

Supporting Information

Flexible laser-induced graphene-based electrodes modified with cobalt-manganese hexacyanoferrate as cathode materials for asymmetric supercapacitors

Evgeniia Khairullina^{a,b}, Alexandra Levshakova^a, Maxim Fatkullin^c, Maxim Tenevich^d, Alexandr Shmalko^e, Maxim Panov^{a,f}, Alina Manshina^a, Artem Lobinsky^d, Raul D. Rodriguez^d, Maria Kaneva^{*d}

^a St. Petersburg State University, Institute of Chemistry, St. Petersburg 199034, Russia

^b ITMO University, Department School of Physics and Engineering, St. Petersburg 191002, Russia

^c Tomsk Polytechnic University, Research School of Chemistry & Applied Biomedical Sciences, Tomsk 634050, Russia

^d Ioffe Institute, Hydrogen Energy Laboratory, St. Petersburg 194021, Russia

^e Saint Petersburg Academic University, Nanotechnology Research and Education Centre RAS, St. Petersburg 194021, Russia

^f St. Petersburg State Chemical Pharmaceutical University, Center for physical sciences and information technologies, St. Petersburg 197022, Russia

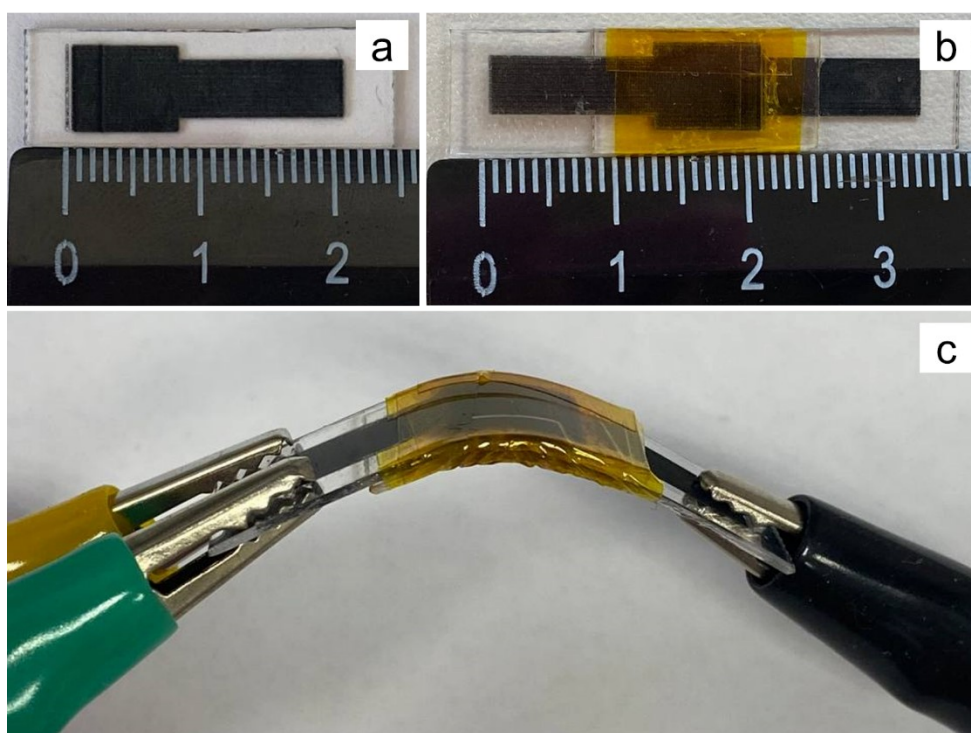


Fig. S1. (a) Photo of the LIMPC electrode, (b) FACS and (c) bending resistance of FASC.

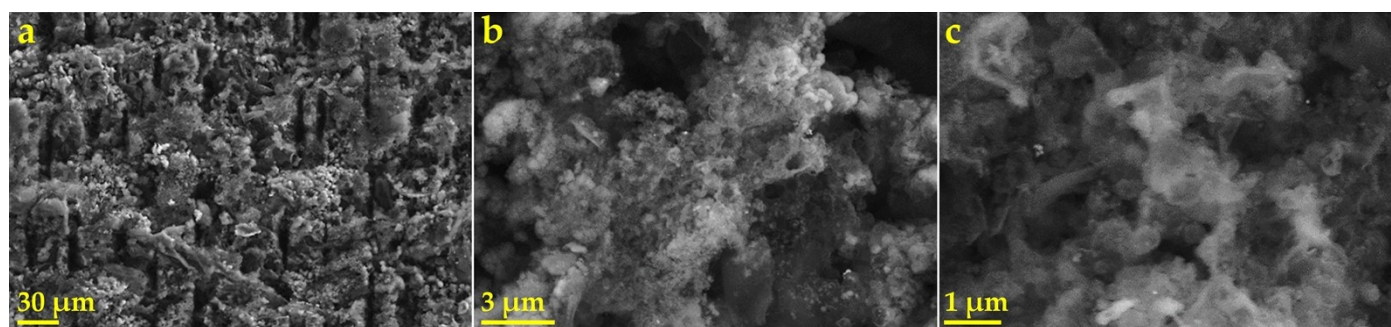


Fig. S2. SEM images of LIMPC electrode at various magnifications.

