

Supporting information

Self-cleaning and fully polymer-based super moisture-resistant gas barrier coating films with 2D polymer for flexible electronic devices and packaging applications

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1. Synthesis and characterization of 2D polymer

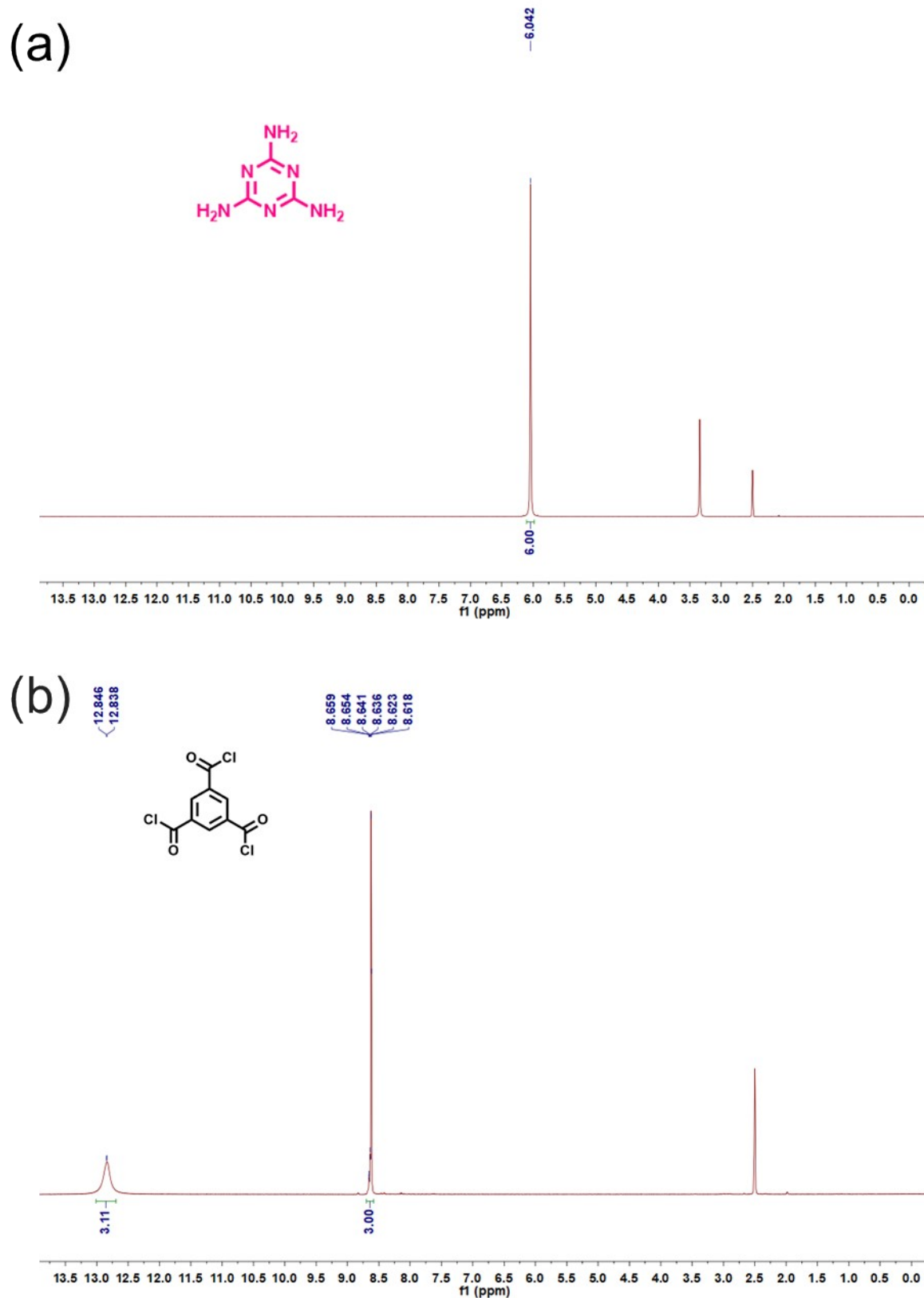


Fig. S1 ^1H NMR spectrum (500 MHz, 298K, DMSO-d_6) of monomers (a) melamine and (b) trimesic acid trichloride.

