Supplementary Material

Continuous production prototype of scaling graphene oxide/carbon nanotubes composite synthesis towards efficient hydrogen storage

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Figure S1. The photo of 100 g composite products.

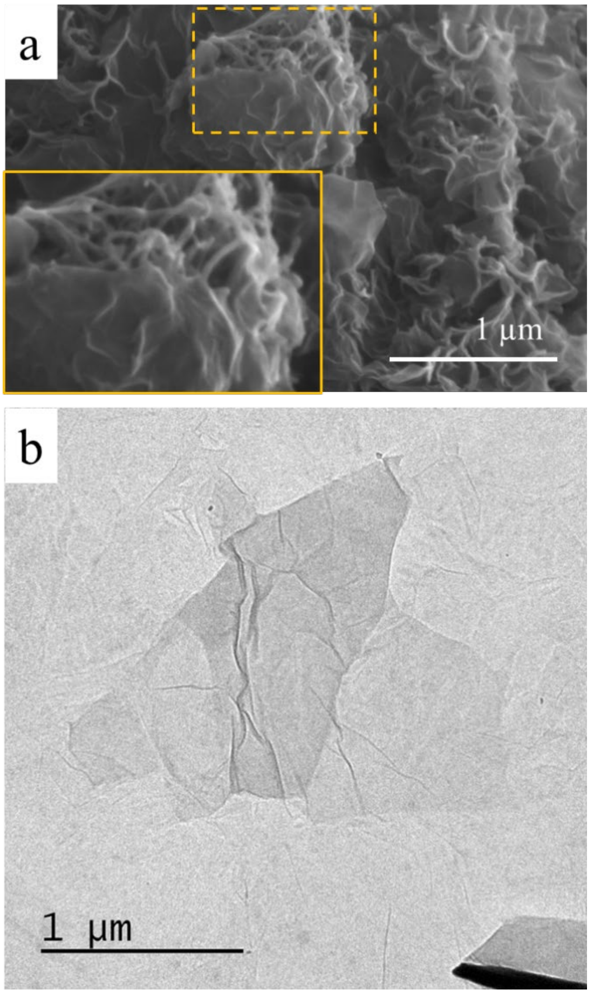

Figure S2. High-magnification (a) SEM image of GO/MWCNTs (inset: zoomed in of orange part) and (b) TEM image of GO.

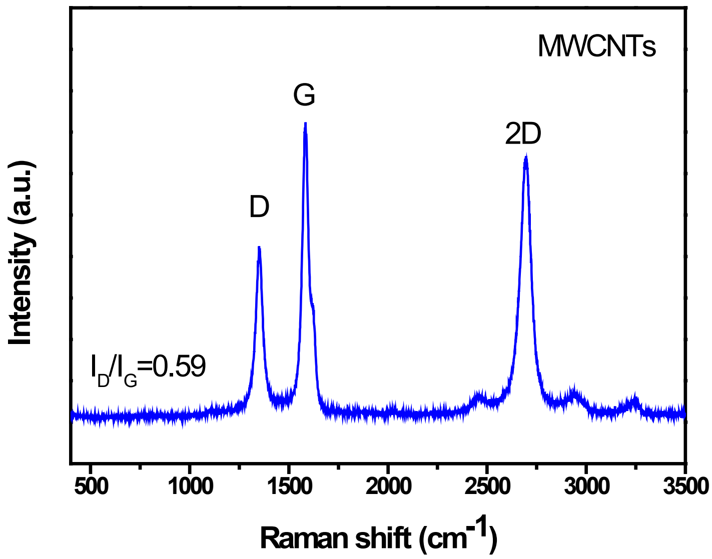

Figure S3. Raman spectra of MWCNTs.

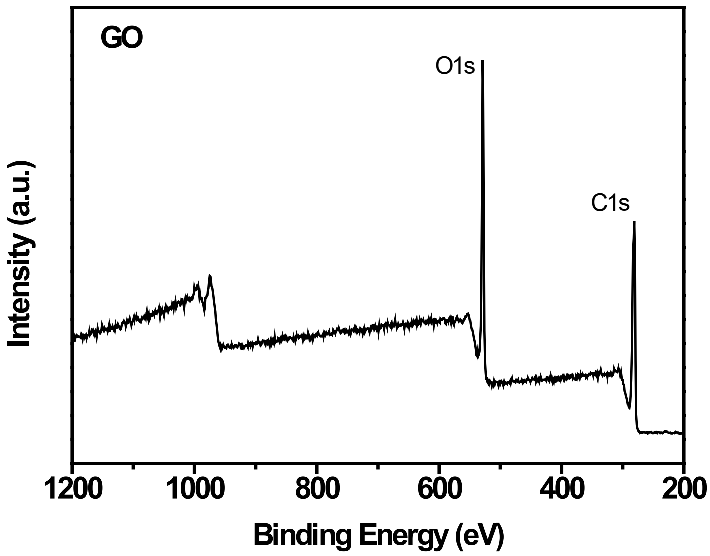

Figure S4. XPS survey spectrum of GO.

