

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



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Authors and Contact Information



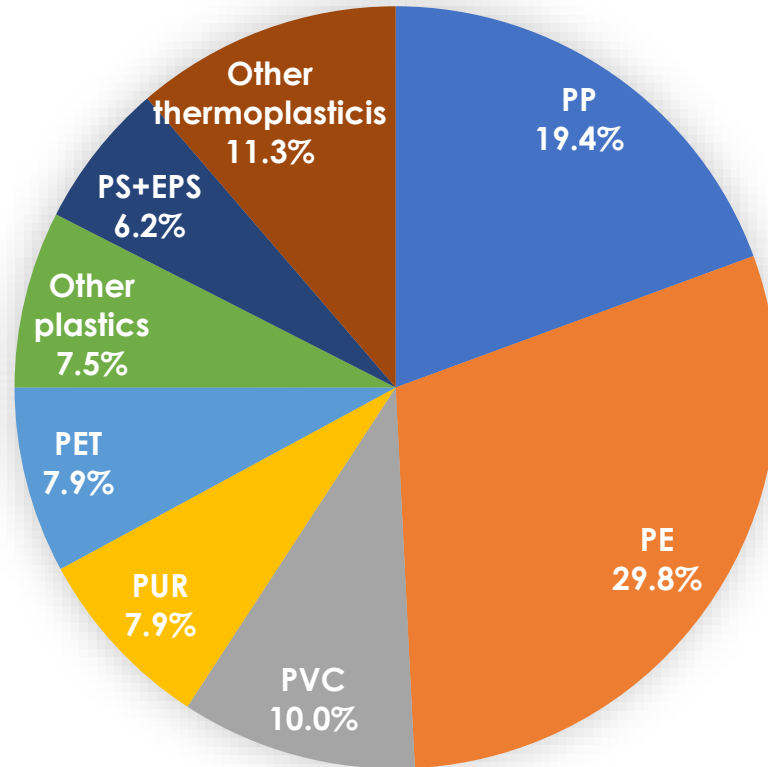
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1. **Background - Plastic Waste Recycling and Upcycling**
2. **Upcycling Polyethylene (PE) Waste into Turbostratic Graphene**
3. **PE-Derived Graphene as Electrode Material for High Mass Loading Supercapacitor**
4. **Recycling Solid Additives**
5. **Summary**

