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Supporting Information

Optimizing Graphene Oxide Membrane for Effective Removing Dyes by Modulating the Reduction Degree and Doped Nitrogen

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Material Characterizations.

The surface and cross-section of NGO-x membranes were evaluated by scanning electron microscopy (HITACHI SU-8010 Field Emission Scanning Electron Microscope). The interlayer spacing of NGO-x membranes was obtained from X-ray diffraction (XRD, Rigaku MiniFlex) using Cu Kα radiation. Raman spectra (Horiba Jobin Yvon LabRAM ARAMIS) with 532 nm laser was used to analyze the structure of GO and NGO-x membranes. Fourier transform infrared spectroscopy (FT-IR, VERTEX 70, Bruker, Germany). X-ray photoelectron spectroscopy (ESCALAB 250Xi, ThermoFisher) were employed to characterize the chemical composition and structure of GO and NGO-x membranes. The contact angle of the membranes were measured to evaluate their hydrophilicity by a contact angle tester (JC20000D1, Shanghai Zhongchen Co., Ltd., China). Test the concentration of the dye solution were determined by using UV–vis spectrophotometer (UV–vis, UV756CRT, You Ke Co., Ltd., China).

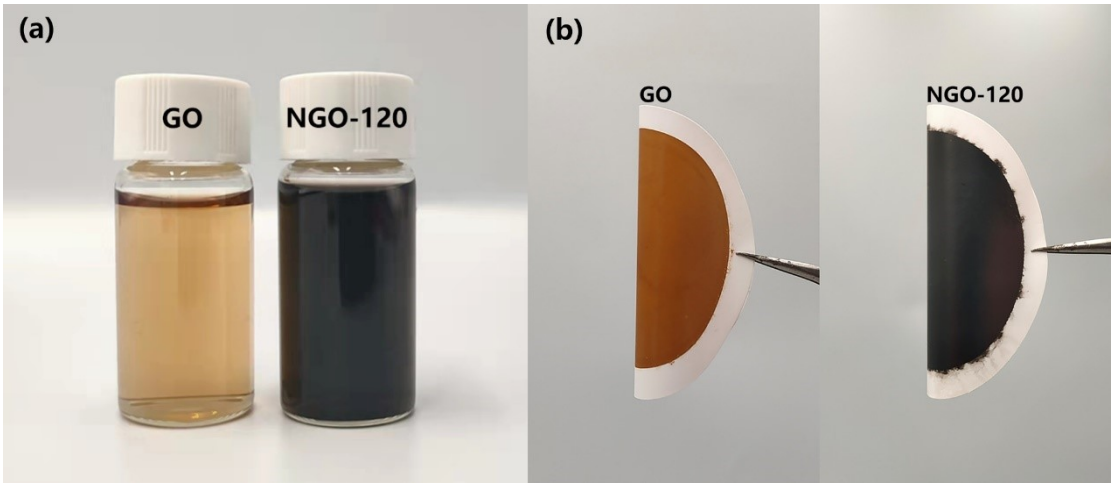

Fig. S1. Digital Photograph of GO and NGO (a) dispersions and (b) membranes.

