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Supporting information

Highly Stretchable, Recyclable, Notch-insensitive, and Conductive

Polyacrylonitrile-Derived Organogel

Xiwei Guo, Changgeng Zhang, Lei Shi, Qi Zhang, and He Zhu*

School of Science and Engineering, The Chinese University of Hong Kong, Shenzhen,

518172, Shenzhen, Guangdong, China. Email: zhuhe@cuhk.edu.cn

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Figure S1. APANG before and after solidification. Transparent polymer solution was converted into opaque solid gel after solidification.



Figure S2. FTIR spectra of DMF, PAN and APANG in DMF, and their partial enlarged views.¹⁻³



Figure S3. ¹H-NMR and ¹³C-NMR spectra of PAN (a and b) and APANG (c and d). From the ¹H-NMR spectrum of pristine PAN, an obvious peak at 3.7 ppm belonging to $-OCH_3$ of methyl acrylate was observed, indicating that methyl acrylate existed which is a common co-monomer in commercial PAN. The molar ratio of acrylonitrile and methyl acrylate in the pristine PAN was calculated to be 27.83:1). Peak values (¹³C-NMR) of nitrile group, amidoxime and cyclic imide dioxime group⁴⁻⁹ and peak values (¹H-NMR) of $-OCH_3^{10}$, DMF, H₂O, DMSO-d₆¹¹, amidoxime and cyclic imide dioxime group⁴⁻⁹ were obtained from references. All samples were dissolved in DMSO-d₆.

Table S1. The integral area ratios for $-CH_2$ and -OH, and the conversion percentage of -CN group in the APANG.

Samples	$\int -CH_2$	∫ -OH[a]	\int -OH ^[b]	Percentage
APANG	4.41	0.61	1.00	75.63%

^[a] -OH of amidoxime group and ^[b] -OH of cyclic imide dioxime group.

 Table S2. Results of elementary analysis and the calculated molar ratios of N:C in PAN

 and APANG.^[a]

Weight (mg)	Name	C (%)	N (%)
3.3350	Sulfanilamide ^[b]	48.81	16.25
3.5640	PAN	65.40	24.17
2.5020	PAN	65.47	24.09
4.7110	APANG	46.15	22.33
3.5580	APANG	46.19	22.36
Table S2 continued	1:		
Samples	Ν	С	Mole ratio
PAN ^[c]	27.83	1x4+27.83x3 0.318	
PAN[d]	2/ 13/1/	65 / 35/12	0.316

IAN	24.13/14	05.455/12	0.510
APANG ^[c]	(0.61x2)+(1x1.5)=2.72	(4.41/2)x(3x27.83+4x1)/28.83	0.407
		=6.69	
APANG ^[d]	22.345/14	46.17/12	0.415

^[a] The elementary analysis was conducted twice on the same sample and the calculation was based on the average of the obtained two results. ^[b] Sulfanilamide was used as the reference standard material. ^[c] The calculation was based on the NMR data. ^[d] The calculation was based on the elementary analysis data.



Figure S4. Photos of APANG before and after stretching.



Figure S5. Tensile stress–strain curves of APANGs from different batch. The APANG showed good repeatability.



Figure S6. (a) The strain recovery of APANG after being stretched to the designed strain and (b) the strain recovery of APANG with various time interval.



Figure S7. Load-displacement curves from fracture tests with various crack sizes (initial L is about 14 mm).



Figure S8. (a) Nyquist-plot by impedance spectroscopy and (b) the schematic diagram of impedance spectroscopy test for APANG. The conductivity (δ) was calculated by the following formula: $\delta = L / ((R-R') \times S)$, where R is the resistance of stainless steel (3.3 Ω) and APANG, R' is the resistance of stainless steel, L is the thickness of APANG (2.2 mm) and S is the surface area of APANG (12 mm x 16 mm).



Figure S9. Time-dependent changes of the current for (a) stainless steel, (b) APANG, and (c) ionic liquid (60% PDIM TFSI mass content) incorporated polyethylacrylate under applied voltage of 1V, respectively.



Figure S10. Responses of APANG sensor to cyclic stretching at the strain of 30%, 20%, 10%, and 5%, respectively.



Figure S11. (a) Nyquist-plot by impedance spectroscopy for recycled APANG (0.039 S cm⁻¹) and (b) response of recycled APANG sensor to cyclic stretching at a strain of 50%.

References

- 1. M. Sałdyka, Z. Mielke and K. Haupa, *Spectroc. Acta Pt. A-Molec. Biomolec. Spectr.*, 2018, **190**, 423-432.
- 2. Y. B. Peng, F. Guo, Q. Y. Wen, F. C. Yang and Z. G. Guo, *Sep. Purif. Technol.*, 2017, **184**, 72-78.
- R. Tan, A. Wang, R. Malpass-Evans, R. Williams, E. W. Zhao, T. Liu, C. Ye, X. Zhou, B. P. Darwich,
 Z. Fan, L. Turcani, E. Jackson, L. Chen, S. Y. Chong, T. Li, K. E. Jelfs, A. I. Cooper, N. P. Brandon,
 C. P. Grey, N. B. McKeown and Q. Song, *Nat. Mater.*, 2019, DOI: 10.1038/s41563-019-0536-8.
- 4. P. Akkaş Kavaklı, C. Uzun and O. Güven, *React. Funct. Polym.*, 2004, **61**, 245-254.
- 5. J. Mravljak and Z. Jakopin, *Antioxidants*, 2019, **8**.
- A. Chylewska, M. Biedulska, A. Glebocka, E. D. Raczynska and M. Makowski, J. Mol. Liq., 2019, 276, 453-470.
- 7. N. Mehio, B. Williamson, Y. Oyola, R. T. Mayes, C. Janke, S. Brown and S. Dai, *Ind. Eng. Chem. Res.*, 2016, **55**, 4217-4223.
- C. X. Ma, J. X. Gao, D. Wang, Y. H. Yuan, J. Wen, B. J. Yan, S. L. Zhao, X. M. Zhao, Y. Sun, X. L. Wang and N. Wang, *Adv. Sci.*, 2019, 6.
- 9. F. Karipcin, I. Karatas and H. I. Ucan, *Turk. J. Chem.*, 2003, **27**, 453-460.
- 10. D. W. Shen and H. Yong, *Polymer*, 2004, **45**, 7091-7097.
- 11. H. E. Gottlieb, V. Kotlyar and A. Nudelman, J. Org. Chem., 1997, **62**, 7512-7515.