Background - Plastic Recycling and Upcycling

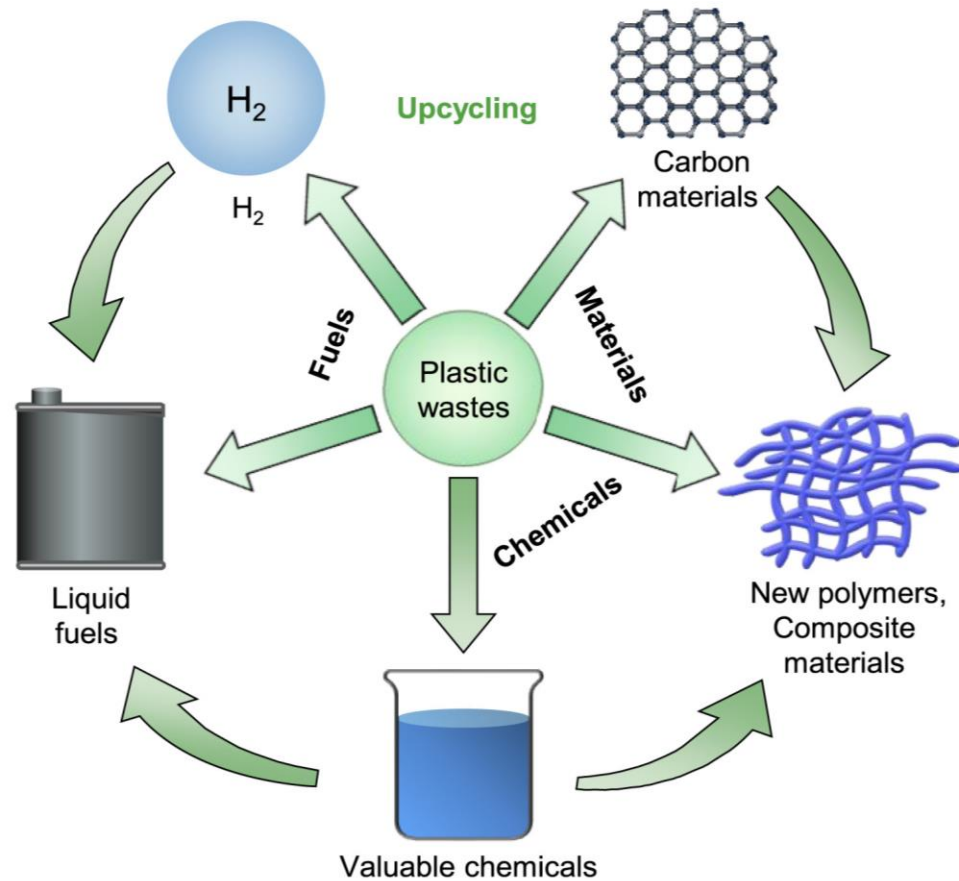


Distribution of global plastic production in 2019, by type.

(<https://plasticseurope.org/knowledge-hub/plastics-the-facts-2020/>)

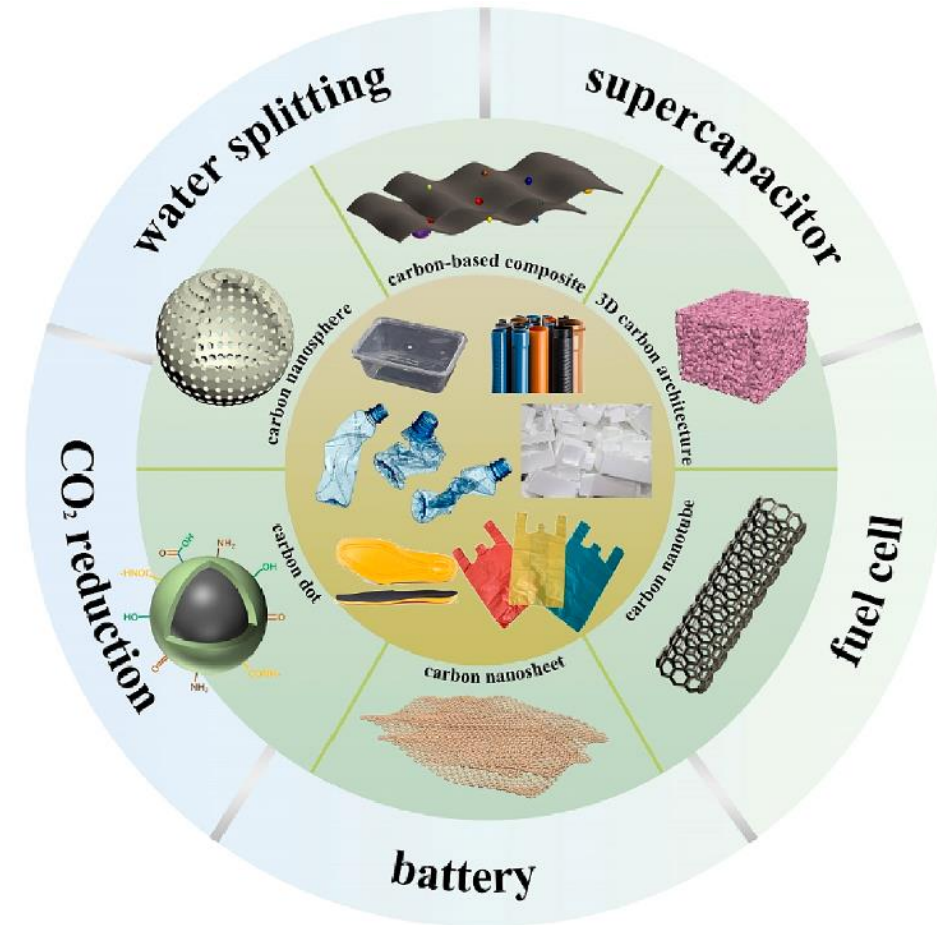
- 8 billion tons of plastic waste produced since 1950, but only 6-7% recycled.
- Current recycling downgrades technical and economic value of plastic, limiting utility.
- PE is difficult to recycle/reuse; decomposes during thermal processing.

Background - Plastic Recycling and Upcycling



Upcycling toward value-added products.

