Supporting information for:

Industrial Waste-Derived Vanadium Recovery for High-Performance $Na_3V_2(PO_4)_2F_3$@C Sodium-ion battery application

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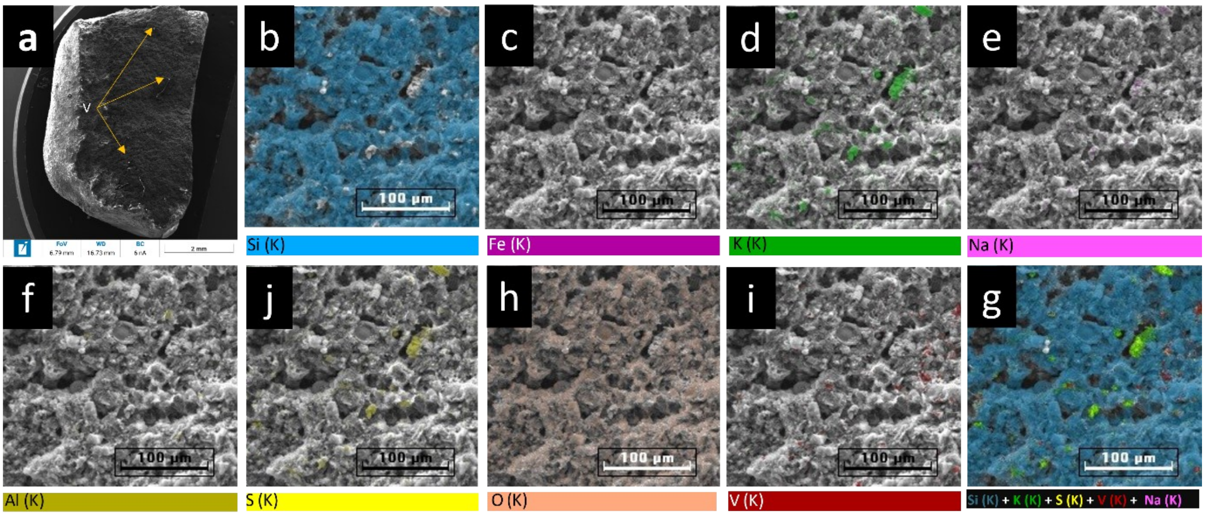

Fig. S1 (a) Sem image and (b-g) EDX mapping of vanadium spent catalyst

Table S1 ICP analysis of the recycled V_2O_5 synthesized via sol-gel method from stripped solution compared to the commercial one

Element	**Unit**	**V_2O_5 powder**	
		(Commercial >98%)	(Recycled)
V	**wt%**	55	55.996
Al	**ppm**	<5	17
Co		<10	<10
Cr		46	232
Fe		10	50
Na		30	47
Ca		<10	73
K		12	57

