

Supporting Information

Vanillin-grafted organosilicon backbone polyimide resins with low dielectric, reprocessing and monomer recovery

Guoming Yuan^{a,b,c,d}, Hui Yang^{a,b,c,d}, Zhijun Liu^{a,b,c,d}, Yuemiao Zhang^{a,b,c,d}, Yanhan Tao^{a,b,c,d}, Bo Yang^{a,b,c,d}, Kun Wu^{a,d*}, Jun Shi^{a,d}, Li Yang^e

a Guangzhou Institute of Chemistry, Chinese Academy of Sciences, Guangzhou 510650, PR China.

b CAS Engineering Laboratory for Special Fine Chemicals, Guangzhou 510650, China.

c CASH GCC Shaoguan Research Institute of Advanced Materials, Nanxiong 512400, PR China.

d University of Chinese Academy of Sciences, Beijing 101408, PR China.

e Nanxiong Science and Technology Development Service Center, Shaoguan 512023, PR China.

Section 1. Supplementary Figures.

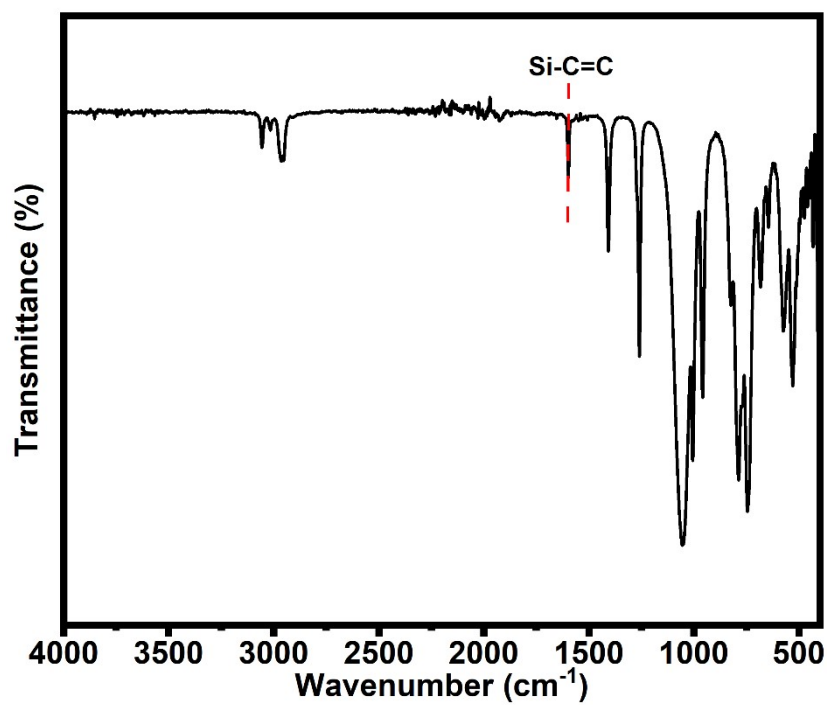


Figure S1. FTIR spectrum of D₄^{Vi}

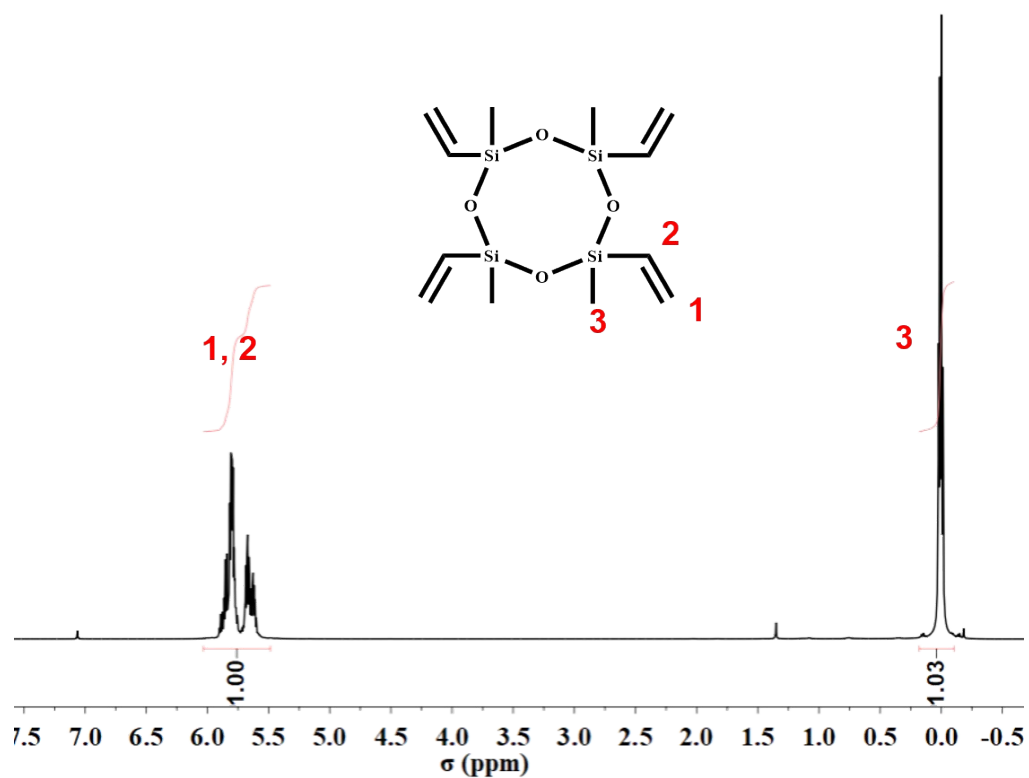


Figure S2. ¹H NMR spectrum of D₄^{Vi}

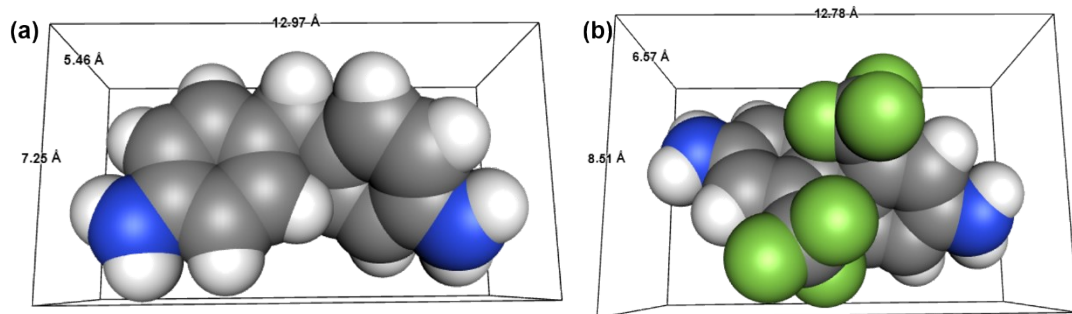


Figure S3. molecular size of (a) DDM and (b) TFDB

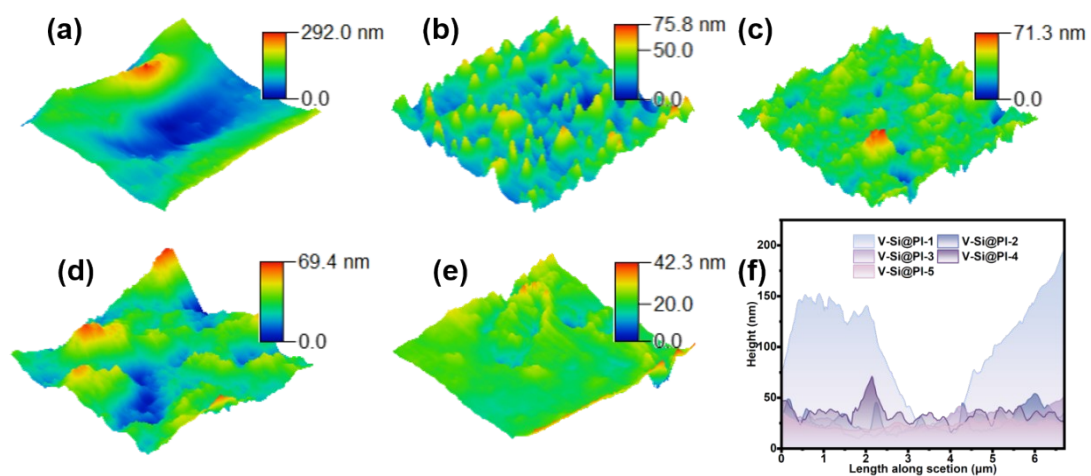


Figure S4. AFM-3D of (a) V-Si@PI-1 , (b) V-Si@PI-2, (c) V-Si@PI-3, (d) V-Si@PI-4, and (e) V-Si@PI-5. (f) Surface height curves of the series V-Si@PI-x

Section 2. Supplementary Tables.

