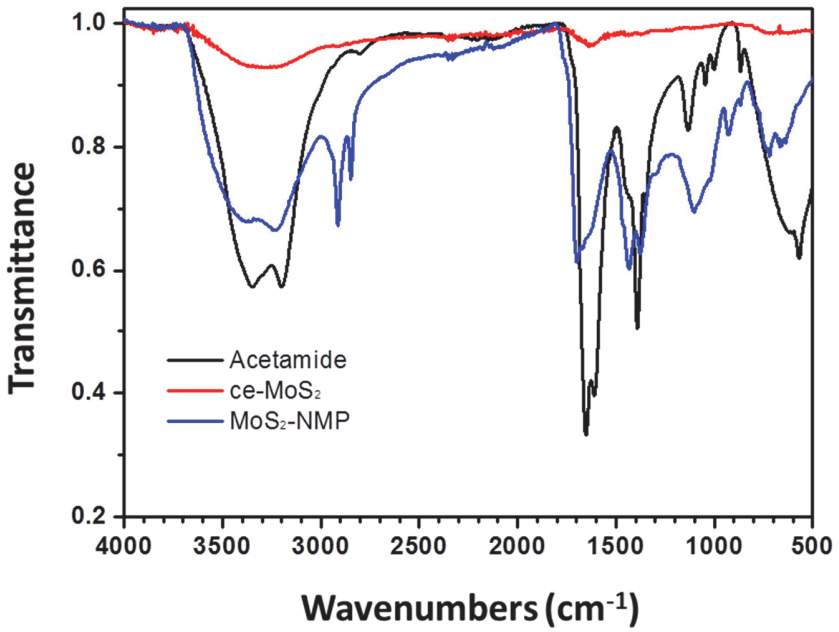
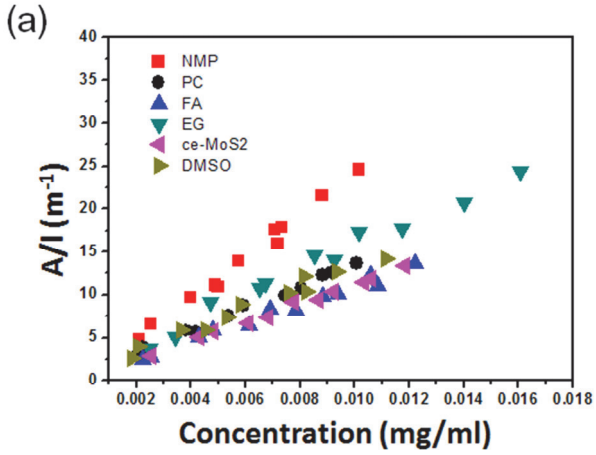


Figure S10. (a) Lambert-Beer plots to obtain the extinction coefficient value. (b) The extracted extinction coefficient values of as-prepared ce-MoS2 and 2H-MoS_2 samples obtained by solvent thermal treatment (c) Concentration of 2H-MoS_2 in the supernatant of each dispersion as a function of storage time.

(a)



(b)

	Slope (ml/mg/m)
NMP	2441.54 ±82.21
PC	1299.24±35.55
Formamide	1079.02±43.15
EG	1465.23±68.09
DMSO	1232.93±69.02
ce-MoS_2	1106.64±26.46

(c)

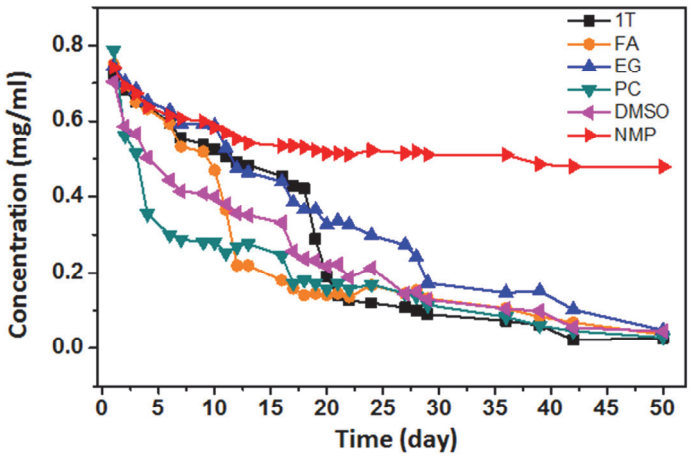


Table S3. Calculated concentration of MoS_2 in the supernatant of the aqueous dispersion as prepared and after 50 days.

	Concentration(mg/ml)	
	0 days	After 50 days
NMP	0.74219	0.47871
PC	0.74642	0.02966
FA	0.75070	0.03722
EG	0.78816	0.04874
DMSO	0.70338	0.04584
1T	0.72256	0.02731

Table S4. Zeta potential values of pristine 2H-MoS_2, exfoliated 1T-MoS_2 and functionalized MoS_2 (NMP)

Entry	Zeta potential (mV)
2H-MoS_2 (bulk)	-20.2±0.48
1T-MoS_2 (Exfoliated MoS_2)	-43.1±0.64
MoS_2-NMP	-41.2±0.72

Figure S11. (a) Photographs of water contact angle taken after 2 s after dropping 1.5 ul water on prepared MoS_2 thin films. (b) Sensitivity of humidity sensor based on ce-MoS_2, MoS_2-EG, and MoS_2-NMP at 80 relative humidity (RH) %