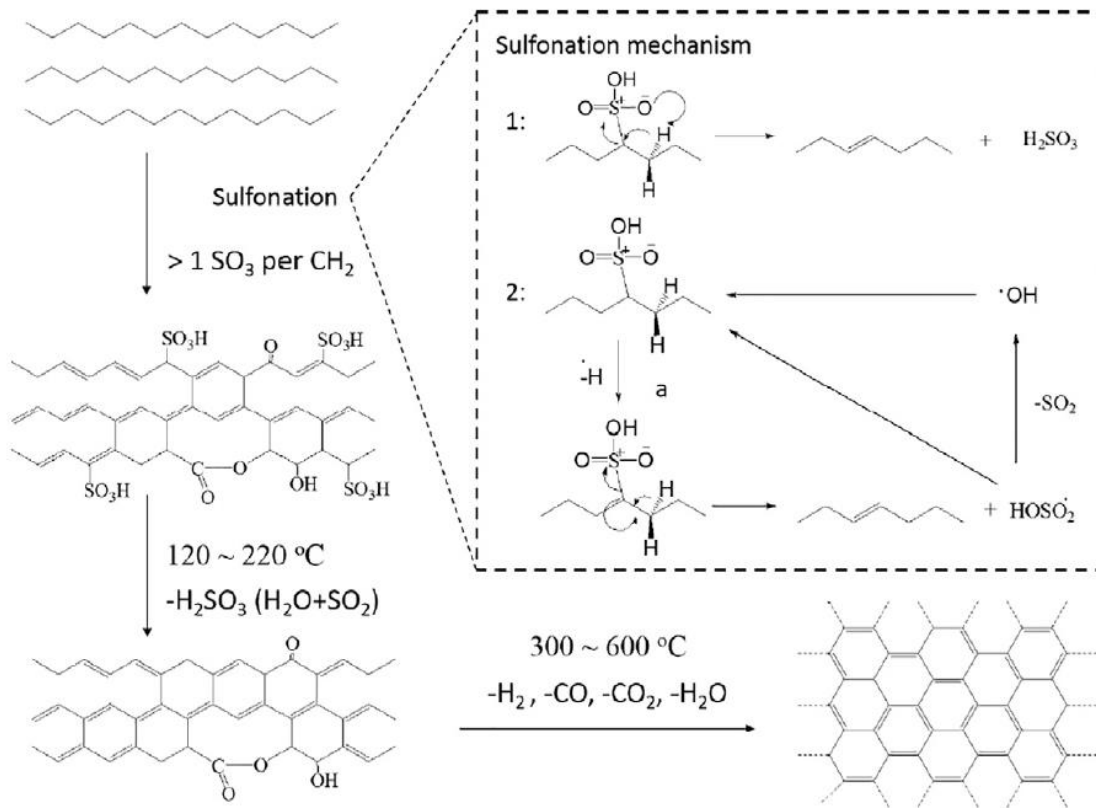
(Cell Reports Physical Science, 2021, 2, 100514)



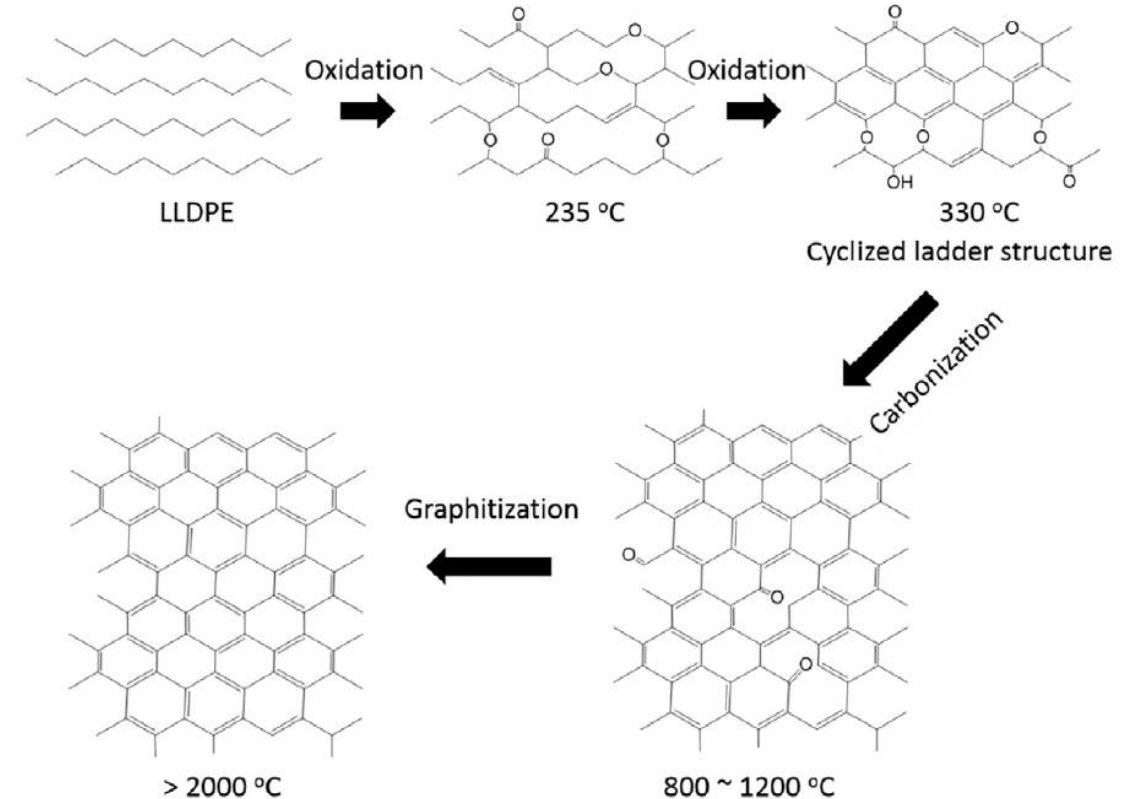
Carbon-based materials derived from typical plastics waste and their applications. *(Chemical Engineering Journal 2023, 461, 141962)*

