

Supporting Information

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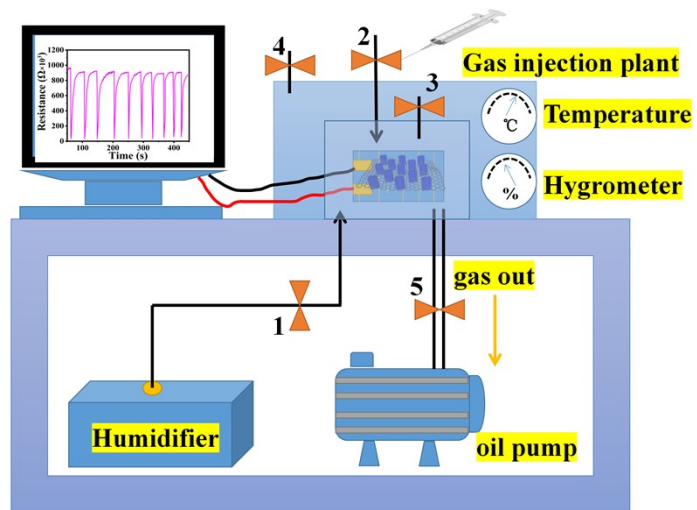


Fig.S1 Self-assembled gas sensor diagram

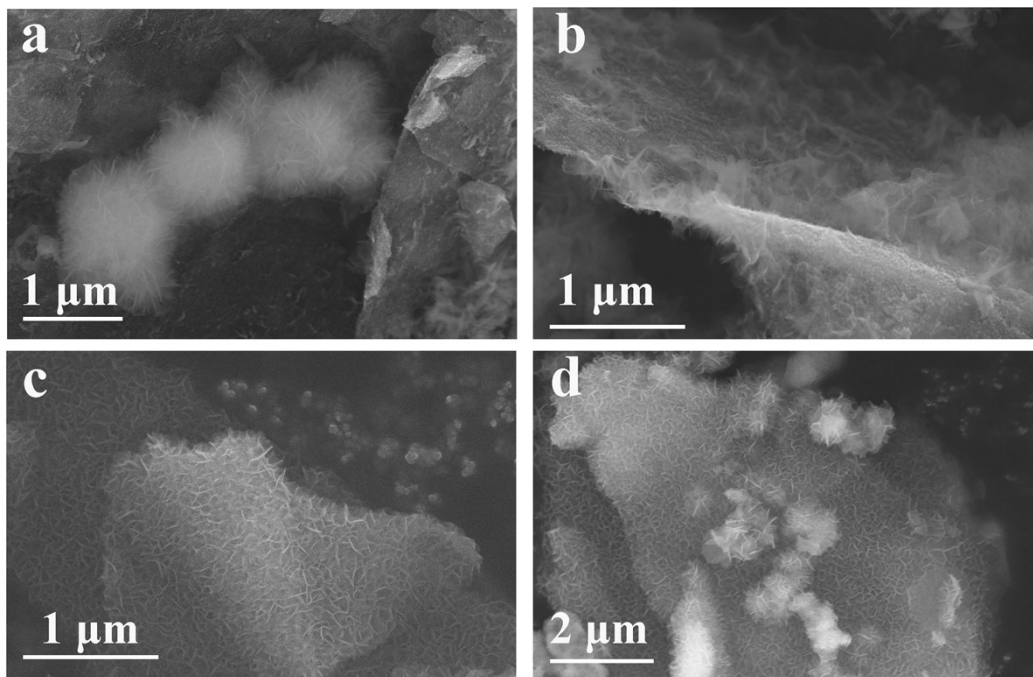


Fig.S2 SEM images of NF/rGOz;NF/rGO1; NF/rGO2; NF/rGO3

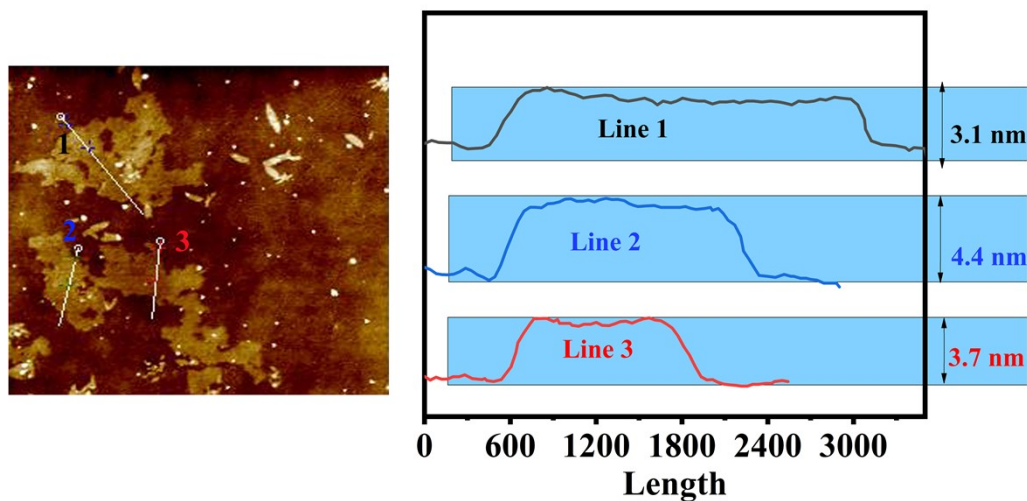


Fig S3 AFM images height profiles of GO

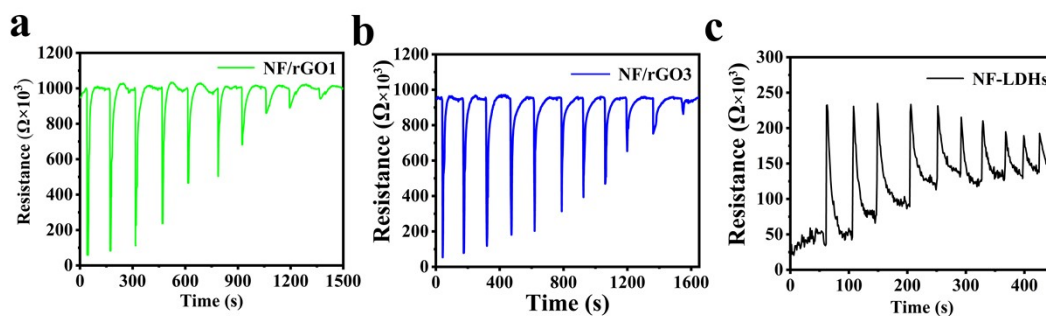


Fig. S4 The dynamical response/recovery transient curves of