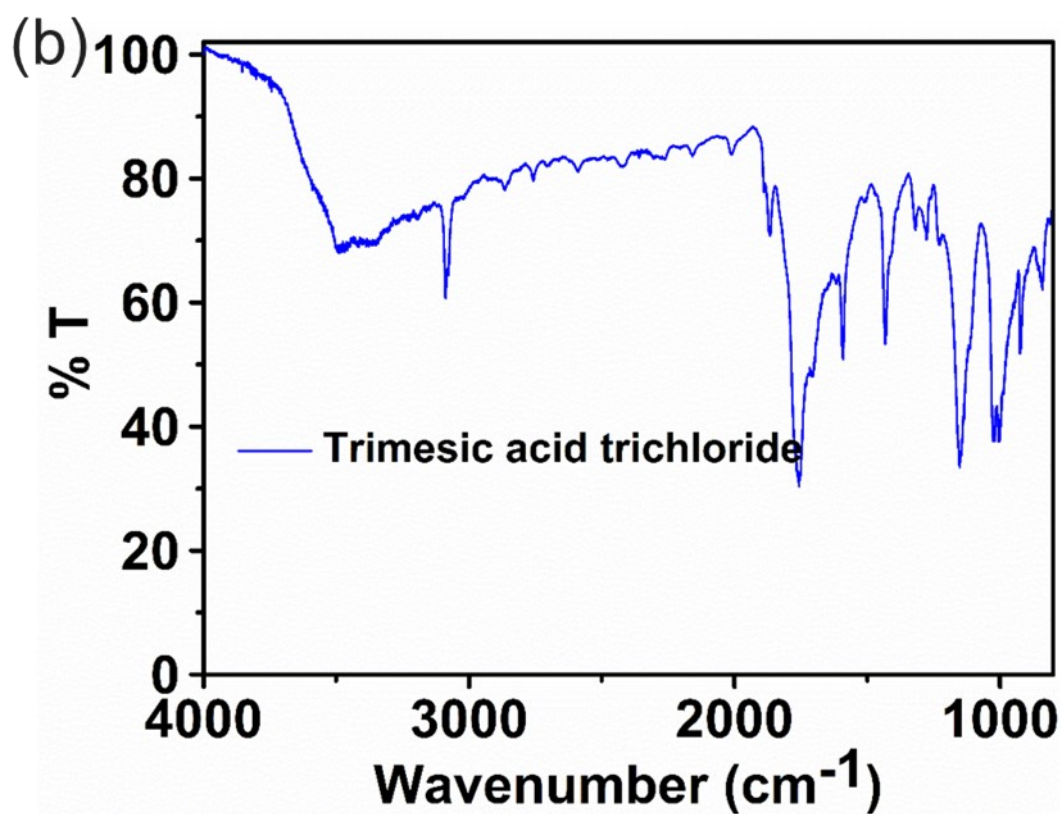
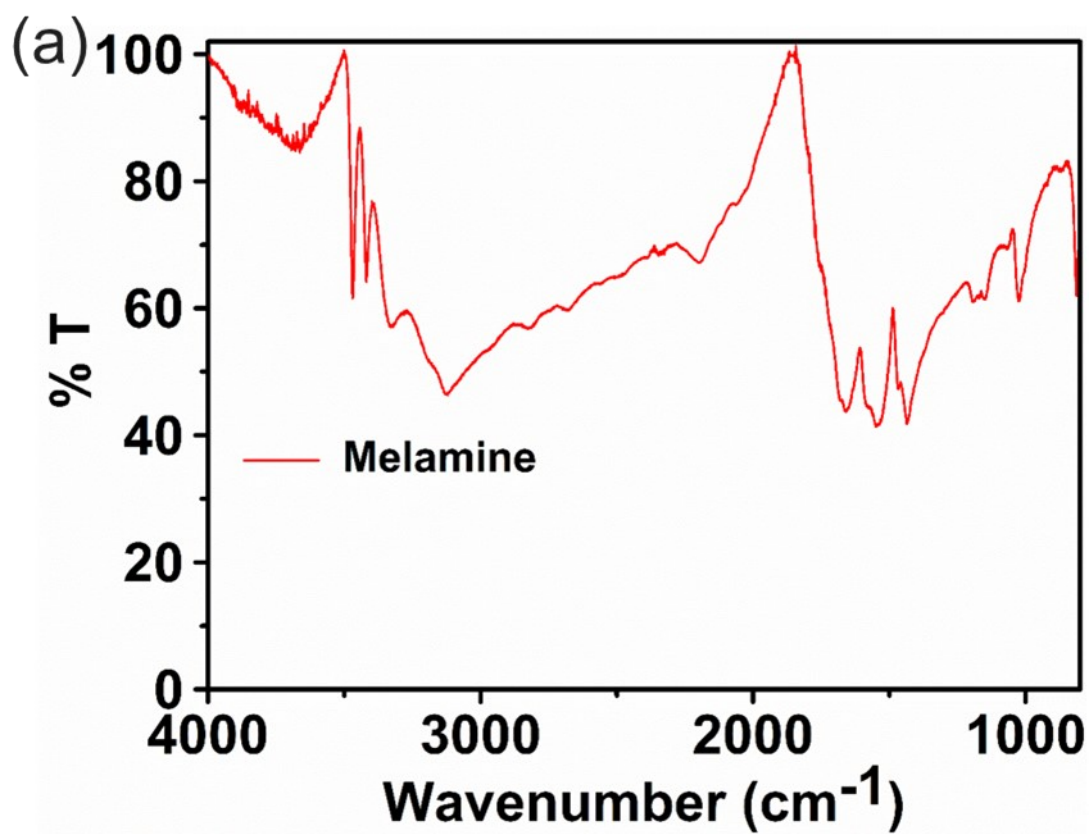