Upcycling PE Waste into Turbostratic Graphene

Thermal Crosslinking and Carbonization of PE



Sulfonation and carbonization of PE.

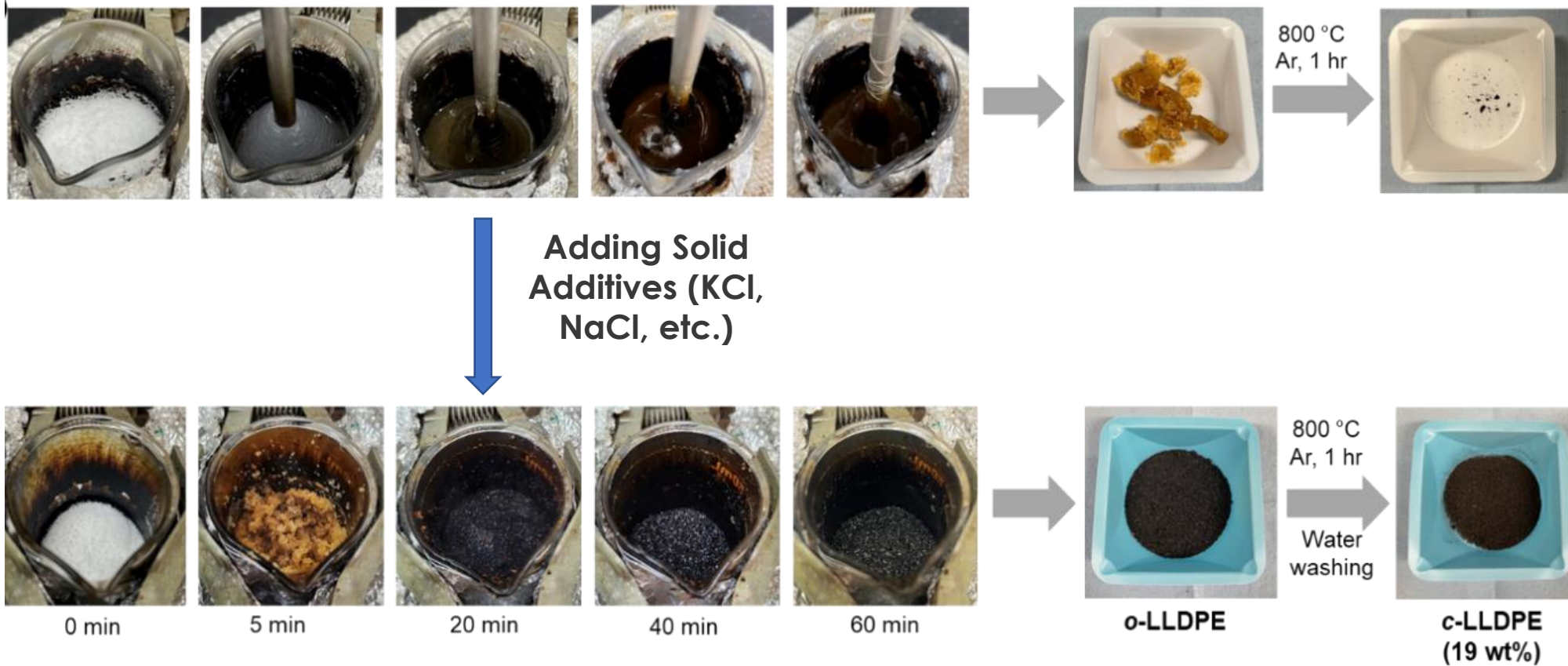


Thermal oxidation, carbonization and graphitization of PE.