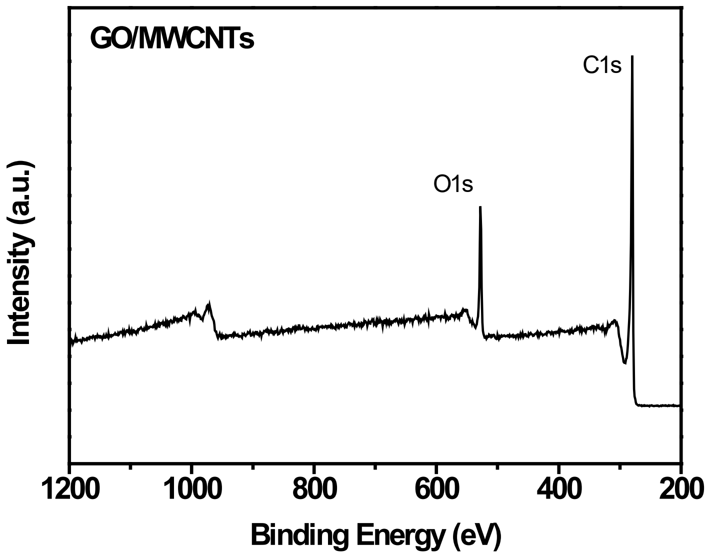

Figure S5. XPS survey spectrum of GO/MWCNTs.

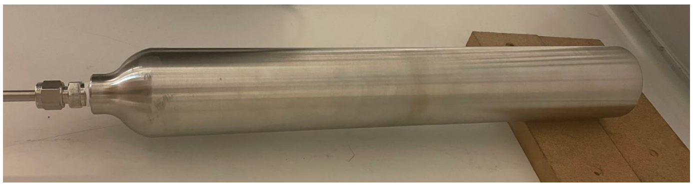

Figure S6. The photo of 500 mL cylinder.

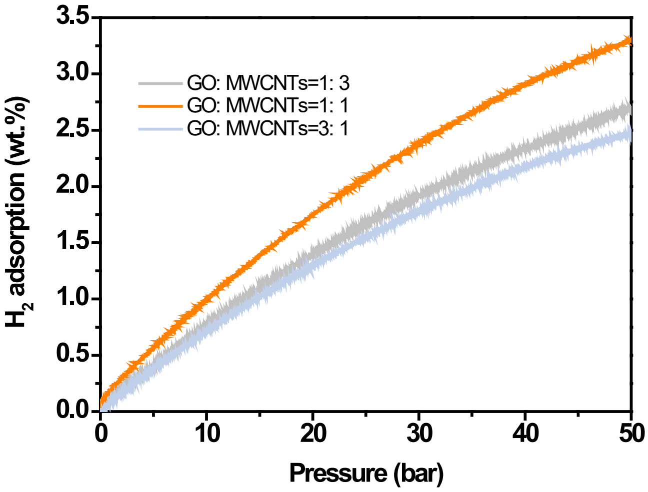

Figure S7. Hydrogen adsorption isotherms of different GO/MWCNTs ratios at 298 K and 50 bar.