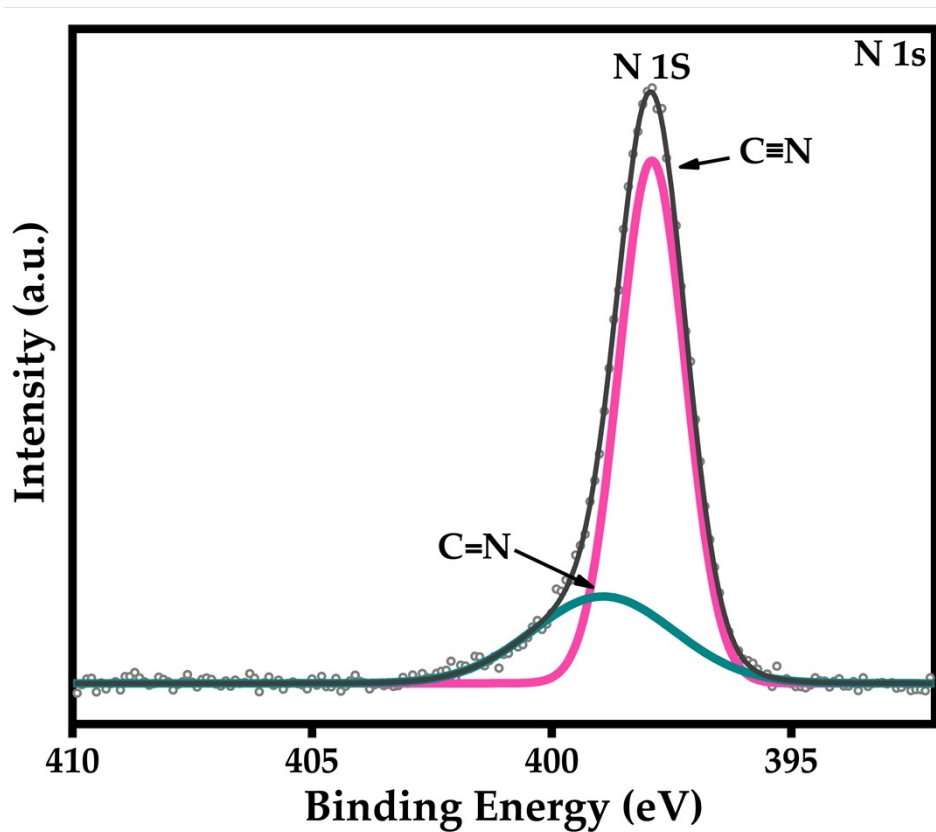
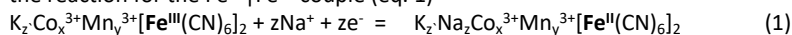
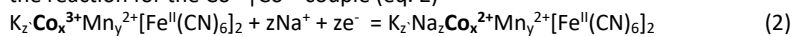


Fig. S3. X-ray photoelectron spectra of the N 1s.

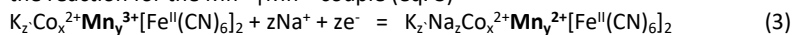
the reaction for the $\text{Fe}^{3+} | \text{Fe}^{2+}$ couple (eq. 1)



the reaction for the $\text{Co}^{3+} | \text{Co}^{2+}$ couple (eq. 2)



the reaction for the $\text{Mn}^{3+} | \text{Mn}^{2+}$ couple (eq. 3)



$$z' + x + y = 3$$

For the purpose of clarity and simplification, the hydration water and counter anions have been omitted.

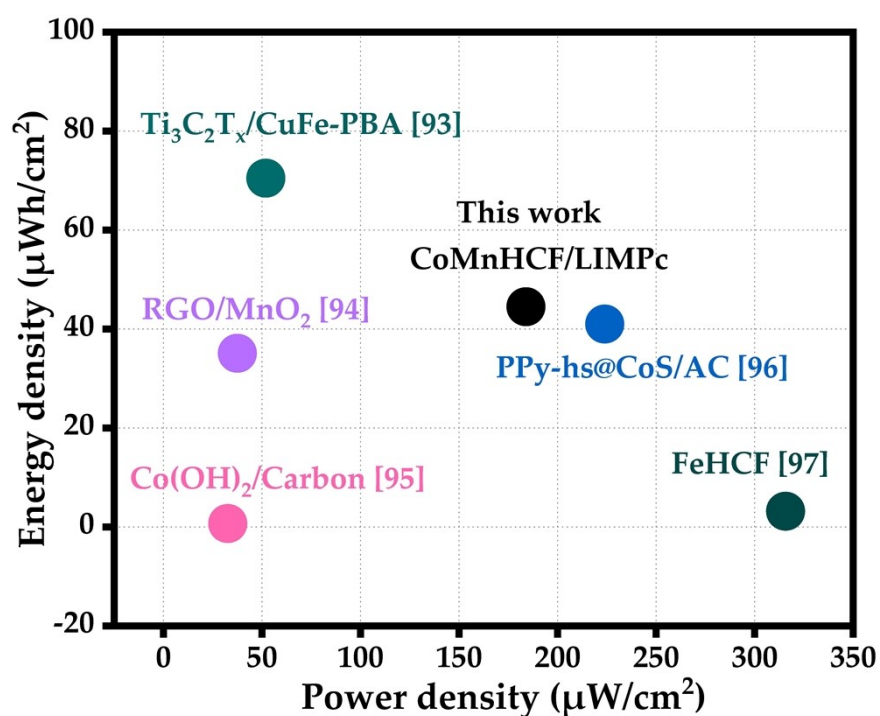
Scheme S1. The possible chemical equations for redox processes in CoMnHCF.