(Science of the Total Environment, 2020, 710, 136250)

Upcycling PE Waste into Turbostratic Graphene

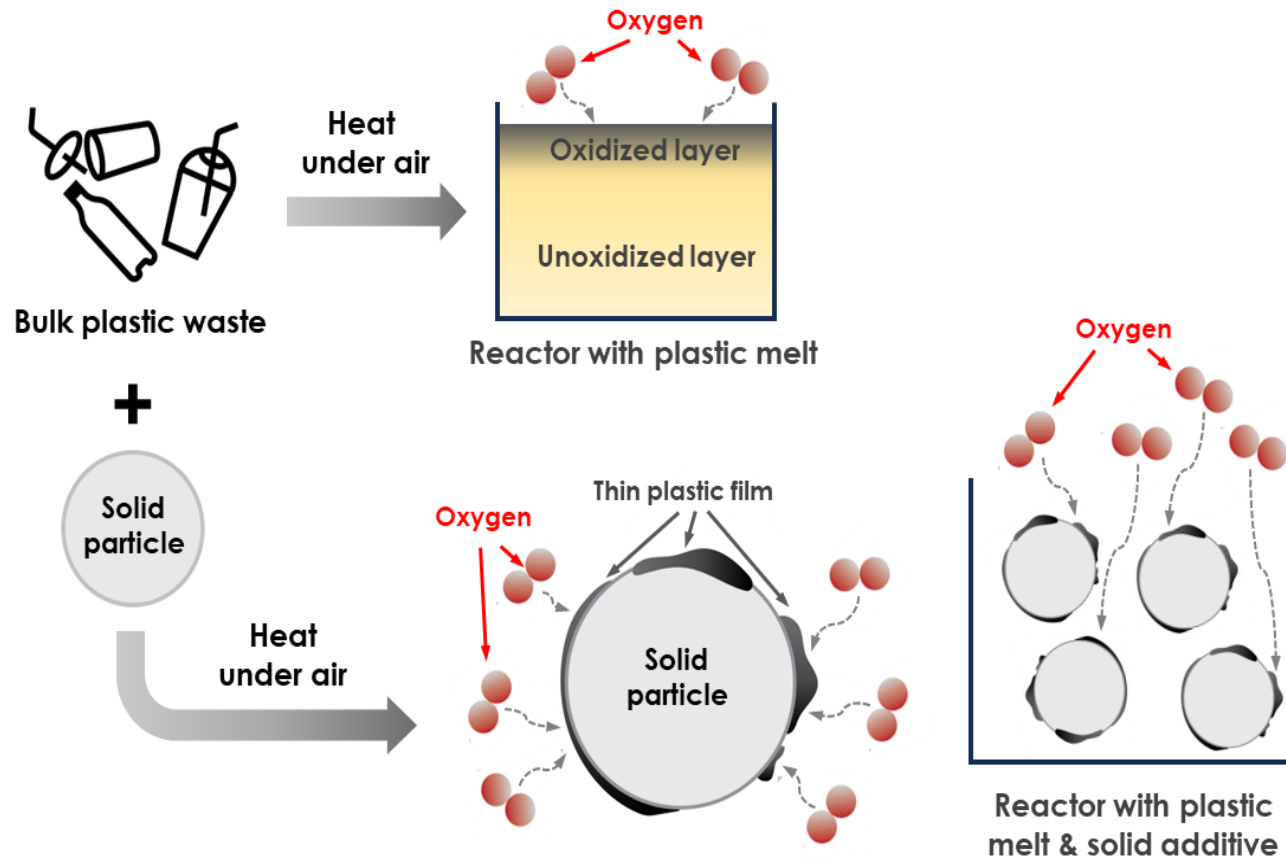
Thermal Crosslinking and Carbonization of PE



Solid additive effect during the bulk thermal oxidation process.