Table S1. ingredients proportion of the series V-Si@PI-x

	V-D ₄ (mmol)	DDM (mmol)	TFDB (mmol)
Si@PI-1	2	4	—
Si@PI-2	2	3	1
Si@PI-3	2	2	2
Si@PI-4	2	1	3
Si@PI-5	2	—	4

Table S2. Swelling ratio (W_{SR}) and gel content W_G of the series V-Si@PI-x

	Si@PI-1	Si@PI-2	Si@PI-3	Si@PI-4	Si@PI-5
W_{SR} (%)	5.1±0.21	6.4±0.35	6.9±0.28	8.2±0.46	10.8±0.43
W_G (%)	86.9±3.8	87.6±4.2	90.3±5.1	91.7±4.8	93.8±4.6

Table S3. Glass transition temperature, storage modulus, crosslink density and Tensile modulus of the series V-Si@PI-x

	V-Si@PI-1	V-Si@PI-2	V-Si@PI-3	V-Si@PI-4	V-Si@PI-5
T_g (°C)	89.78	91.4	91.78	95.36	97.09
Storage modulus (Mpa, 40 °C)	1222	1273	1331	1412	1500
Storage modulus (Mpa, T_{g+30} °C)	18.4	16.44	15.84	14.71	12.75
V_e^a (mol/m ³)	5632.39	5011.76	4824.20	4439.80	3831.23
Tensile modulus (GPa, RT)	1.01	1.19	1.41	1.38	1.58

$$V_e^a = \frac{G_{Tg+30^\circ C}}{RT}$$

Table S4. $T_{d,5\%}$, $T_{d,30\%}$, T_{max} , and Residual mass of the series V-Si@PI-x

	V-Si@PI-1	V-Si@PI-2	V-Si@PI-3	V-Si@PI-4	V-Si@PI-5
$T_{d,5\%}$	238.61	243.79	257.36	264.28	268.79
$T_{d,30\%}$	397.71	397.30	380.30	383.77	382.79
T_{max}	322.41	330.23	339.67	338.32	349.93
Residual mass	52.27	51.16	46.82	44.09	42.30

Table S5. Elemental content of the series V-Si@PI-x

	Atomic content (%)				
	C	N	O	F	Si
V-Si@PI-1	64.37	14.08	14.77	0.44	6.34
V-Si@PI-2	64.61	15.86	13.07	2.89	3.57
V-Si@PI-3	63.67	15.61	13.26	4.19	3.28
V-Si@PI-4	58.87	13.80	15.46	4.59	7.28
V-Si@PI-5	58.45	13.17	14.22	6.60	7.56

Table S6. D_k , D_f , d-spacing, and Ra of the series V-Si@PI-x

	V-Si@PI-1	V-Si@PI-2	V-Si@PI-3	V-Si@PI-4	V-Si@PI-5
D_k (at 100 MHz)	3.08	2.70	2.45	2.38	2.33
D_f (at 100 MHz)	0.0101	0.0073	0.0047	0.0038	0.0030
d-spacing (Å)	4.048	4.076	4.109	4.157	4.219
Ra (nm)	39.73	8.60	8.54	6.26	3.09

Table S6. Dielectric properties, recycling and hydrophobicity of V-Si@PI-3 and published low dielectric materials containing dynamically reversible bonds

Samples	D_k	D_f	Reprocess ability	Monomer recovery	WCA (°)	Ref
OBBBMA	3.06	0.030				29
CIP1.0	4.50	0.045	Yes	No		53
DGEBA/TAI	3.37	0.013	Yes	No		54
C80-PSVMb	2.55	0.0019	Yes	No		55
POF-0.20	2.25	0.018	Yes	No		56
ETOD-SA	2.75	0.016	Yes	Yes		19
V-Si@PI-3	2.45	0.0047	Yes	Yes	119.6	This work

References

- 19 M. Chen, W. Luo, S. Lin, B. Zheng and H. Zhang, *Compos. Part B Eng.*, 2023, **257**, 110666.
- 29 X. Zhang, J. Wu, Z. Qin, L. Qian, L. Liu, W. Zhang and R. Yang, *ACS Appl. Polym. Mater.*, 2022, **4**, 2604–2613.
- 53 G. Ye, C. Wang, Q. Zhang, P. Song, H. Wang, S. Huo and Z. Liu, *Int. J. Biol. Macromol.*, 2024, **278**, 134933.
- 54 J. Liu, X. Liu, X. Cui, J. Qin, M. Shi, D. Wang, L. Liang and C. Yang, *ACS Appl. Polym. Mater.*, 2023, **5**, 10042–10052.
- 55 S. Wang, L. Wang, B. Wang, H. Su, W. Fan and X. Jing, *Polymer*, 2021, **233**, 124214.
- 56 K. Luo, G. Song, Y. Wang, J. Yu, J. Zhu and Z. Hu, *ACS Appl. Polym. Mater.*, 2019, **1**, 944–952.