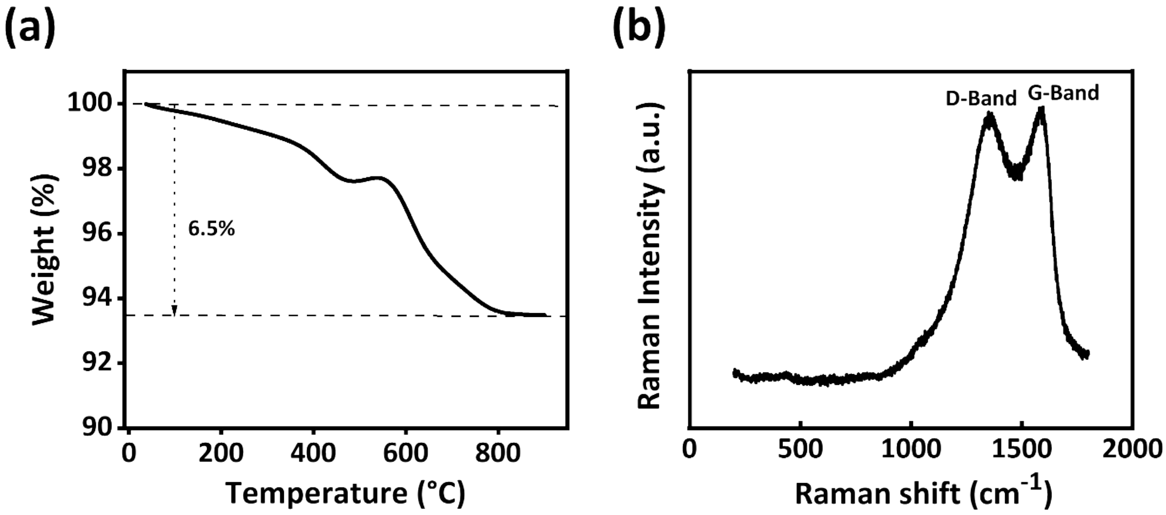

Fig. S2 (a) Thermogravimetric analysis, and (b) Raman spectroscopy of the as synthesized NVPF@C

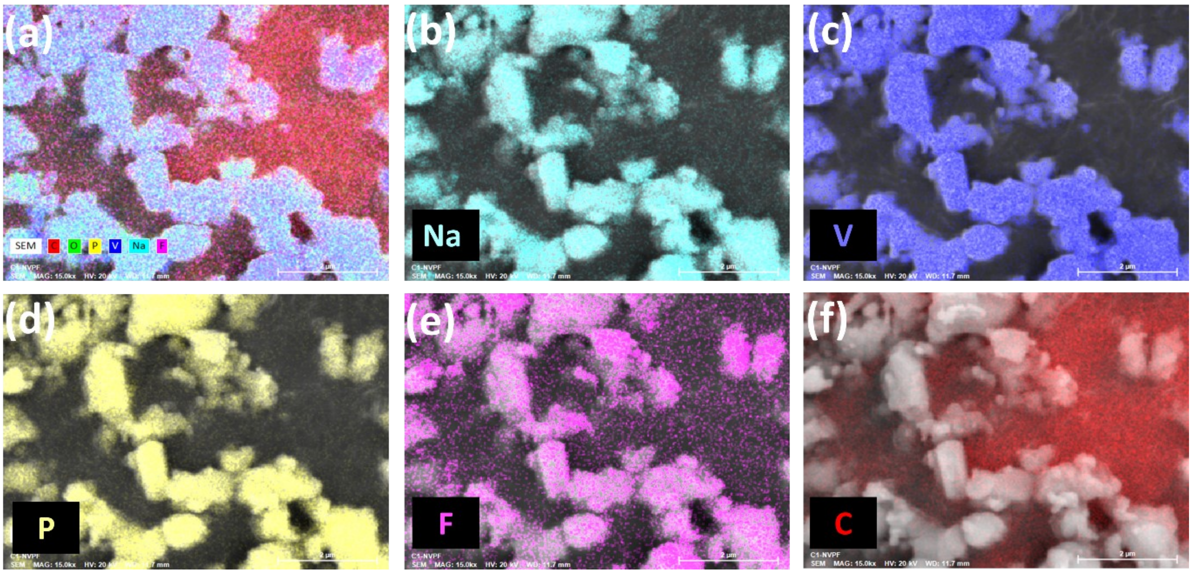

Fig. S3 EDX mapping of the as prepared NVPF@C from recycled vanadium pentoxide

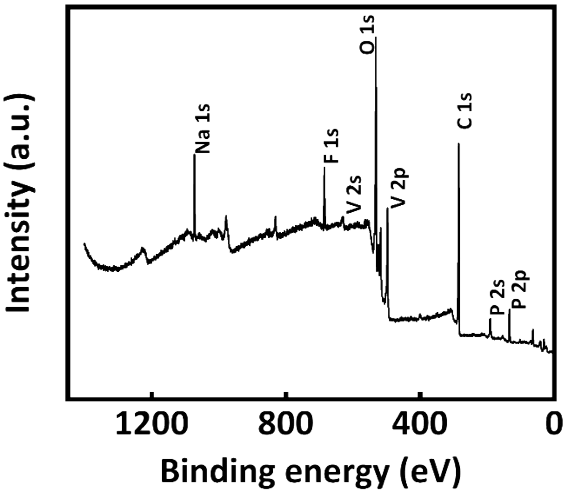

Fig. S4 Survey XPS spectrum of NVPF@C synthesized from recovered V_2O_5.