Fig. S2 FTIR spectrum of monomers (a) melamine and (b) trimesic acid trichloride.

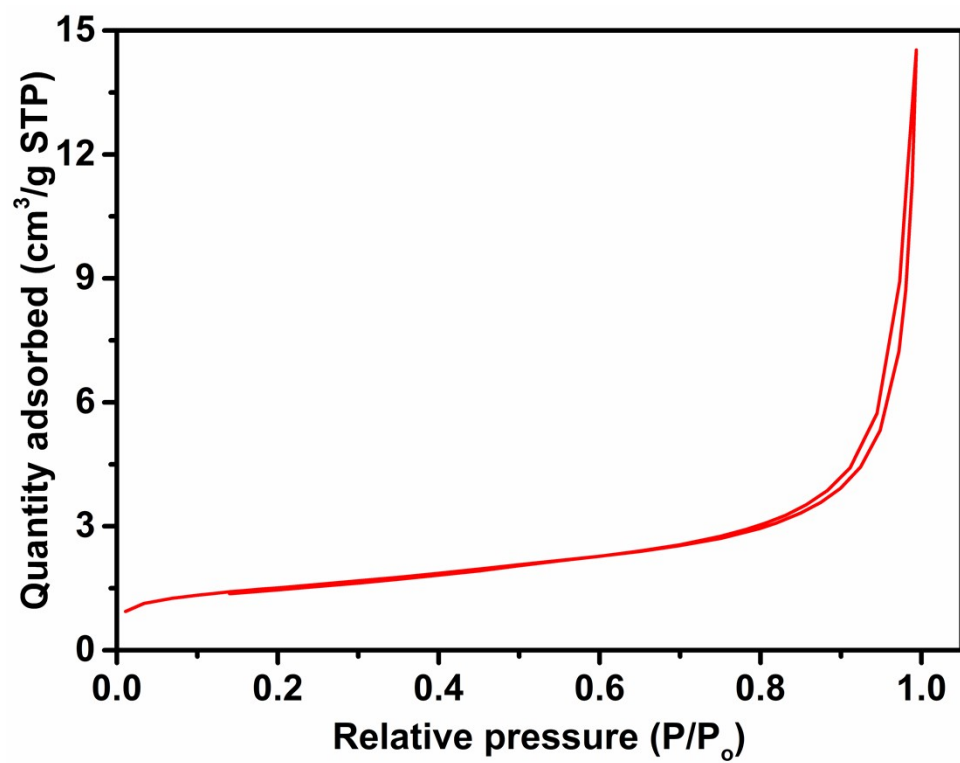


Fig. S3 BET adsorption-desorption curves of 2D polymer.

