Upcycling Linear Low-Density Polyethylene Waste into Graphene for High Mass Loading Supercapacitors

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Introduction

Upcycling plastic into advanced carbons, such as **graphene** and **porous carbon**, offers attractive options to manage waste streams by converting the plastic into carbon electrode materials for energy storage devices. Linear low-density polyethylene (LLDPE) is first bulk-oxidized with a facile and scalable method and then carbonized and catalytically graphenized into porous graphene materials. The LLDPE derived graphene (LLDPE-G) has a specific surface area up to 1,800 m^2/g and Raman I_D/I_G ratio of 0.85. When used as electrode material for symmetric supercapacitor, LLDPE-G possesses outstanding specific capacitance and excellent areal capacitance. Moreover, LLDPE-G exhibits exceptional cycling stability with capacitance retention of 95.8% after 100,000 cycles. Last, but not least, potassium chloride (KCl) is recycled and reused over 3 cycles with material quality and electrocapacitive performance of LLDPE-G retained and verified after each cycle.

Method of Upcycling LLDPE Waste Into Graphene

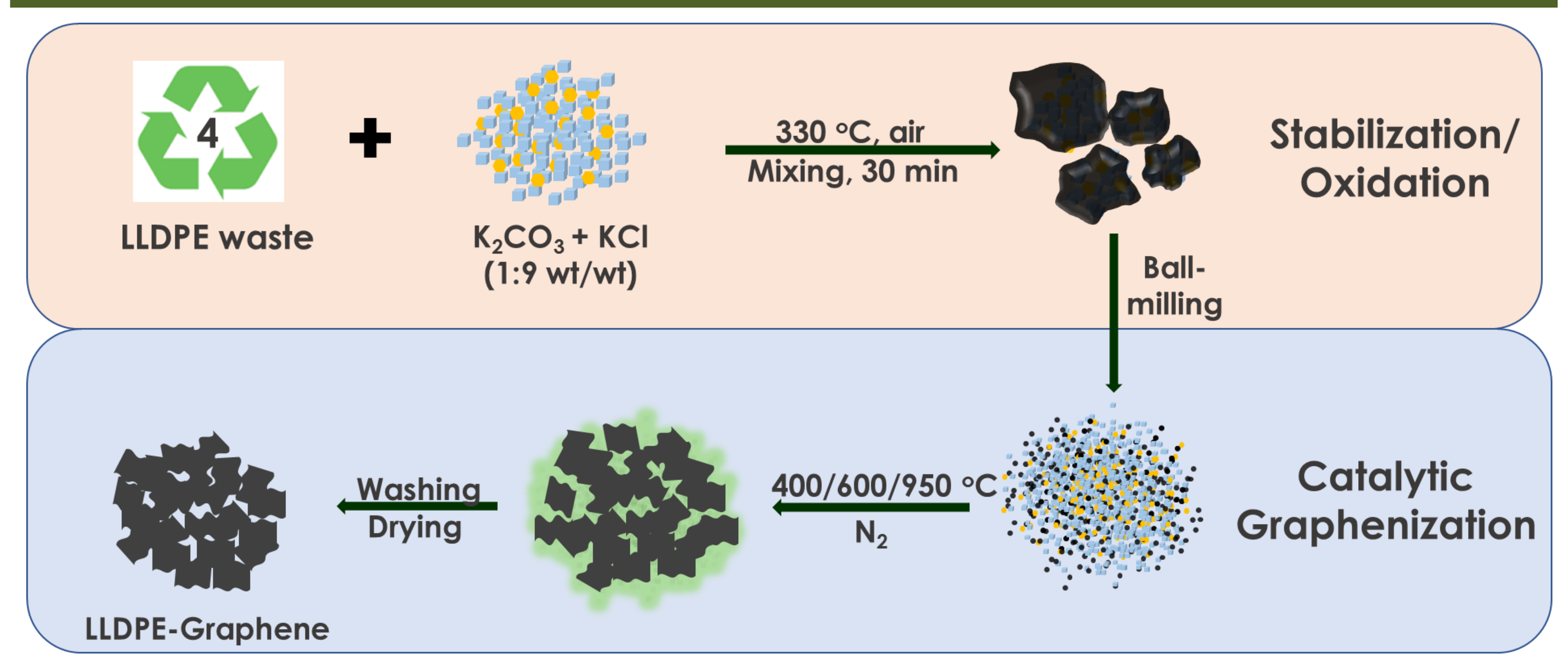


Figure 1. Schematic illustration of synthesis procedure of upcycling LLDPE waste into graphene.[1]