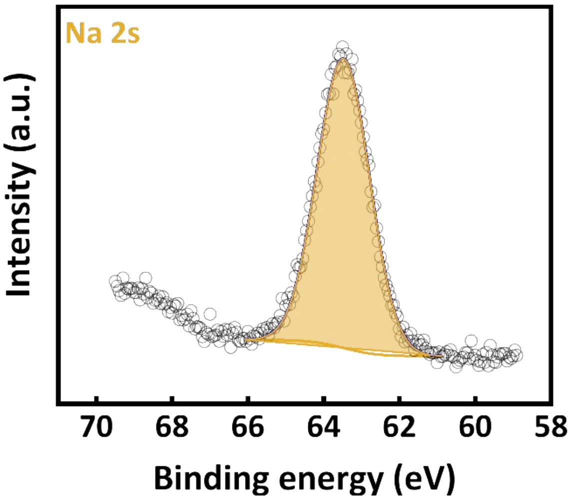

Fig. S5 High-resolution XPS spectra of NVPF@C : Na 2s synthesized from recovered V_2O_5.

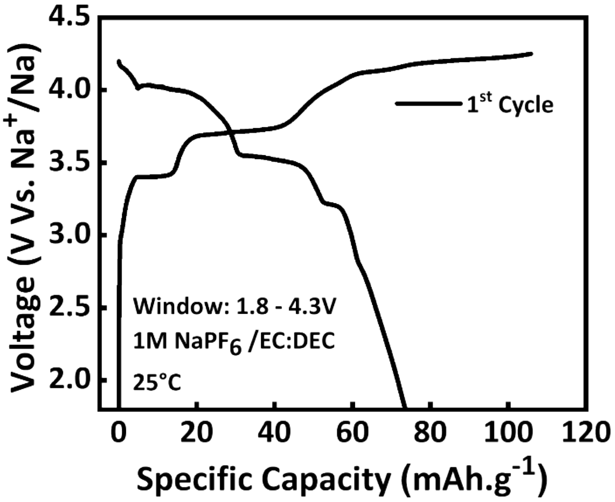

Fig. S6 Charge discharge profile of NVPF@C synthesized from commercial V_2O_5

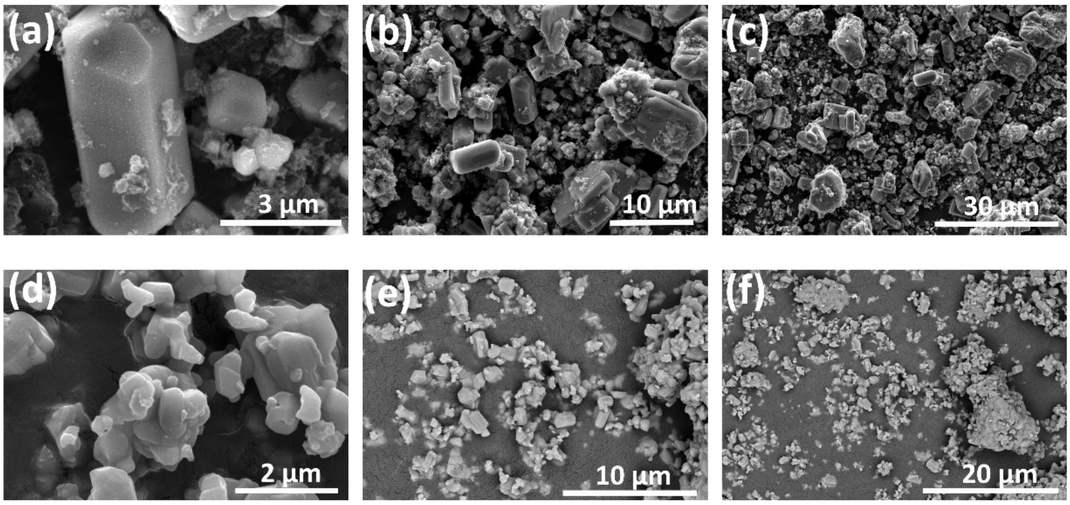

Fig. S7 (a-c) SEM images of the NVPF@C materials synthesized from recycled V_2O_5 and (d-f) commercial V_2O_5