2. Characterization of 2D polymer and (BL)_n films

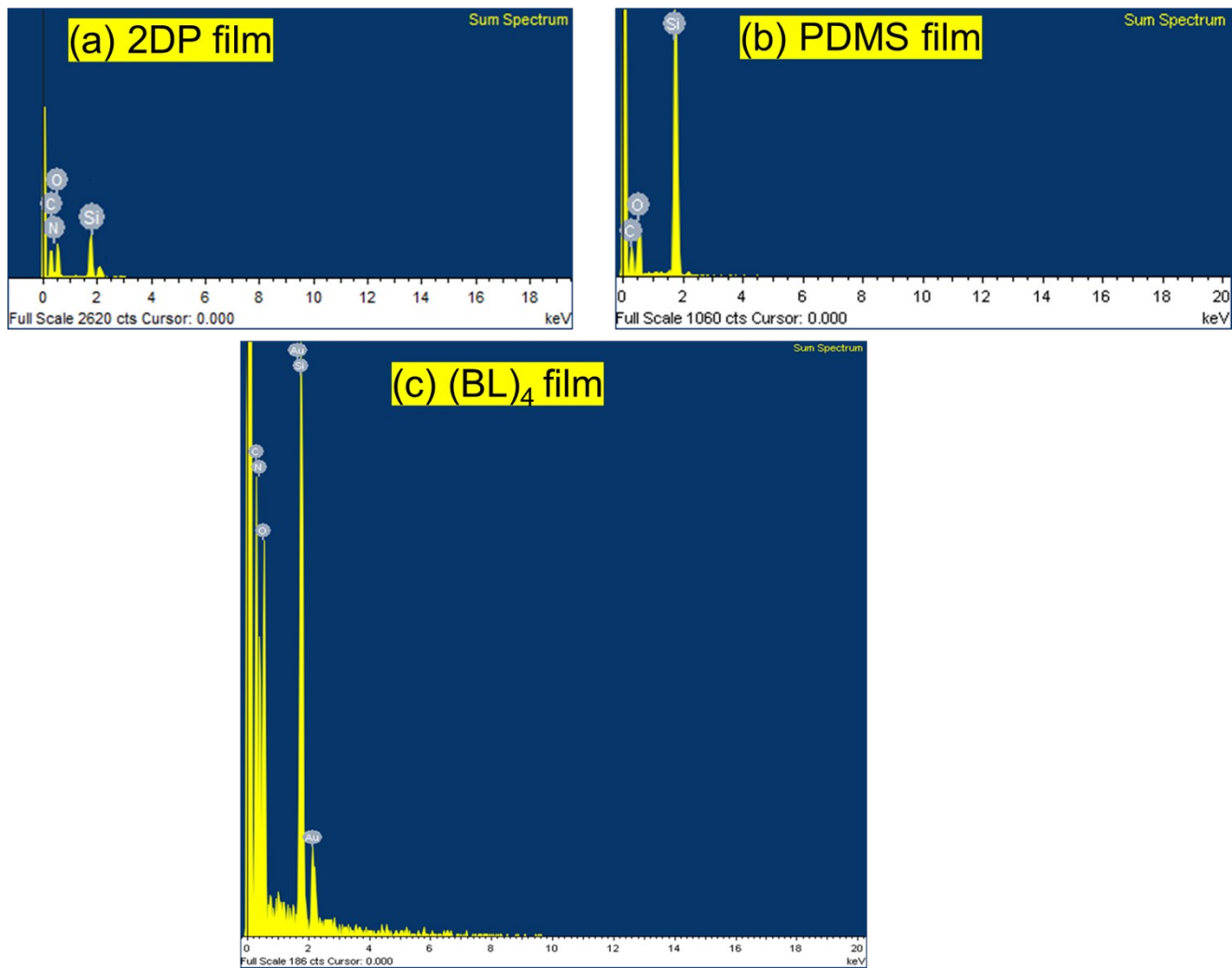


Fig. S4 EDX profile of (a) 2DP, (b) PDMS, and (c) (BL)₄ films.