Step 1: Stabilization/oxidation: Polyethylene is notoriously difficult to upcycle because it decomposes into light gases at ~350-400 °C which prevents processing it at higher temperatures to convert it into electrode materials. We address this challenge by using an air-based, thermo-oxidative, process, along with a solid surface area enhancer, which heats LLDPE to 290-310 °C to initiate oxidation and cross-linking of its alkyl chains.

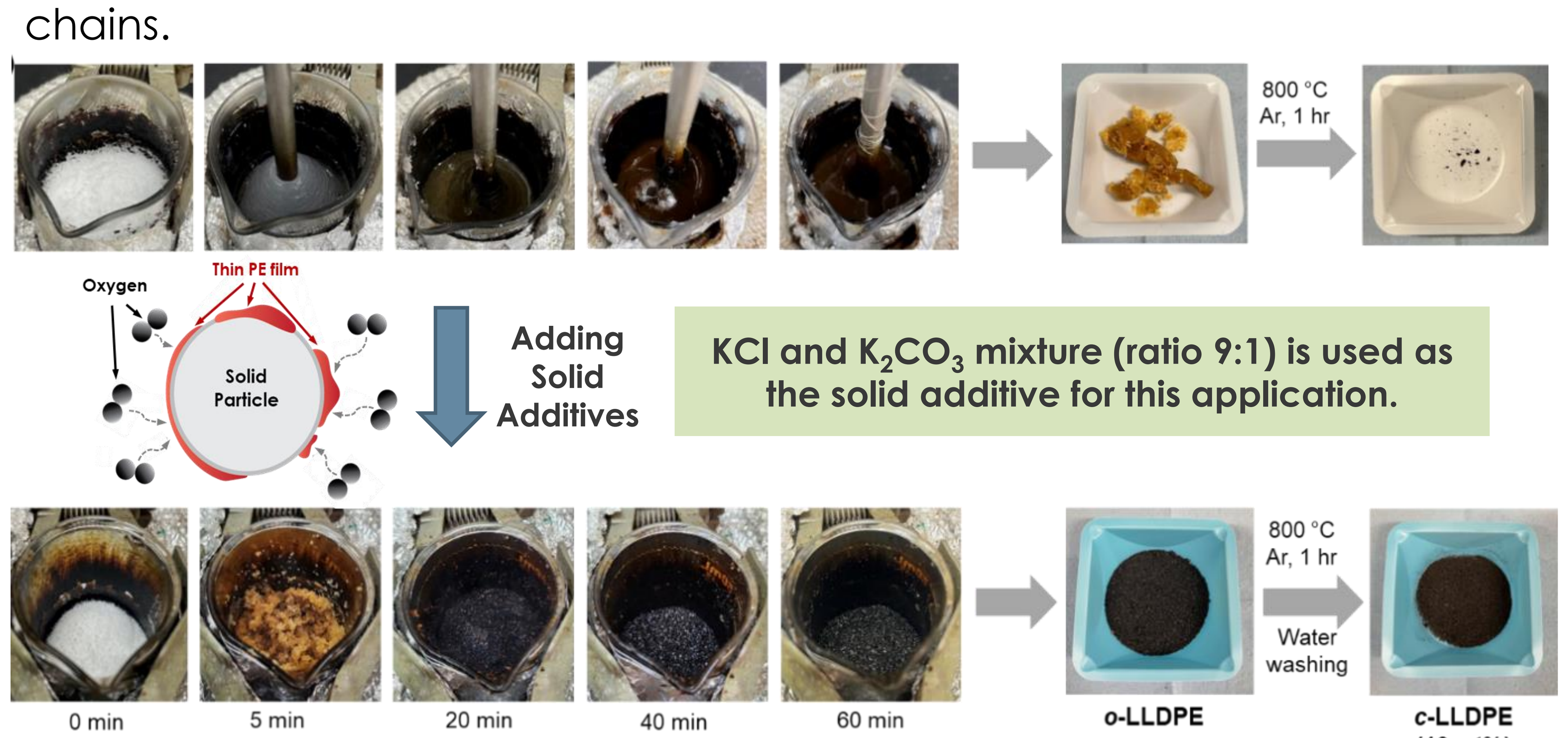
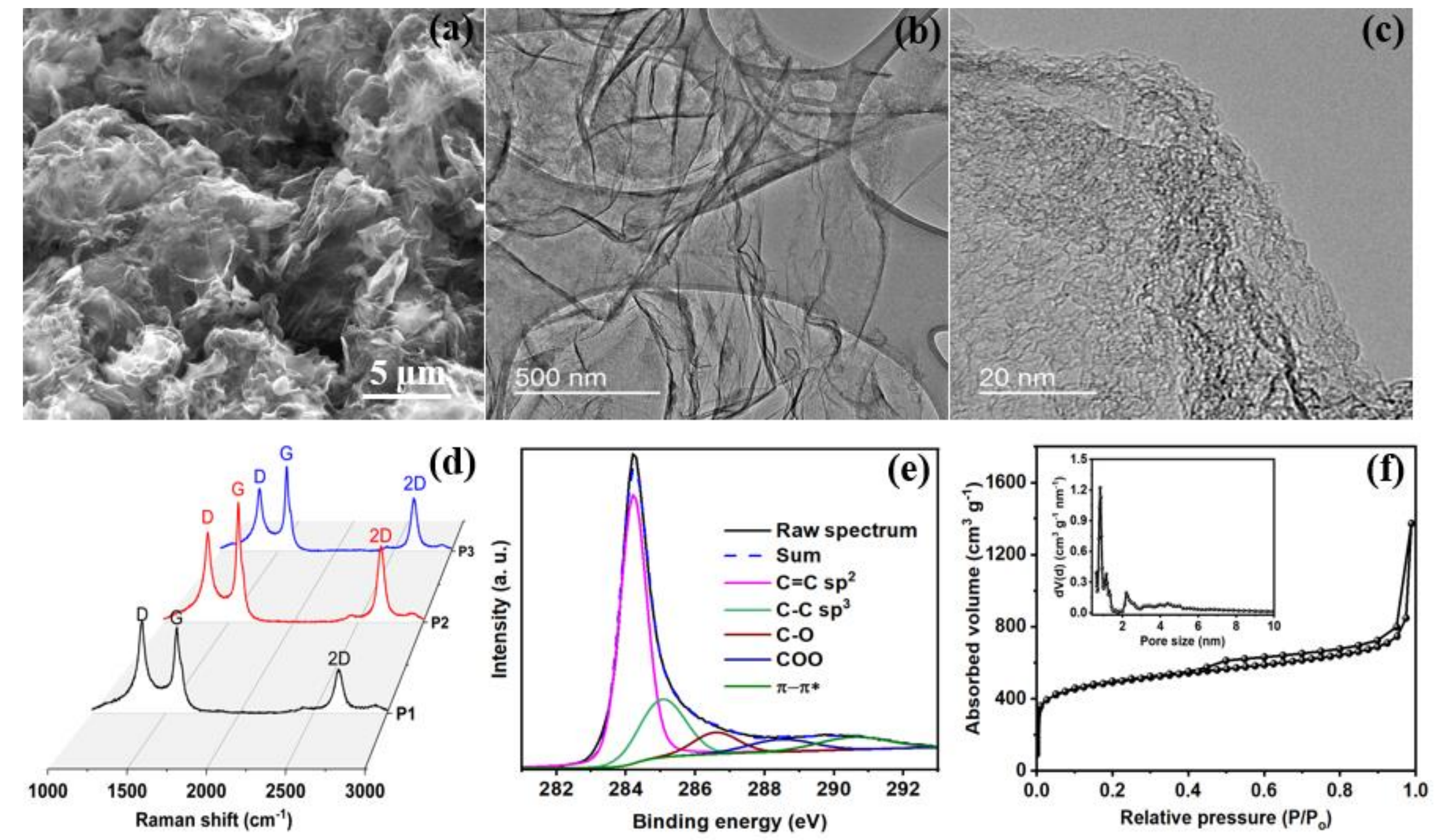