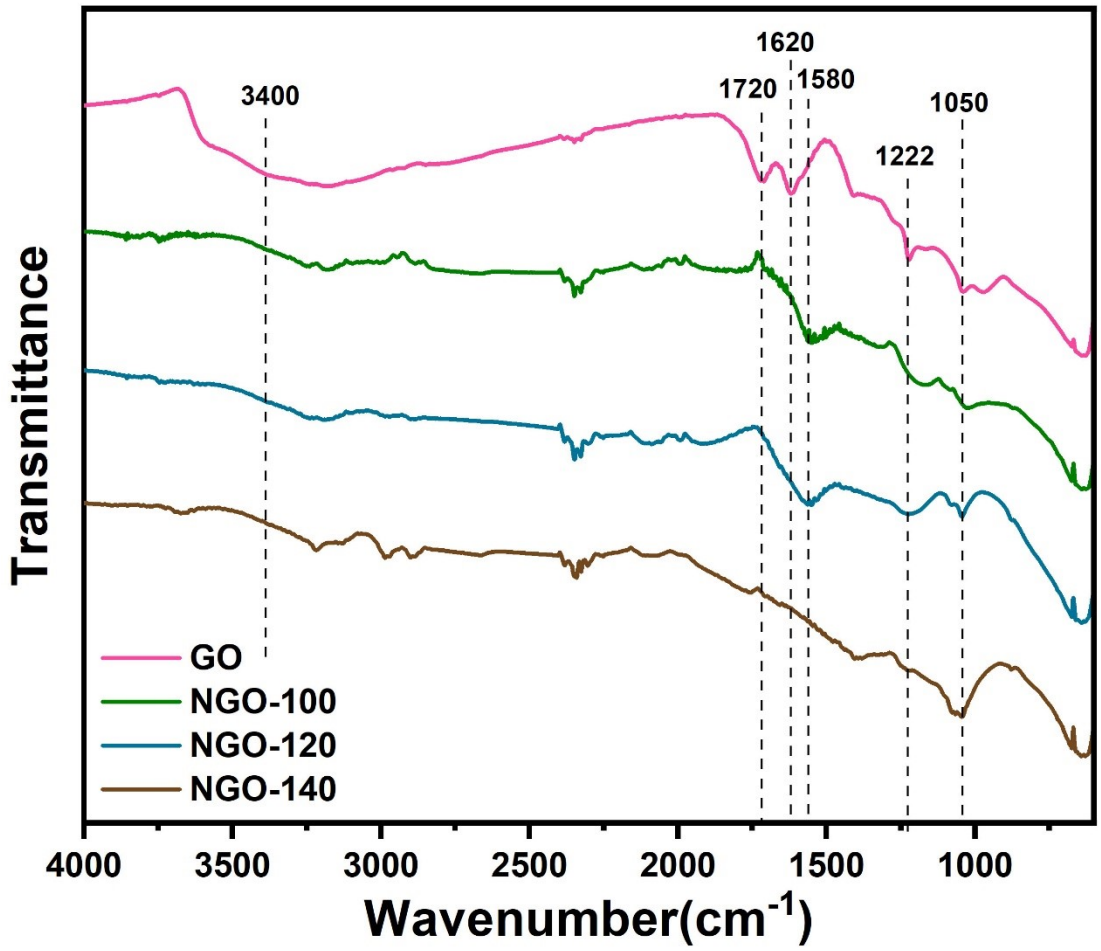

Fig. S2. FT-IR spectra of GO and NOG-x membranes.

The characteristic bands of the infrared spectrum of GO are located at 3204 cm^{-1} (-OH stretched), 1720 cm^{-1} (C=O stretched from the carbonyl group), 1620 cm^{-1} (C=C stretched), 1220 cm^{-1} (C-OH stretched) -O vibration) and 1050 cm^{-1} (C-O vibration of epoxy group).[1] With the increase of the hydrothermal temperature, the bands of -OH, C=O and C-OH gradually weakened or even disappeared, indicating that some oxygen-containing functional groups in GO decreased. At the same time, the appearance of a new peak at ~1050 cm^{-1} in the NGO-x sample indicates the formation of a C-N bond.[2] The peak at ~1580 cm^{-1} is attributed to the N-H bending vibration in the NGO-x samples.[3] The formation of the N-C bond and the remaining C-O group are confirmed by the appearance of a new peak at ~1100 cm^{-1}.[4] All these results confirm the partial reduction and nitrogen doping on graphene.