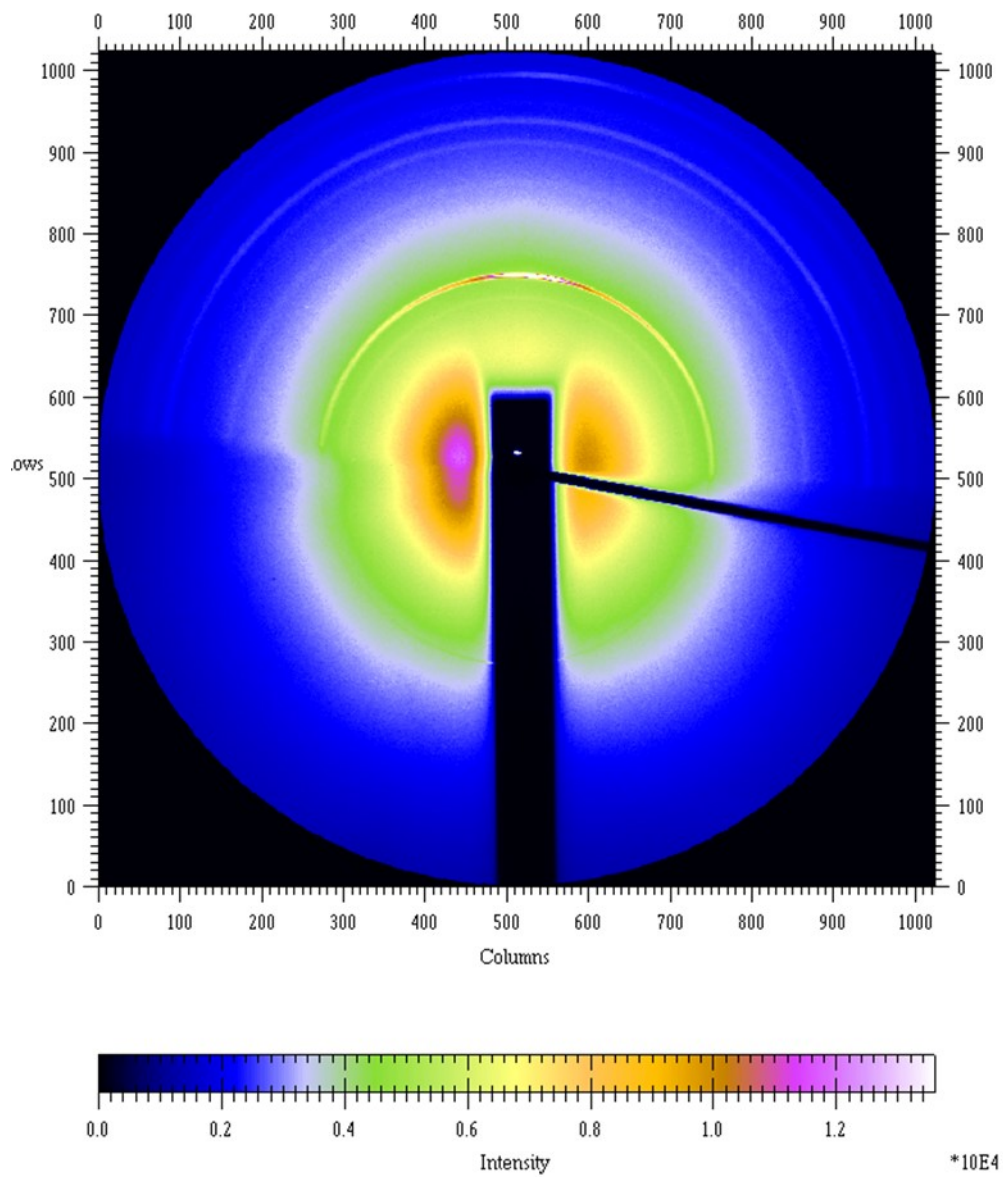


Fig. S5 2D profile of 2DP film obtained using GIWAXS.