Figure 2. Solid additive effect during the bulk oxidation process.[2]

Step 2: Catalytic graphenization: After the pre-treatment step, the mixture of oxidized LLDPE, K_2CO_3, and KCl was ball milled to create a homogeneous powder, followed by thermal treatment in inert nitrogen atmosphere for additional stabilization, carbonization, and catalytic graphenization. The K_2CO_3 from the salt mixture is the catalyst for the graphenization.

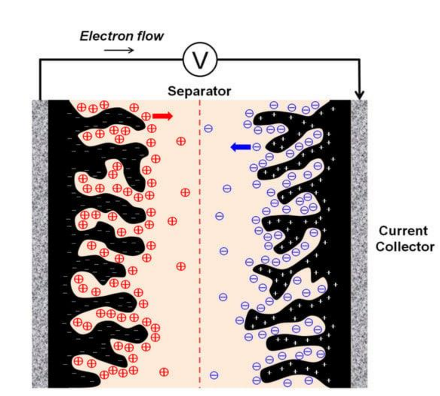
LLDPE Derived Porous Graphene Product

The physical and chemical properties of LLDPE-G is specifically tuned for the supercapacitor application.



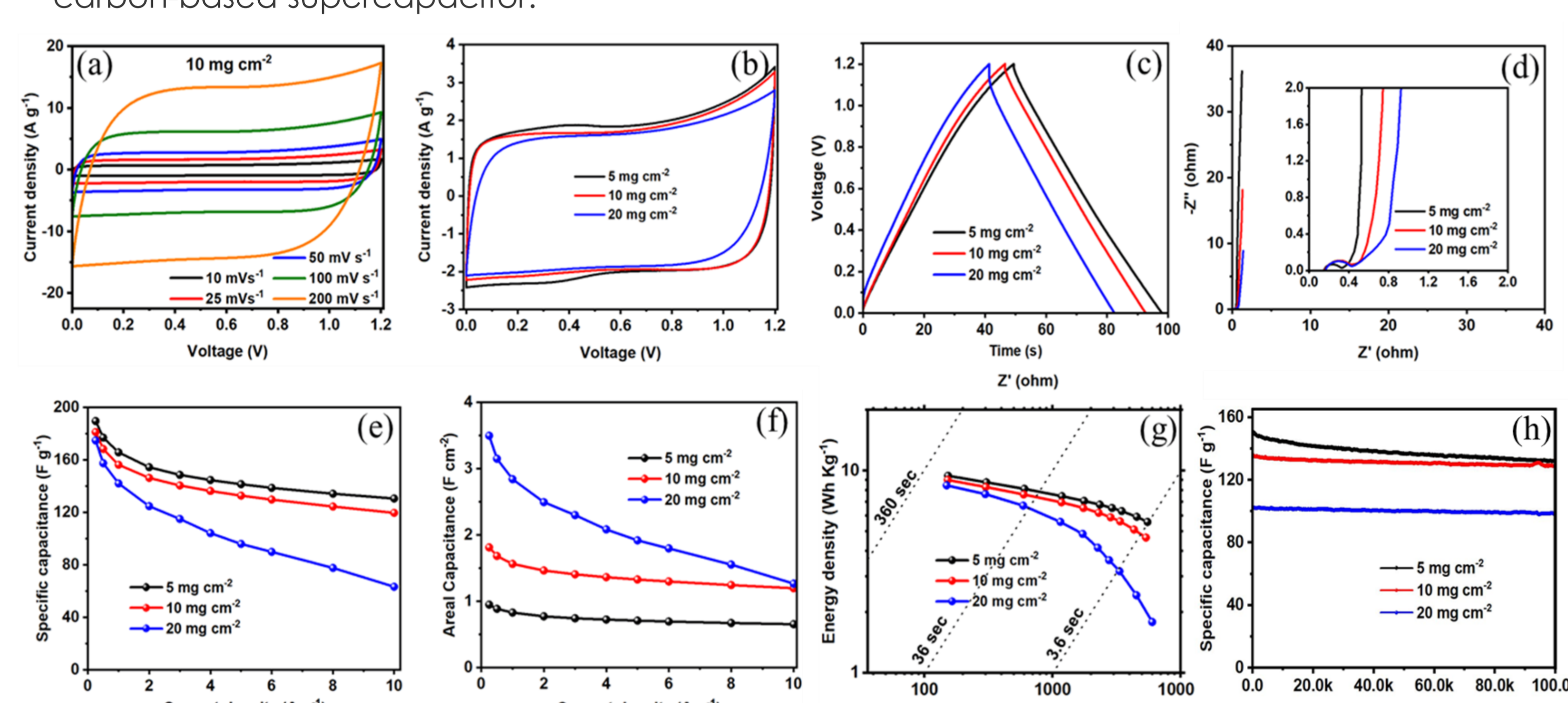
	Yield	BET Surface Area (m^2 g^{-1})	Raman I_d/I_g	Oxygen Content at%
LLDPE-G	7.5 wt.%	1,800	0.85	2