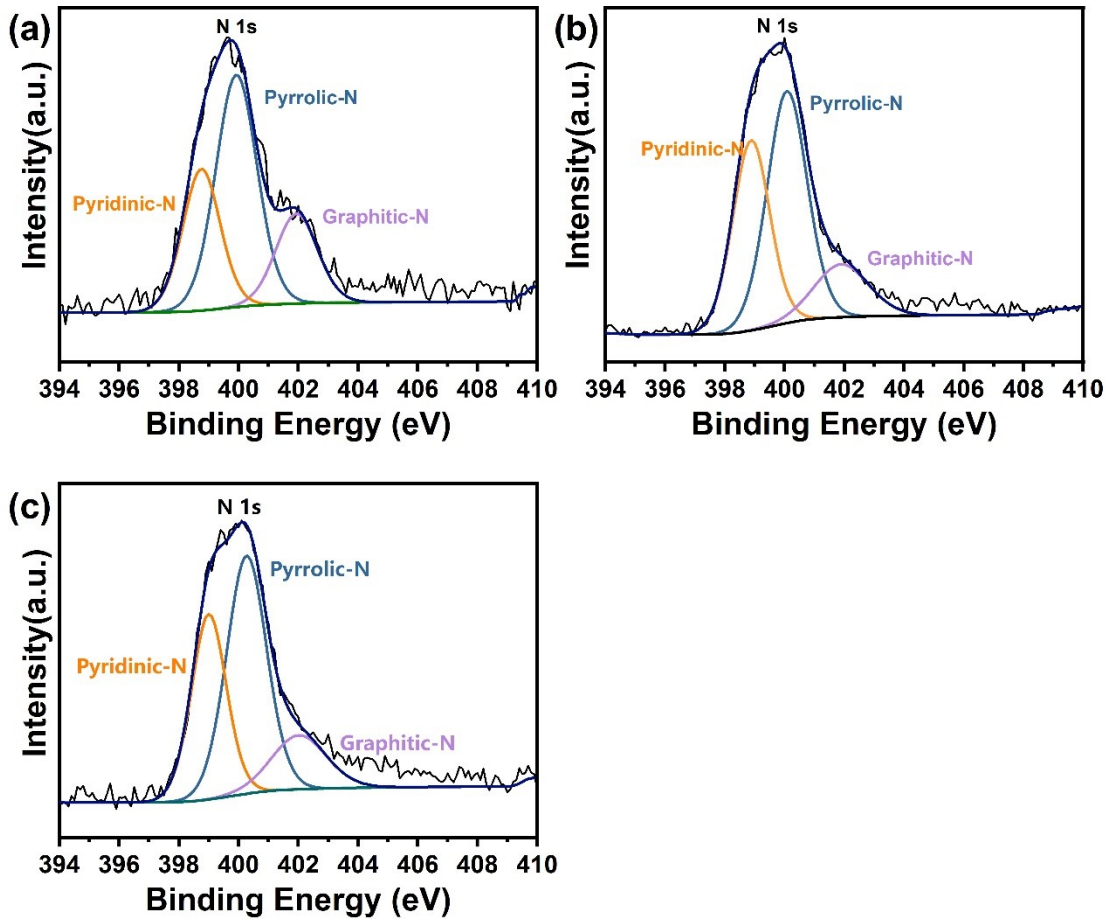

Fig. S3. XPS spectra. N1s of (a) NGO-100, (b) NGO-120 and (c) NGO-140.