3. Gas barrier properties

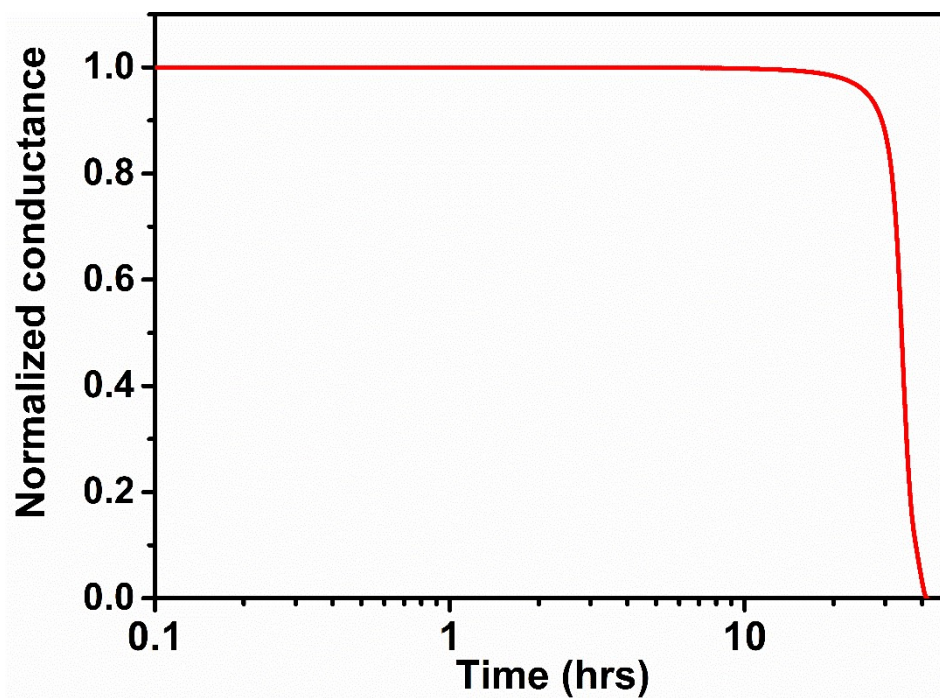


Fig. S6 Graphic plot of normalized conductance vs. time for SB-PDMS/PET/(BL)₄.

Table S1. Barrier Properties of Gas-Diffusion-Barrier-Films (GDBFs)

GDBFs	Lag Time ^a (hours)	Ca Sensor Lifetime (hours)	WVTR (g m⁻² day⁻¹)
PET	0.2	1	1.4
PET/2DP	5	12	4.0*10 ⁻²
PET/(BL) ₁	11	14	9.0*10 ⁻³
PET/(BL) ₂	15	22	6.0*10 ⁻³
PET/(BL) ₄	23	35	9.0*10 ⁻⁴
SB-PDMS/PET/(BL) ₄	25	37	8.5*10 ⁻⁴

^a Lag time: The initial region in the normalized conductance vs. time graph where the conductivity does not change with time.