(a)NF/rGO1,(b)NF/rGO3 and (c) NF-LDHs sensor to 100-0.1 ppm NO₂.

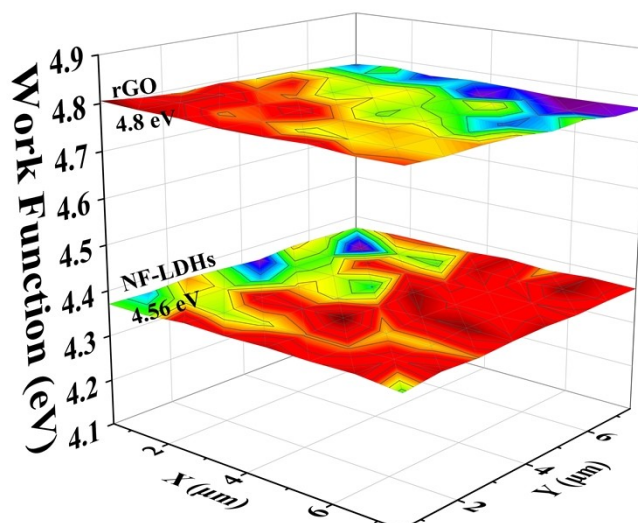


Fig. S5 Work function of NF-LDHs and rGO

Table S1 XRD parameters of NF/rGO series and NF-LDHs

| Samples | D ₀₀₃ /nm | D ₁₁₀ /nm | I ₁₁₀ /I ₀₀₃ |
|---------|----------------------|----------------------|------------------------------------|
| NF-LDHs | 24.55 | 36.875 | 0.18 |
| NF/rGO1 | 6.145 | 23.779 | 0.24 |
| NF/rGO2 | 7.769 | 13.433 | 0.35 |
| NF/rGO3 | 7.946 | 8.9123 | 0.21 |