Supercapacitor Test



Schematic illustration of the porous carbon-based supercapacitor.[4]

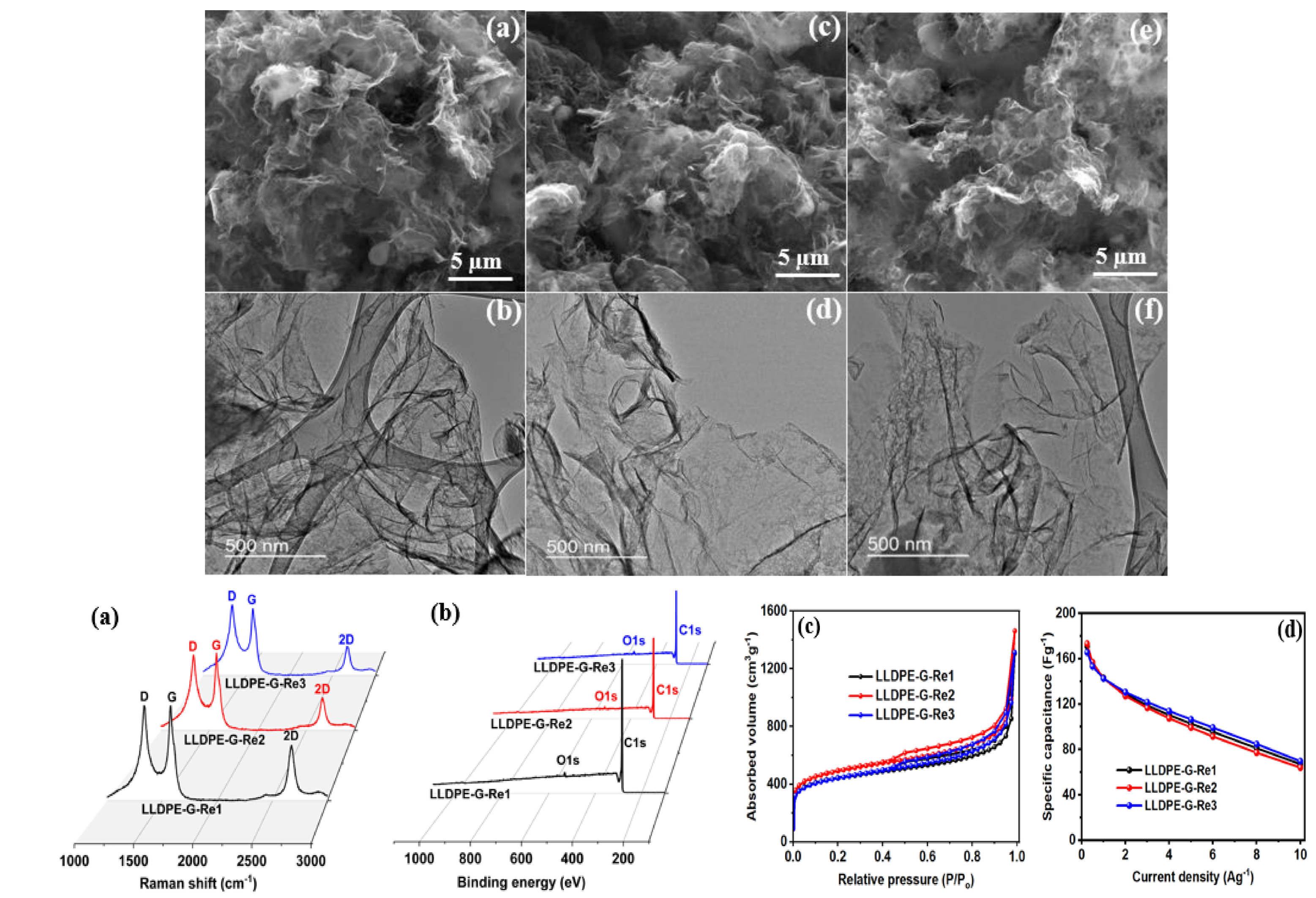
To be relevant for practical applications, the LLDPE-G was evaluated as a supercapacitor electrode in two-electrode symmetric cells using 1M H_2SO_4 aqueous electrolyte with an areal mass loading up to 20.0 mgcm^{-2}. PE-derived graphene has comparable or even better electrochemical capacitive properties, which demonstrates both high areal capacitance and high power density[3].



	BET Surface Area (m^2 g^{-1})	Specific Capacity at 0.25 A/g (F g^{-1})	Energy Density (Wh kg^{-1})	Capacity Retention After 100,000 Cycles
LLDPE-G	1,800	175	7.86	95.8
Commercial SOTA YP-50F	1,760	150	6.27	88.5

Recycle/Reuse the KCl

The filtrate collected after Step 2 of graphenization was neutralized with $KHCO_3$ and then dried to recover the mixture of KCl and K_2CO_3 for reuse. The recovery rate of the salt mixture is up to 95%. The subsequent graphene products have comparable qualities compared to original porous graphene.



Summary

1. Polyethylene materials are upcycled into porous graphene with a facile and scalable bulk stabilization/oxidation method followed by an inexpensive catalytic graphenization synthesis.
2. The porous graphene has a BET surface area up to 1,800 m^{2}g^{-1} and a hierarchical pore size distribution.
3. The graphene demonstrates excellent electrochemical capacitive properties as a supercapacitor electrode material, which outperforms the commercial porous carbon electrode.
4. The salt mixture (KCl and K_2CO_3) used in the process can be easily recycled and reused for at least 3 cycles.

References

1. Y. Gao et al., Chemical Engineering Journal(accepted) ***2.*** *K. Kim. et al., ACS Sustainable Resource Management (in review)* ***3.*** *V. H. Pham et al., Small Methods 2024, 2301426.* ***4.*** *H.-C. et al., Materials 2020, 13(18), 4215*

Disclaimer

Acknowledgement

This work was performed in support of the U.S. Department of Energy's (DOE) Laboratory Directed Research & Development (LDRD) program and executed through the National Energy Technology Laboratory (NETL) Research & Innovation Center consistent with the authorizing legislation and DOE LDRD Order 413.2C.

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