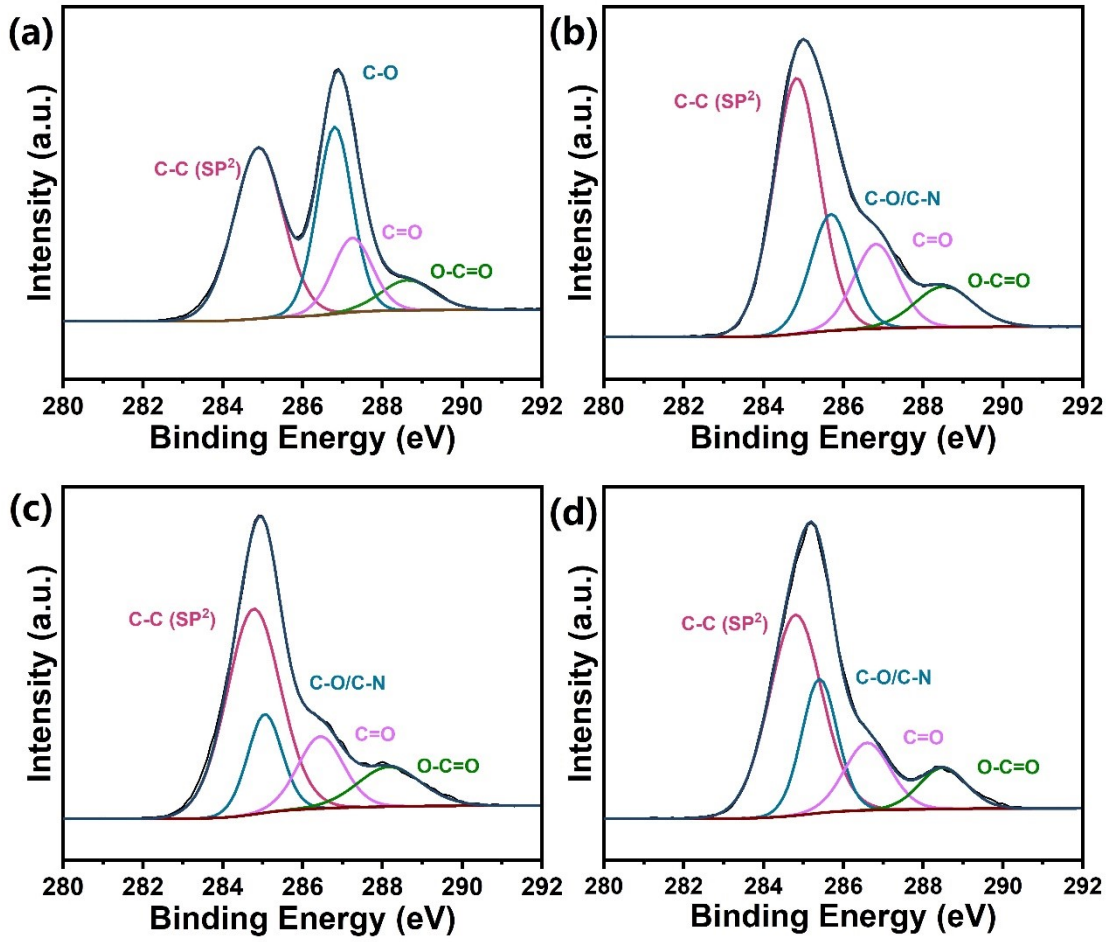

Fig. S4. XPS spectra. C1s of (a) GO powder, (b) NGO-100, (c)NGO-120 and (d) NGO-140.