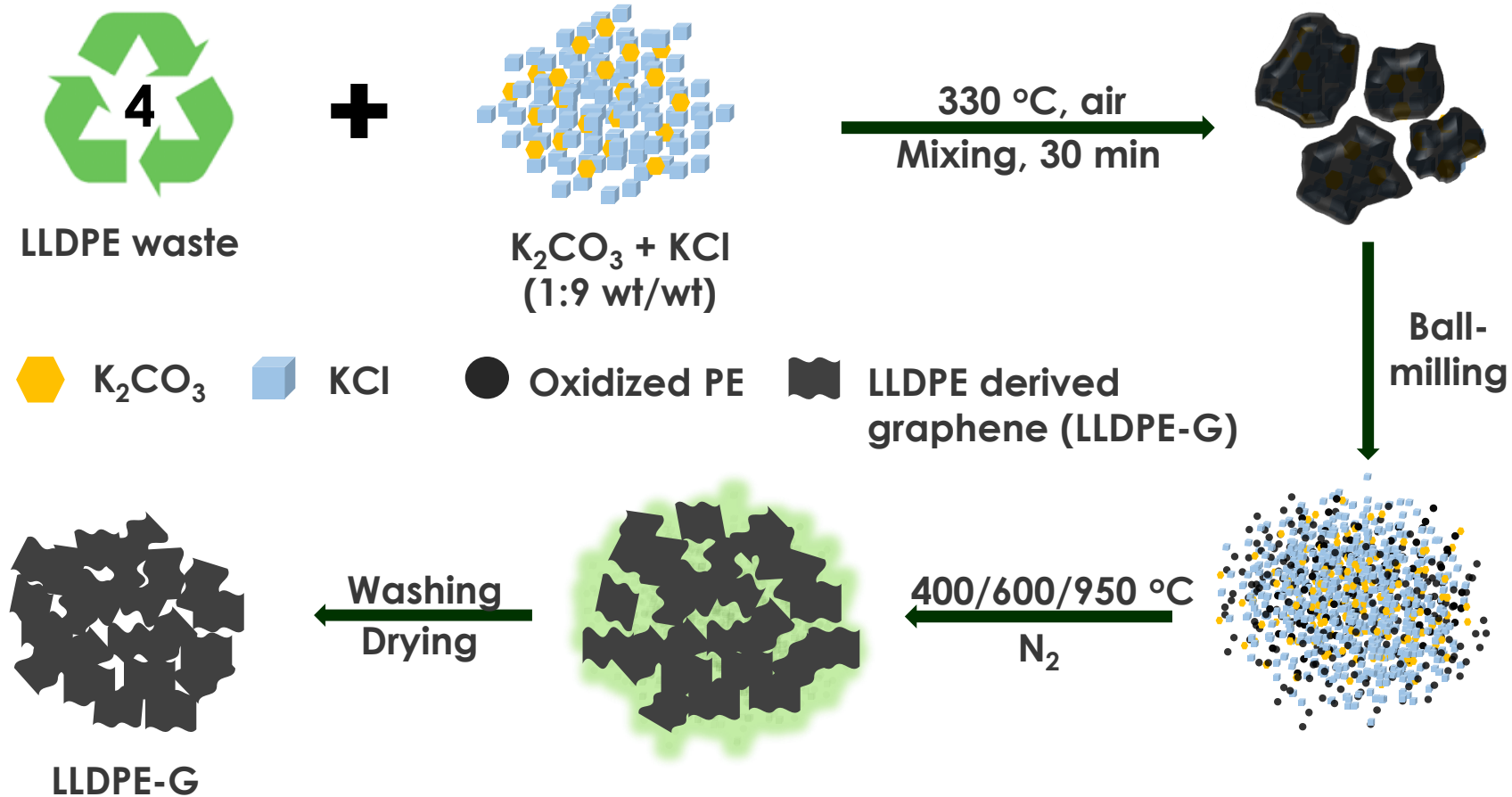
Upcycling PE Waste into Turbostratic Graphene

Thermal Crosslinking and Carbonization of PE



Schematic illustration of enhancing thermal oxidation of PE using solid additive.

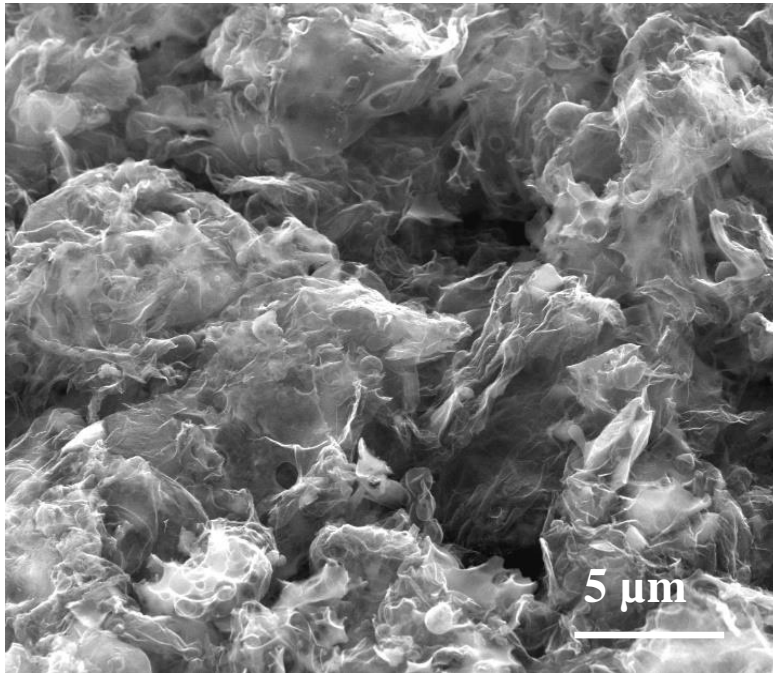
Upcycling PE Waste into Turbostratic Graphene