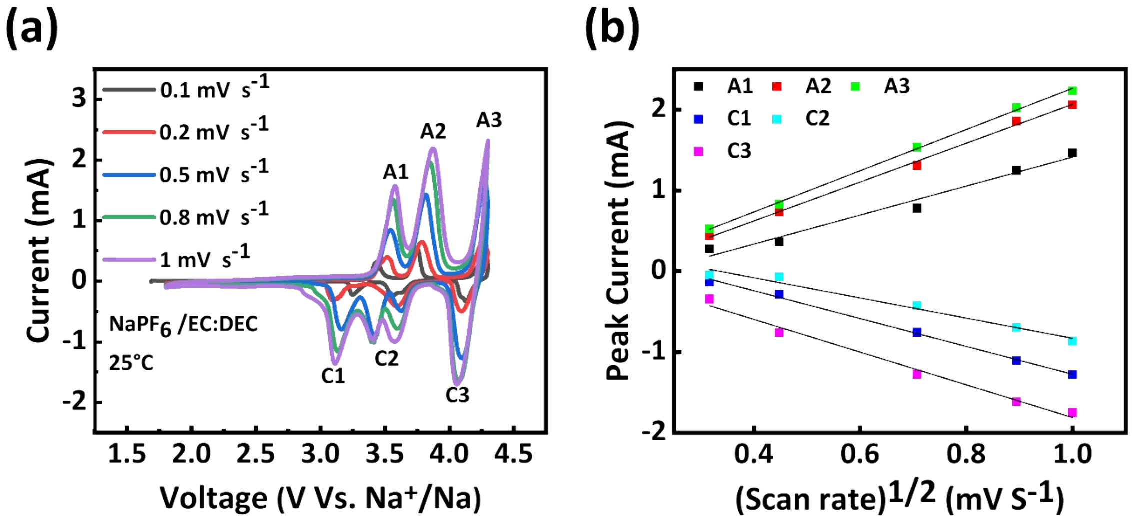

Fig. S8 (a) Cyclic voltammetry multi-rate of NVPF@C cathode material and (b) Peak current (Ip) plotted against the square root of the scan rate ($v^{1/2}$))

Environmental assessment of reductive leaching process

In this environmental assessment, only the reductive leaching step was incorporated into the OpenLCA model, as it represents the most sensitive stage of the recycling process. This step directly influences gaseous outputs such as CO_2 and SO_2 due to the acid reactions and the use of reducing agents. In this study, we compared the environmental assessment of the leaching section, which is the core of wastes treatment with two other leaching systems already reported in the literature.

Calculation Method

In this work, the environmental assessment was conducted using OpenLCA software, starting first by flow creation, process creation, and product system creation. In our model, the spent catalyst was classified as hazardous waste (Deposited goods/Hazardous waste-chemical), while the raw chemicals were defined as manufacturing chemicals. The water input was considered as industrial water, and the electricity mix (energy carriers and technologies/Electricity) was included as the main electricity source. The environmental impact assessment employed the ReCiPe Midpoint (H) method, with detailed input and output parameters and calculation results are provided in **Table S2 and S3**. The comparison was done using the already reported data of $NaHSO_3$ and oxalic acid leaching system.

1. **In this analysis work**, 1 kg of spent catalyst was used as the feedstock, with a (S/L) ratio of 5, a sulfuric acid concentration of 0.5 M, and a hydrazine monohydrate ($N_2H_4 \cdot H_2O$) to V_2O_5 molar ratio of 6. The leaching reaction was conducted at 80°C for 3 hours using a 600 W hot plate, resulting in a total energy consumption of 6480 kJ (1.8 kWh). Upon completion of the experiment, comprehensive material balances were performed to quantify all inputs and outputs. The collected process data were subsequently entered into the openLCA software to perform a detailed environmental impact assessment. The detailed input and output used in this analysis is summarized in **table S2**.
2. **Sulfuric acid/$NaHSO_3$ leaching**: A total of 1 kg of spent catalyst was subjected to leaching using a S/L ratio of 10 g/L in a 1 mol/L sulfuric acid solution. The leaching process was conducted at 80 °C for 4 hours, utilizing a 1500 W water bath, resulting in an energy consumption of 21,600 kJ.
3. **Oxalic acid leaching:** For the oxalic acid leaching process, 1 kg of spent catalyst was used as the feedstock, with a S/L ratio of 20 g/L. A 1 mol/L oxalic acid solution was employed as the leaching agent, and comprehensive material accountancy was conducted throughout the procedure. The leaching reaction was performed at 90 °C for 3 hours using a water bath operating at 1500 W.