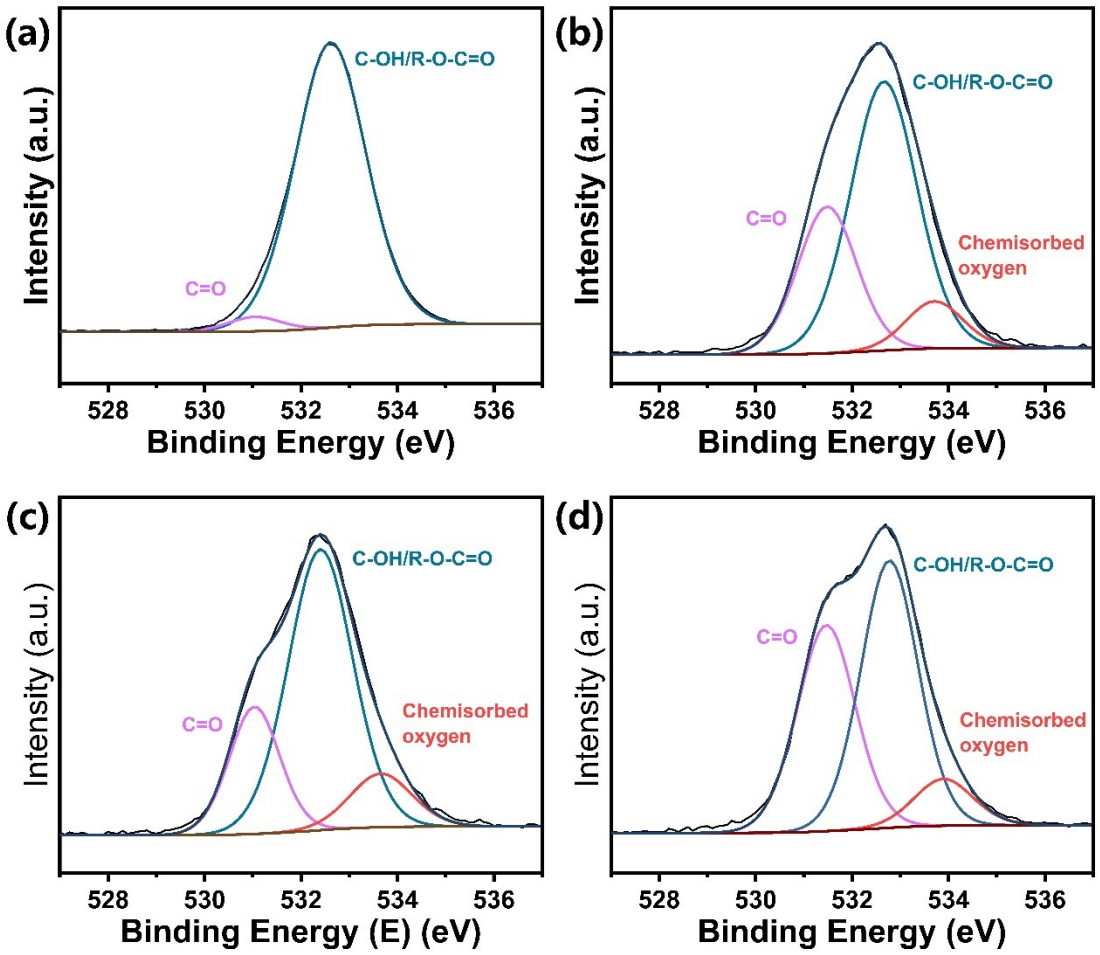

Fig. S5. XPS spectra. O1s of (a) GO powder, (b) NGO-100, f (c) NGO-120 and (d) NGO-140.

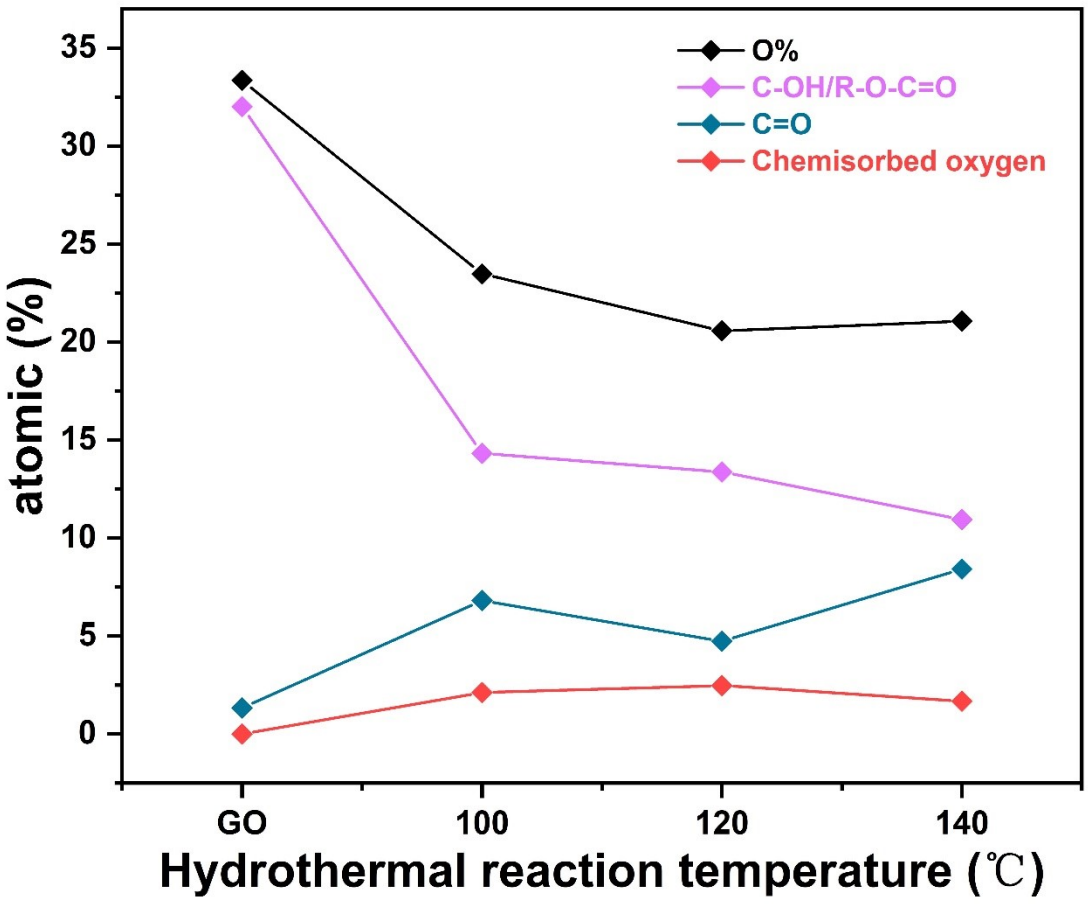

Fig. S6. The relationship between functional oxygen-containing groups and hydrothermal temperature.