*Based on Scherrer equation $D=0.89 \lambda / \beta \cos \theta$, D is Lattice dimension (nm); β is the full width at half-maximum(in radian); θ is diffraction angle; λ is the wavelength of X-rays 0.1542 nm

*The diffraction intensity ratio of (110) line to (003) line

Table S2 O1s peaks position and peak area ratio of NF-LDHs and NF/rGO

| Samples | NF-LDHs | | | NF/rGO2 | | |
|----------------------|----------------|----------------|----------------|----------------|----------------|----------------|
| | O _a | O _b | O _c | O _a | O _b | O _c |
| Energy position (eV) | 530.7 | 531.7 | 533.6 | 531.2 | 532.0 | 533.3 |
| Peak area ratio (%) | 38.06 | 45.03 | 16.9 | 18.96 | 49.42 | 31.6 |

Table S3 The response, response time and recovery time of the three samples expose to NO₂ concentrations at room temperature (RH: 26%)

| Sample | NF/rGO1 | | | NF/rGO2 | | | NF/rGO3 | | | NF-LDHs | | |
|-----------------------|---------|------|------|---------|------|------|---------|------|------|---------|-----|------|
| NO ₂ (ppm) | R | Ts | Tr | R | Ts | Tr | R | Ts | Tr | R | Ts | Tr |
| 100 | 16.5 | 2.4 | 56.9 | 22.3 | 2.3 | 46 | 17.6 | 4.3 | 63.3 | 6.6 | 2.4 | 32 |
| 50 | 11.96 | 3 | 46 | 18.6 | 3.2 | 34 | 12.03 | 11.1 | 45.2 | 5.1 | 2.5 | 29.3 |
| 30 | 8.63 | 3.5 | 32 | 15.6 | 4.8 | 29.6 | 7.96 | 14.4 | 35.4 | 3.25 | 2.5 | 26.2 |
| 10 | 4.2 | 5.2 | 26 | 12.7 | 6.32 | 24.5 | 5.21 | 17.6 | 29.2 | 2.57 | 2.6 | 23.6 |
| 5 | 2.13 | 6.2 | 21.3 | 9.24 | 8.01 | 21.6 | 4.67 | 18.4 | 25.4 | 2.05 | 2.8 | 20.2 |
| 3 | 1.98 | 7.3 | 18.1 | 7.57 | 8.6 | 16.6 | 3.02 | 23.6 | 21.9 | 1.51 | 3.1 | 18.8 |
| 1 | 1.46 | 9.4 | 15.6 | 4.26 | 9.3 | 14.3 | 2.41 | 24.3 | 16.5 | 1.50 | 3.2 | 17.5 |
| 0.5 | 1.16 | 10.2 | 14.3 | 3.17 | 11.3 | 14.3 | 1.96 | 26.1 | 14.4 | 1.49 | 5.4 | 15.7 |
| 0.3 | 1.12 | 11.3 | 12.6 | 2.45 | 12.3 | 12.8 | 1.45 | 27.5 | 12.3 | 1.42 | 6.8 | 12.3 |
| 0.1 | 1.06 | 12.1 | 11.3 | 1.92 | 13.6 | 11.6 | 1.26 | 29.1 | 11.2 | 1.41 | 7.1 | 10.2 |
| 0.05 | | | | 1.32 | 13.8 | 10.3 | 1.09 | 30 | 10.7 | | | |
| 0.01 | | | | 1.12 | 13.9 | 10.2 | | | | | | |