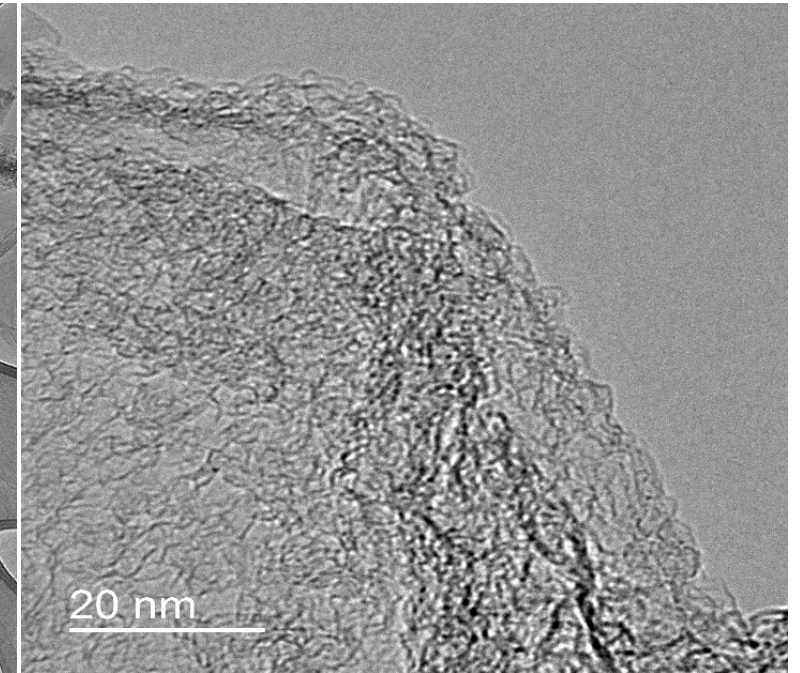
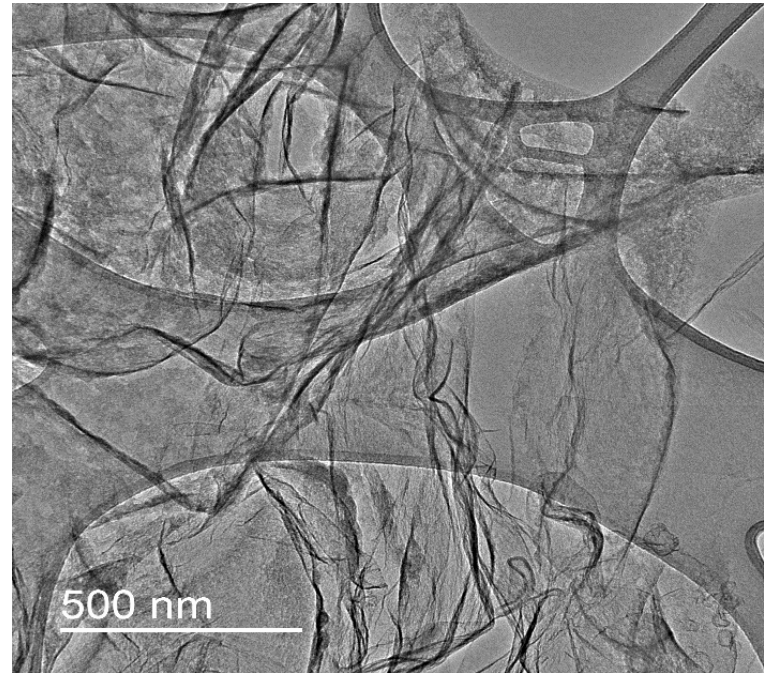
Schematic illustration of synthesis procedure of upcycling LLDPE waste into graphene.