Table S1. Impedance parameters obtained by data fitting with the electrical equivalent circuit and calculation.

| | LIMPc | CoHCF | MnHCF | CoMnHCF | CoMnHCF after 10000 cycles |
|----------|-------|-------|-------|---------|----------------------------|
| R_s | 8.1 | 7.5 | 8.6 | 7.4 | 8.75 |
| R_{CT} | 6.7 | 2.2 | 5.4 | 1.7 | 22.25 |

Table S2. Characteristics of cathode materials and ASCs

| Cathode material | Areal capacitance (mF/cm ²) | | Electrolyte | Retention | Method synthesis | Ref. |
|--|---|--|---------------------------------------|--------------------------|--|--------------|
| | Cathode | ASC | | | | |
| WO ₃ /WS ₂ | 55.3 at 5 mV/s | 32.5 at 5 mV/s 31 at 1 mA/cm ² | 0.1 M Na ₂ SO ₄ | 100% after 10 000 cycles | electrochemical deposition and chemical vapor deposition | [89] |
| rGO/MnO ₂ | 274 at 2 mV/s | 152 at 2 mV/s | 0.5 M Na ₂ SO ₄ | - | hydrothermal method | [9] |
| CuO/MnO ₂ | 261.4 at 1 mA/cm ² | 152.7 at 1 mA/cm ² | 1 M Na ₂ SO ₄ | 90% after 1000 cycles | electrodeposition method and SILAR | [90] |
| CC/MoO ₂ /Au/MnO ₂ | 112 at 5 mV/s | - | 1 M Na ₂ SO ₄ | 95% after 3000 cycles | electrodeposition method | [91] |
| Ag/V ₃ O ₇ | 274.5 F/g at 0.3 A/g | 322.5 at 0.5 mA/cm ² | 1 M Na ₂ SO ₄ | 90.8% after 5000 cycles | screen printing, template-solvothermal route | [92] |
| CoMnHCF_90/LIMP | 224.5 at 0.5 mA/cm ² | 142.3 at 0.25 mA/cm ² | 1 M Na ₂ SO ₄ | 93% after 1000 cycles | laser-assisted integration and SILAR | This article |

**Fig. S4.** Ragone plot of the CoMnHCF/LIMPc FASC compared with the reported supercapacitors

- [89] K. S. Kumar, N. Choudhary, D. Pandey, L. Hurtado, H. S. Chung, L. Tetard, Y. Jung, J. Thomas, *Nanotechnology* **2020**, *31*, DOI 10.1088/1361-6528/aba305.
- [90] B. Liu, L. Tian, X. Zheng, Z. Xing, *J. Alloys Compd.* **2022**, *911*, DOI 10.1016/j.jallcom.2022.165003.
- [91] X. Zhang, Y. Xu, Y. Ma, M. Yang, Y. Qi, *Eur. J. Inorg. Chem.* **2015**, *2015*, DOI 10.1002/ejic.201500501.
- [92] B. Lin, Y. Zheng, J. Wang, Q. Tu, W. Tang, L. Chen, *Nanomaterials* **2023**, *13*, DOI 10.3390/nano13162282.
- [93] Y. Lei, W. Zhao, Y. Zhu, U. Buttner, X. Dong, H. N. Alshareef, *ACS Nano* **2022**, *16*, DOI 10.1021/acsnano.1c06552.
- [94] A. Sumboja, C. Y. Foo, X. Wang, P. S. Lee, *Adv. Mater.* **2013**, *25*, DOI 10.1002/adma.201205064.
- [95] P. Navaneeth, V. K. V, A. K. P, A. R. S, B. G. Nair, S. B. T G, P. V. Suneesh, *Electrochim. Acta* **2022**, *430*, DOI 10.1016/j.electacta.2022.141096.
- [96] Y. Zhao, J. Zheng, J. Yang, W. Liu, F. Qiao, J. Lian, G. Li, T. Wang, J. Zhang, L. Wu, *Nano Res.* **2023**, *16*, DOI 10.1007/s12274-022-5201-x.
- [97] S. C. Vu Van Thuy, Vu Thao Trang, *J. Nanoparticle Res.* **2024**, *26*, DOI 10.1007/s11051-024-05984-7.