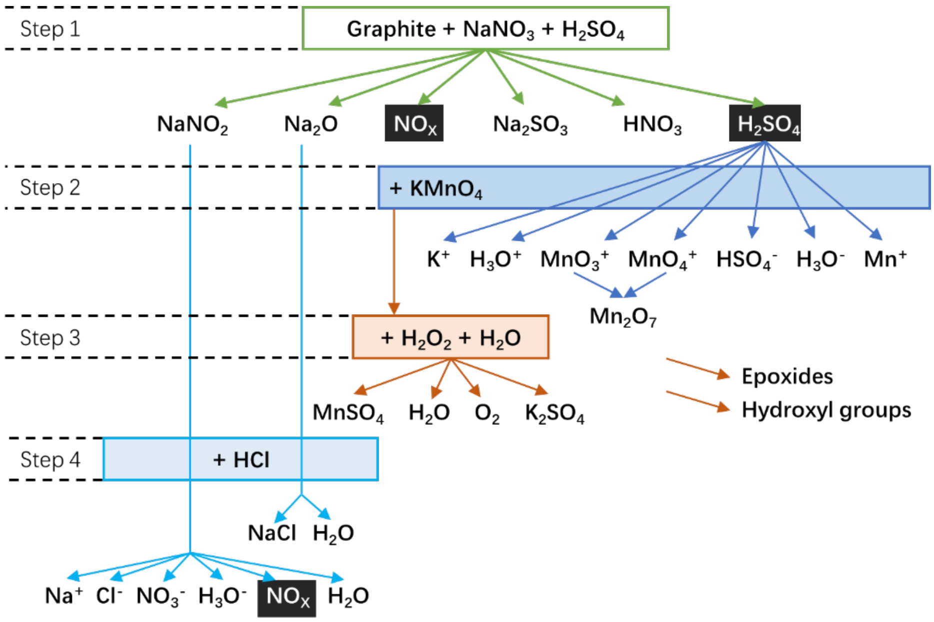

Figure S8. Reactions that occurred from Hummers' method for the production of the reduced graphene oxide. The chemicals in black background are those that are considered to be emitted in the scenario defined as "worst case" [1].

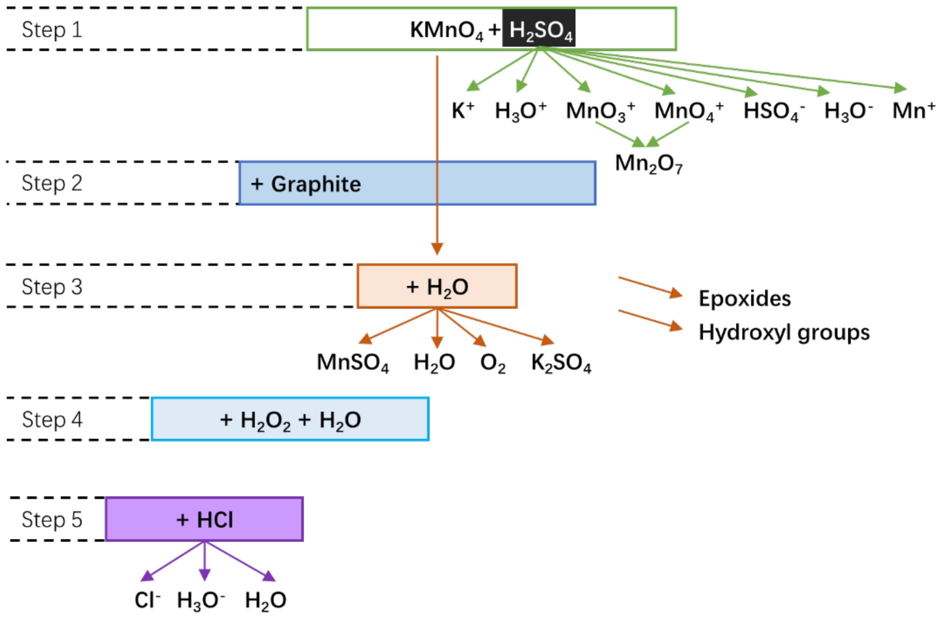

Figure S9. Reactions that can take place during our continuous production of the reduced graphene oxide. (To rate the environmental impact, our continuous production prototype, which generates fewer hazardous substances, has a relatively lower environmental impact and can be assigned an environmental impact index of 1. In contrast, the conventional Hummers' method, which produces a greater amount of hazardous substances, has a significantly higher environmental impact and can be assigned an index of 2.)

Table S1. The content of function groups in GO, MWCNTs, and GO/MWCNTs.

	GO	Binding energy	MWCNTs	Binding energy	GO/MWCNTs	Binding energy
C=C (sp^2)	45.15%	284.5 eV	43.07%	284.5 eV	46.48%	284.5 eV
C-C (sp^3)	10.09%	285.4 eV	27.71%	285.1 eV	22.85%	285.1 eV
C-O	31.34%	286.7 eV	13.99%	286.4 eV	19.80%	286.4 eV
C=O	9.95%	288.2 eV	9.42%	288.5 eV	4.55%	288.5 eV
O-C=O	1.61%	289.1 eV	2.42%	289.6 eV	2.14%	289.1 eV
π-π*	1.85%	290.9 eV	3.39%	291.4 eV	4.18%	290.1 eV

Table S2. Elemental analysis of GO, MWCNTs, and GO/MWCNTs.