Table S2. GDBF Performances (WVTR Values) Reported in Literature

GDBFs	WVTR (g m ⁻² day ⁻¹)	Film Thickness	Temperature (°C) & Relative Humidity (RH)	References
PI/PDMS	1	38 μm/130 nm	40°C & 90 % RH	1
PP/(LDH/PSS) ₃₀	1.6 * 10 ⁻¹	179 μm/65 nm	23 °C & 100 % RH	2
PET/PU-Na-hec nanocomposite	<5 * 10 ⁻²	100 μm/1.5 μm	25 °C & 50 % RH	3
PET/(PEI/VMT) ₂₀	6.5 * 10 ⁻¹	179 μm /160 nm	23 °C & 100 % RH	4
PET/(LDH-80/PDMS) ₁₅	5 * 10 ⁻²	N.A/12.95 μm	N.A	5
APTES@LDH(10%)/PLA	2.6 * 10 ⁻²	60 μm	30 °C & 90 % RH	6
PET/(PDDA/h-BN) ₃₀	1.3 * 10 ⁻²	179 μm/130.6 nm	23 °C & 50 % RH	7
PES/(PEI/PMA/PEI/MTM) ₅	3.8 * 10 ⁻²	100 μm /50 nm	20 °C & 60 % RH	8
PET/PVA-LDH nanocomposite	4 * 10 ⁻²	12 μm/1.1 μm	23 °C & 50 % RH	9
PEN/PDDA(GO)/PVA(MMT) ₁₀	3 * 10 ⁻³	125 μm/143 nm	23 °C & 50 % RH	10
BOPP/(PEI/VMT) ₃₀	1.4	17.8 μm/ 226 nm	23 °C & 100 % RH	11
PDMS/PDDA (GO)/PVA (MMT)	2.5 * 10 ⁻²	300 μm/150 nm	23 °C & 50 % RH	12
PET/(PEI/PAA) ₅₀		179 μm/19.5 μm	23 °C & 100 % RH	13
PUA ₈ PU ₂ -PDMS	1.5	125 μm		
PUA ₈ PU ₂ -PDMS/(HL) ₈	8 * 10 ⁻³	125/ 3 μm	23 °C & 50 % RH	14
PET/(HL) ₈ /PUA ₈ PU ₂ -PDMS	7.1* 10 ⁻³	123/3/1 μm		
PET/2DP	4*10⁻²	125 μm/400 nm		
PET/(BL)₁	9*10⁻³	125 μm/1.1 μm	23 °C & 50 % RH	This Work
PET/(BL)₂	6*10⁻³	125 μm/1.1 μm		
PET/(BL)₄	9*10⁻⁴	125 μm/1.1 μm		

Abbreviations used in Table S2: Polydimethylsiloxane (PDMS), Polyimide (PI), Polyethylenimine (PEI), Polyethylene terephthalate (PET), Vermiculite (VMT), Poly(sodium-p-styrenesulfonate) (PSS), Layered double hydroxides (LDH), Polypropylene (PP), Na_{0.5}-fluorohectorite (Na_{0.5}(Mg_{2.5}Li_{0.5})Si₄O₁₀F₂, denoted as Na-hec), Polydiallyldimethylammonium (PDDA), Polyurethane (PU), Graphene oxide (GO), Montmorillonite (MMT), Poly(vinyl alcohol) (PVA), Polyethylene naphthalate (PEN), 3-Aminopropyltriethoxysilane (APTES), Polylactic acid (PLA), LDH nanoplatelets modified by APTES (APTES@LDH), Montmorillonite (MTM), Hexagonal boron nitride (h-BN), Poly(methyl acrylic acid) (PMA), Biaxially oriented polypropylene (BOPP), Poly(ethersulfone) (PES), Amphiphilic surfactant (Tween 80) modified LDH nanoplatelets (denoted as LDH-80).