Table S2 Input and output parameters of the reductive acidic leaching using hydrazine monohydrate and sulfuric acid.

		Category	unity	Amount
Input	**Electricity mix**	Energy carriers and technologies/Electricity	kWh	1.8
	Spent catalyst	Deposited goods/Hazardous waste	Kg	1
	Hydrazine monohydrate	Inorganic Chemical		0.051
	Sulfuric acid, at plant	Inorganic Chemical		0.138
	Water for industrial use	Water		4.862
Output	**Vanadium solution**	Inorganic Chemical		4.95
	Chemical waste, inert	Wastes/Production residues		0.95
	Water vapour	Elementary flows/Emission to air		0.151

Calculation results

Table S3 Environmental gas emission data of the proposed process compared to the reported one.

Technology	Raw materials	CO_2(g)	SO_2(g)	Ref.
This Work	1kg spent catalyst	267	0.74	**This Work**
Reductive leaching	1kg spent catalyst	850.0	41.533	2
Oxalic acid leaching	1kg spent catalyst	8097.4	49.383	1

Improvements in the leaching process directly reduced environmental emissions. By optimizing the S/L ratio, acid concentration, reaction conditions, and the choice of reductive agent, both energy consumption and process duration were minimized. For instance, hydrazine-assisted leaching at 80 °C for 3 h consumed only 1.8 kWh, markedly lower than the 6 kWh and 21.6 kWh required for sulfuric acid/$NaHSO_3$ and oxalic acid leaching, respectively. These reductions in energy and reagent use directly translate to lower CO_2 and SO_2 emissions, as confirmed by OpenLCA, highlighting that more efficient process conditions significantly decrease the environmental impact of vanadium recovery.

Table S4 Main works investigated the leaching of vanadium spent catalyst using different reductive acidic leaching system compared to our work

Leaching System	**Vanadium Recovery (%)**	**Leaching Conditions**	**By-products / Gas Emissions**	**References**
H_2SO_4 + $N_2H_4 \cdot H_2O$ (This work)	97.8	80 °C, 3 h, 0.5 M H_2SO_4 $N_2H_4 \cdot H_2O$ to V_2O_5 molar ratio of 6, L/S ratio of 5 mL/g,	Recyclable silica/ Mainly N_2 (non-toxic)	**This work**
H_2SO_4 + $NaHSO_3$	93.6	80°C, 0.5 mol/L H_2SO_4, n ($NaHSO_3$): n (V_2O_5) = 3, L/S ratio of 10 mL/g.	CO_2/SO_2 (toxic gas),	2
H_2SO_4 + Na_2SO_3	88.26	90 °C, 1 h, 2 M H_2SO_4, Na_2SO_3 excess coefficient of 1.5, L/S of 8:1 mL/g.	SO_2 (toxic gas)	3
Oxalic acid	84	90°C, 3 h, 1 M $C_2H_2O_4$, L/S ratio of 20mL/g.	Enormous CO_2 emissions from oxalic acid decomposition; significant SO_2 and NMVOC emissions	1
Citric acid +H_2O_2	95	50°C, 2 h, 0.1 M $C_6H_8O_7$, 0.1 M H_2O_2	CO_2 emissions from citric acid decomposition	4
Oxalic +H_2SO_4	90.13	90°C, 4 h, 4 m H_2SO_4, L/S ratio of 3	Vanadium rich slag (Require more recycling)/ CO_2 emissions from oxalic acid decomposition	5

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