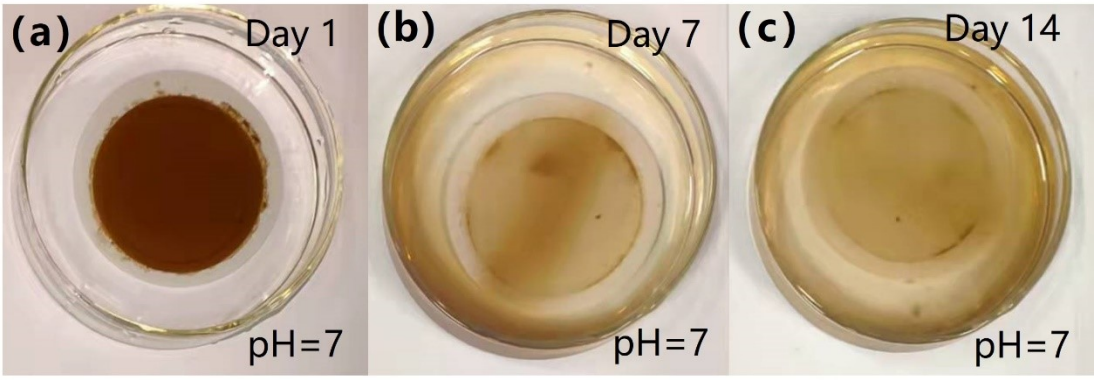

Fig. S7. Physical photos of 14-day stability of GO in water

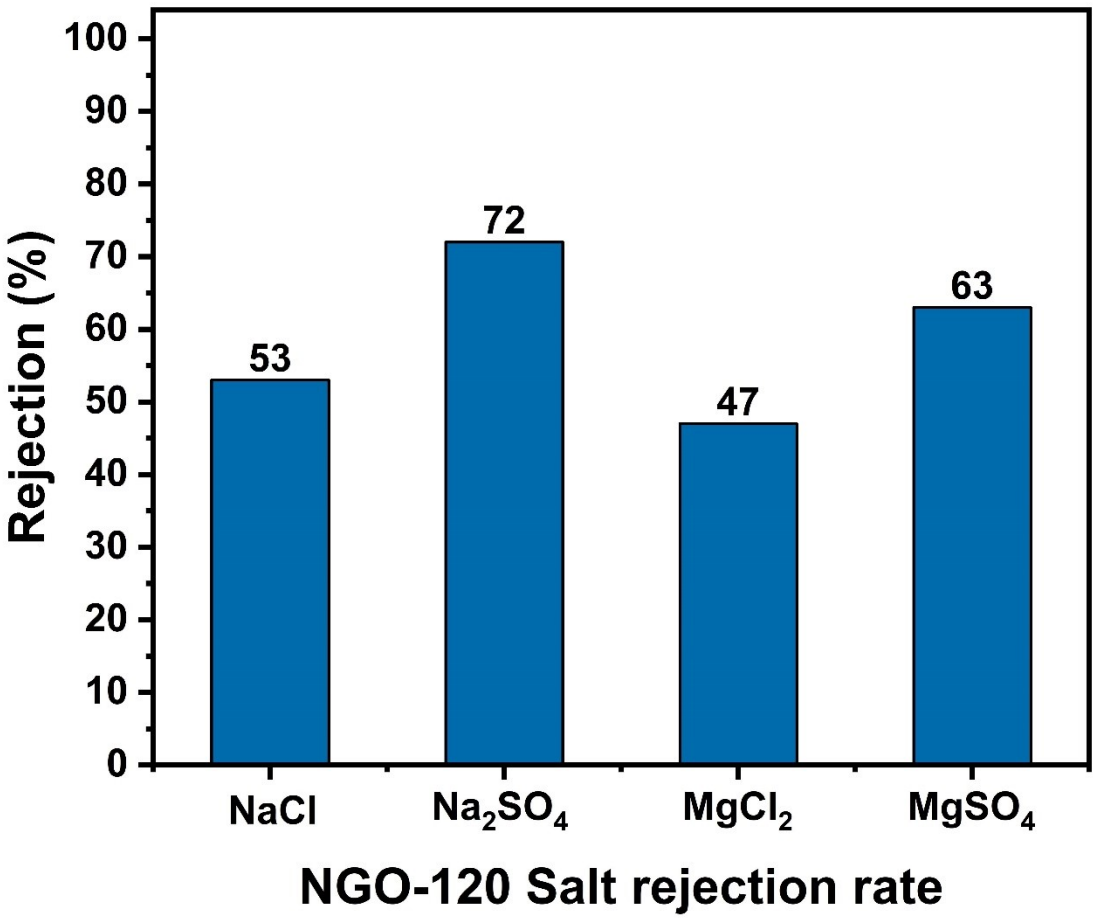

Fig. S8. NGO-120 Salt rejection rate (Feed salt concentration: 2000 ppm. Feed pressure: 1 bar.).