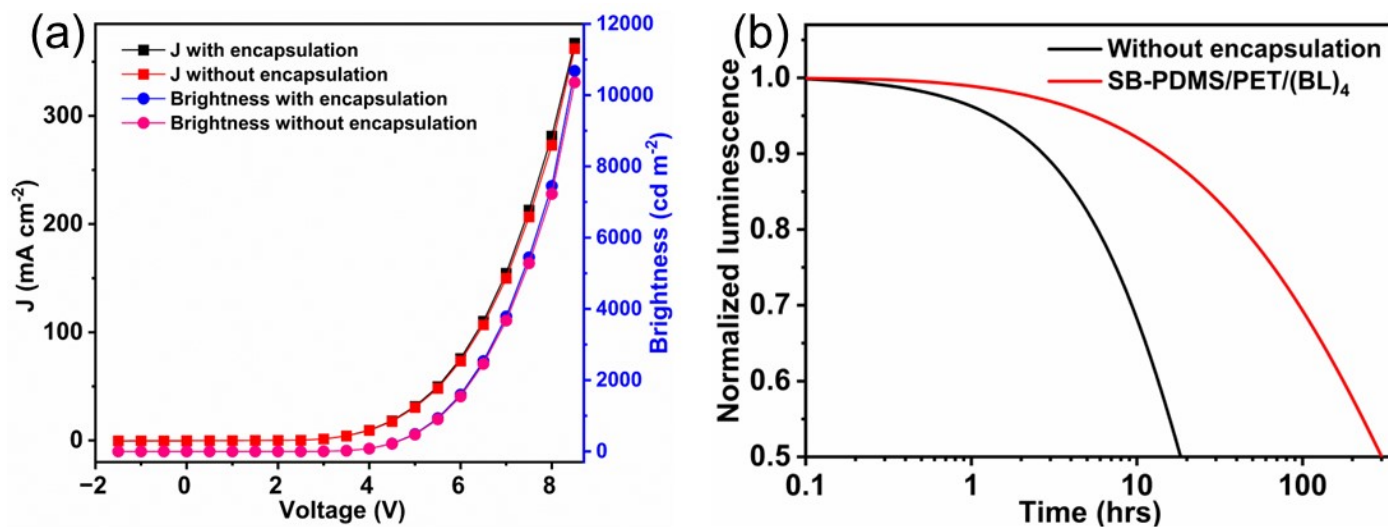


Fig. S7 (a) Initial performances (current density-voltage-luminescence (J-V-L) characteristics) of OLED devices with/without encapsulation and (b) Performance degradation curves of normalized luminescence ($\approx 1000 \text{ Cd m}^{-2}$) versus continuous operation time (up to half-lifetime $L_{50} \approx 300 \text{ h}$) for OLED devices with/without encapsulation of SB-PDMS/PET/(BL)₄.

References

- 1 T. Sasaki, L. Sun, Y. Kurosawa, T. Takahashi and Y. Suzuri, *Adv. Mater. Interfaces*, 2022, **2201517**, 1–8.
- 2 J. Wang, X. Xu, J. Zhang, M. Chen, S. Dong, J. Han and M. Wei, *ACS Appl. Mater. Interfaces*, 2018, **10**, 28130–28138.
- 3 D. A. Kunz, J. Schmid, P. Feicht, J. Erath, A. Fery and J. Breu, *ACS Nano*, 2013, **7**, 4275–4280.
- 4 M. A. Priolo, K. M. Holder, S. M. Greenlee and J. C. Grunlan, *ACS Appl. Mater. Interfaces*, 2012, **4**, 5529–5533.
- 5 J. Wang, T. Pan, J. Zhang, X. Xu, Q. Yin, J. Han and M. Wei, *RSC Adv.*, 2018, **8**, 21651–21657.
- 6 Z. Yang, K. Shi, Z. Jin, Z. Liu, Y. Li, Y. Huang, F. Gao and J. Han, *Ind. Eng. Chem. Res.*, 2022, **61**, 1367–1374.
- 7 T. Y. Kim, E. H. Song, B. H. Kang, S. J. Kim, Y. H. Lee and B. K. Ju, *Nanotechnology*, 2017, **28**, 12LT01.
- 8 J. H. Choi, Y. W. Park, T. H. Park, E. H. Song, H. J. Lee, H. Kim, S. J. Shin, V. Lau Chun Fai and B. K. Ju, *Langmuir*, 2012, **28**, 6826–6831.
- 9 J. Yu, K. Ruengkajorn, D. G. Crivoi, C. Chen, J. C. Buffet and D. O’Hare, *Nat. Commun.*, 2019, **10**, 1–8.
- 10 S. J. Kim, T. Y. Kim, B. H. Kang, G. H. Lee and B. K. Ju, *RSC Adv.*, 2018, **8**, 39083–39089.
- 11 Y. Song, P. Tzeng and J. C. Grunlan, *Macromol. Rapid Commun.*, 2016, **37**, 963–968.
- 12 S. J. Kim, T. Kim, D. Kim and B. K. Ju, *Appl. Sci.*, , DOI:10.3390/app11135794.
- 13 Y. H. Yang, L. Bolling, M. Haile and J. C. Grunlan, *RSC Adv.*, 2012, **2**, 12355–12363.
- 14 S. Mahmood, A. Khan, C. Kant, C. W. Chu, M. Katiyar and H. C. Lin, *Adv. Mater. Interfaces*, DOI:10.1002/admi.202202093.