***R: Response; Ts: Response time; Tr: Recovery time**

Table S4 Parameters obtained by fitting experimental curve to equivalent circuit

| Raw materials | N_d | $R_1(\Omega)$ | $C_1(F)$ | $R_2(\Omega)$ | $C_2(F)$ |
|---------------|-----------------------|--------------------|------------------------|--------------------|------------------------|
| NF-LDHs | 2.89×10^{15} | 1.63×10^5 | 1.52×10^{-12} | 4.97×10^6 | 8.96×10^{-11} |
| NF/rGO1 | 2.19×10^{16} | 7.32×10^5 | 1.14×10^{-10} | 1.45×10^5 | 9.81×10^{-12} |
| NF/rGO2 | 2.78×10^{16} | 7.06×10^6 | 1.20×10^{-10} | 1.44×10^5 | 1.10×10^{-11} |
| NF/rGO3 | 2.01×10^{16} | 5.78×10^5 | 1.33×10^{-10} | 1.94×10^5 | 1.00×10^{-11} |

* $N_d = - (2/e_0 \varepsilon \varepsilon_0) [d(1/C^2)/dV]^{-1}/C^2$ is the slope of the curve, q is the charge constant ($q=1.69 \times 10^{11}$), ε_0 is the vacuum dielectric constant ($8.859 \times 10^{-14} \text{ F cm}^{-1}$), and ε is the relative dielectric constant of NiFe-LDHs ($\varepsilon = 20$)

Table S5 Current performance of typical gas sensors based on bimetallic composites in literature reports

| Samples | Application | Operation temperature | Response/concentration | References |
|--------------------------|--------------------|-----------------------|------------------------|------------|
| Ag/ NiAl-LDHs | Ethanol gas sensor | RT | 10.89/250 ppm | [1] |
| ZnTi-LDHs/ Ti 3C 2 Tx | NH ₃ | RT | 1.26/100 ppb | [2] |
| PANI/ZnTi-LDHs | NH ₃ | RT | 9.91/5 ppm | [3] |
| Zn-Cr-WO-LDHs | Cl ₂ | RT | 60%/100 ppm | [4] |
| Co-AlLDHs | NO ₂ | RT | 17.09/100 ppm | [5] |
| CoAl-LDH | NO ₂ | RT | 26.61/100 ppm | [6] |
| Ni-Al-LDH | O ₃ | RT | 1.84/700 ppb | [7] |
| ZnV/ZnCr-LDHs | SO ₂ | RT | 71.71%/100 ppm | [8] |
| PS@Co-LDH | Ethanol vapour | 200°C | 2.48/4.3 ppm | [9] |
| Zn-TiLDHs/rGO | NO ₂ | RT | 97%/100 ppm | [10] |
| NiFe LDH/rGO | NO ₂ | RT | 22.3/100 ppm | This work |

References

- [1]S. A. Zakaria, S. H. Ahmadi and M. H. Amini, *Materials Chemistry and Physics*,2024, **318**,129216.
- [2]Y. Qin, H. Gui, Y. Bai and S. Liu, *Sensors and Actuators B: Chemical* ,2022, **352**, 131077.
- [3]Y. X. Qin L. P. Wang and X. F. Wang, *Organic Electronics*,2019, **66**, 102-109.
- [4]R. B. Shinde, A. S. Patil, S. Sadavar, V, Y. M. Chitare, V. V. Magdum, N. S. Padalkar, U. M. Patil, S. T. Kochuveedu, V. G. Parale and H. H. Park, *Sensors and Actuators B: Chemical*,2022, **352**,131046.
- [5]D. Wang, Z. Liu, Y. Hong, C. Lin, Q. Pan, L. Li and K. Shi, *RSC Advances*,2020, **10**,34466-34473.
- [6]Z. Liu, L. Teng, L. Ma, Y. Liu, X. Zhang, J. Xue, M. Ikram, M. Ullah, L. Li and K. Shi, *RSC Advances*,2019, **9**, 21911.
- [7]G. Kang, Z. Zhu, B. H. Tang, C. H. Wu and R. J. Wu, *Sensors & Actuators: B. Chemical*,2017, **241**, 1203-1209.
- [8]R. B. Shinde, N. S. Padalkar and S. V. Sadavar, *Materials Today Chemistry*,2022, **24**,100801.
- [9]Y. Li, F. Zhou, L. Gao and G. Duan, *Sensors & Actuators: B. Chemical*,2018, **261**, 553-565.
- [10]Y. Qin, R. Zhao and C. Bai, *New Journal of Chemistry*,2020, **44**, 16985-16994.