Table S1. Comparison of the nanofiltration performance for graphene-based membranes.

Membrane	Water permeance ($L \cdot m^{-2} \cdot h^{-1}$ bar^{-1})	MWCO	Rejection (%)	Pressure (bar)	Ref
NGO-100	4.89	320 (MnB) 800 (MB)	MnB (>97%), MB (99.8%)	1	This work
NGO-120	9.76	320 (MnB) 800 (MB)	MnB (99.6%), MB (99.8%)	1	This work
NGO-140	5.86	320 (MnB) 800 (MB)	MnB (>95%), MB (>97%)	1	This work
$(PDDA/GO)_{4.0}$/PAN	6.42	800 (MB)	MB (99%)	5	[5]
MWCNTs/rGO/PVDF	11.3	327 (MO)	MO (>96%)	5	[6]
rGO	16	800 (MB)	MB (99%)	1	[7]
GO@ PAN	8.2	697 (CR)	CR (99%)	5	[8]
GO/NG	6.13	800 (MB)	MB (>98%)	1	[9]
GO/MB	3.83	327 (MO)	MO (>96%)	1	[10]
PDA / RGO / MOFs	5	697 (CR)	CR (>89%)	1	[11]
uGNMs	21.81	800 (MB)	MB (99.2%)	1	[12]
GO/Ca/CR	17.1	800 (MB)	MB (99.5%)	5	[13]

References

1 H. H. Huang, R. K. Joshi, K. K. H. De Silva, R. Badam and M. Yoshimura, *Journal of Membrane Science*, 2019, **572**, 12-19.

2 R. W. Shu, Z. L. Wan, J. B. Zhang, Y. Wu, Y. Liu, J. J. Shi and M. D. Zheng, *Acs Applied Materials & Interfaces*, 2020, **12**, 4689-4698.

3 M. Song, J. Zhao, Y. Meng, L. Riekehr, P. X. Hou, H. Grennberg and Z. B. Zhang, *ChemNanoMat*, 2019, **5**, 1144-1151.

4 Z. Y. Lin, G. Waller, Y. Liu, M. L. Liu and C. P. Wong, *Advanced Energy Materials*, 2012, **2**, 884-888.

5 L. Wang, N. X. Wang, J. Li, J. W. Li, W. W. Bian and S. L. Ji, *Separation and Purification Technology*, 2016, **160**, 123-131.

6 Y. Han, Y. Q. Jiang and C. Gao, *Acs Applied Materials & Interfaces*, 2015, **7**, 8147-8155.

7 X. T. Fan, C. B. Cai, J. Gao, X. L. Han and J. D. Li, *Separation and Purification Technology*, 2020, **241**, 8.

8 J. Q. Wang, P. Zhang, B. Liang, Y. X. Liu, T. Xu, L. F. Wang, B. Cao and K. Pan, *Acs Applied Materials & Interfaces*, 2016, **8**, 6211-6218.

9 R. T. Hou, Y. He, H. Yu, J. Ma, Y. X. Gao, Y. Bai and J. Y. Chen, *Chemical Physics Letters*, 2021, **775**, 8.

10 J. K. Hou, Y. B. Chen, W. X. Shi, C. L. Bao and X. Y. Hu, *Applied Surface Science*, 2020, **505**, 12.

11 Y. C. Liu, M. Zhu, M. Y. Chen, L. L. Ma, B. Yang, L. L. Li and W. W. Tu, *Chemical Engineering Journal*, 2019, **359**, 47-57.

12 Y. Han, Z. Xu and C. Gao, *Advanced Functional Materials*, 2013, **23**, 3693-3700.

13 S. Zhao, H. T. Zhu, H. Wang, P. Rassu, Z. Wang, P. Song and D. W. Rao, *Journal of Hazardous Materials*, 2019, **366**, 659-668.