	C (wt.%)	H (wt.%)	O (wt.%)	N (wt.%)	S (wt.%)	C/O ratio
GO	47.14	1.77	46.77	0.12	0.44	1.01
MWCNTs	99.27	0.51	0.85	0.27	0.39	116.68
GO/MWCNTs	93.36	0.88	7.41	0.34	0.48	12.60

Table S3. The comparison of different hydrogen materials.

Materials	Yield	Sample amount for test	Conditions (temperature)	Conditions (pressure)	Hydrogen capacity	Ref.
3D graphene	0.4 g	0.1-0.3 g	293 K	120 bar	1.25 wt.%	[2]
Ni-CNTs	0.4 g	N.R.	298 K	100 bar	0.87 wt.%	[3]
Ni_2(m-dobdc)	2.25 g	1.0-2.0 g	298 K	100 bar	1.24 wt.%	[4]
Cu^{I}_2(BBTA)	<0.1 g	0.2595 g	296 K	110 bar	0.48 wt.%	[5]
GO/MWCNTs	100 g[1]	75 g	298 K	50 bar	~3.1 wt.%	This work

[1](single feed batch), continuous production

Table S4. The calculation of unit energy consumption towards four synthesis processes.

Unit energy consumption (kWh/g)	GO			GO/MWCNTs			Ref.
	Electricity	Heat	Total	Electricity	Heat	Total	
Shear exfoliation in liquids	~0.048	-	~0.048	-	-	-	[6, 7]
Conventional Hummers' method	~0.0009	~0.0475	~0.0484	-	-	-	[8]
Lab-scale synthesis process	0.26	1.05	1.31	2.17	2.33	4.50	
Continuous production prototype	0.0065	0.015	0.0215	0.08307	0.08666	0.1697	

Table S5. The calculation of unit carbon footprint towards four synthesis processes.

Unit Carbon footprint (kg CO_2e/g)	GO	GO/MWCNTs
Shear exfoliation in liquids	0.8589	-
Conventional Hummers' method	0.0814	-
Lab-scale synthesis process	0.0887	0.22395
Continuous production prototype	0.0395	0.05835

The carbon footprint refers to the total amount of greenhouse gases (GHGs), primarily carbon dioxide, methane, and nitrous oxide (as defined by The Nature Conservancy), that may be emitted into the atmosphere during various stages of the value chain. These stages include raw material procurement, polymer manufacturing, final product transportation, and consumer use and disposal of the final product. The total carbon footprint (kg CO_2e) can be calculated as follows:

Total carbon footprint (kg CO_2e) = Chemicals + Electricity + Waste treatment + By-product emissions

Herein, the carbon emission factor of each chemical can be provided Ecoinvent database. The carbon emission factor of the Swiss electricity grid is 0.03817 kg CO_2e/kWh [9].

Table S6. The calculation of unit cost towards four synthesis processes.

Unit Cost (CHF/g)	GO			GO/MWCNTs		
	Chemical	Energy	Total	Chemical	Energy	Total
Shear exfoliation in liquids	2921.87	0.013	2921.88	-	-	-
Conventional Hummers' method	7.88	0.0132	7.8932	-	-	-
Lab-scale synthesis process	1.73	0.356	2.056	3.65	1.224	4.874
Continuous production prototype	1.73	0.006	1.736	3.65	0.046	3.696

* The cost of chemical: each chemical price × amount
* 2023 Swiss electricity price: 0.272 CHF /kWh [10, 11]

Table S7. Comparison between the continuous production prototype and the conventional Hummer method.

	Continuous production prototype	Conventional Hummers' method
Production Scale and Consistency	· Larger-scale production, enabling the synthesis of GO in significant quantities without the need for batch processing · Uniform product	· Smaller, laboratory scale, involves batch processing · Variability between different batches
Reaction Conditions	· Allow for more precise control over reaction parameters such as temperature, time, and the concentration of reactants · Improved heat and mass transfer, preventing explosions and temperature runaway	· Inconsistent reaction parameters for each batch processing · Less efficient heat and mass transfer, high explosive potential
Product Quality	· Consistent oxidation level · Uniform distribution of oxygen-containing functional groups	· Different oxidation level between batches and within the same batch · Less uniform distribution of oxygen-containing functional groups
Environmental Impact	· Reduced environmental impact · More energy-efficient	· Less efficient and more environmentally taxing (NO_x) · Higher energy consumption

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