Upcycling PE Waste into Turbostratic Graphene

Morphologies of LLDPE-Derived Graphene



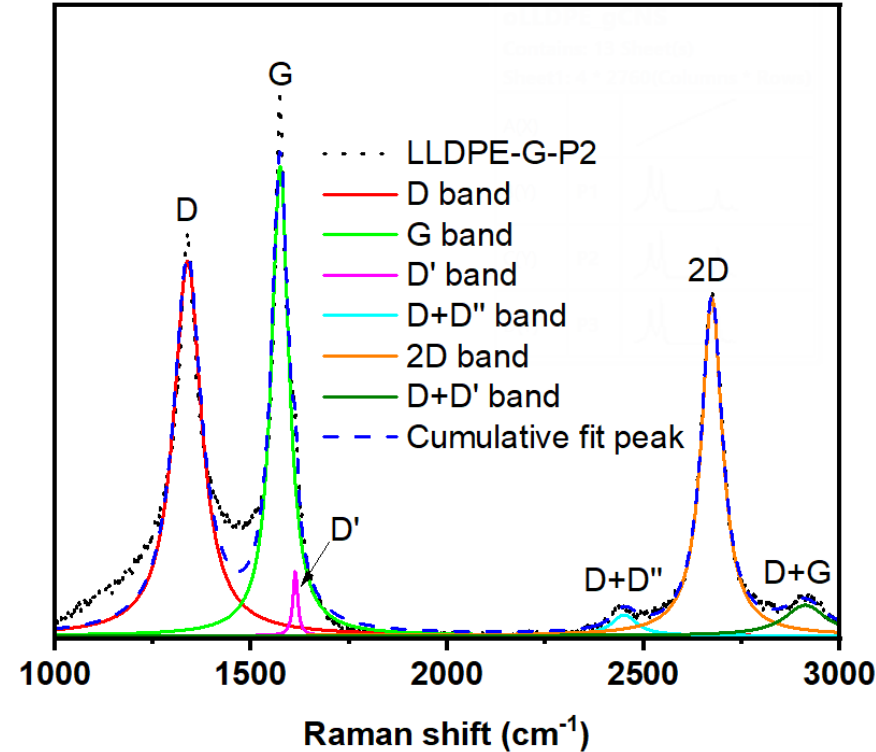
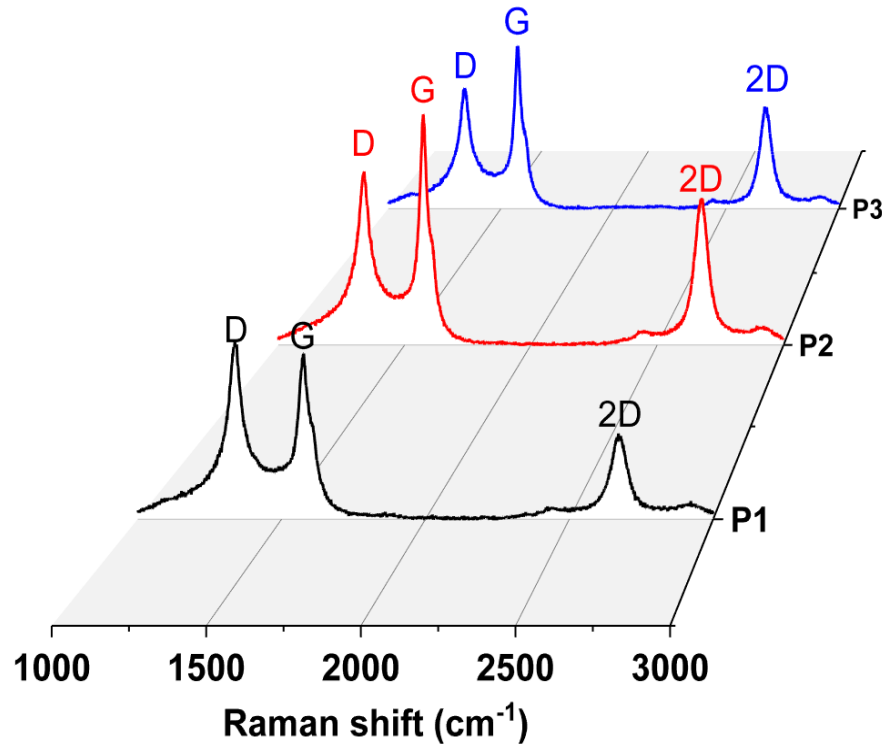
SEM image.



TEM images.

Upcycling PE Waste into Turbostratic Graphene

Physical Properties of LLDPE-Derived Graphene



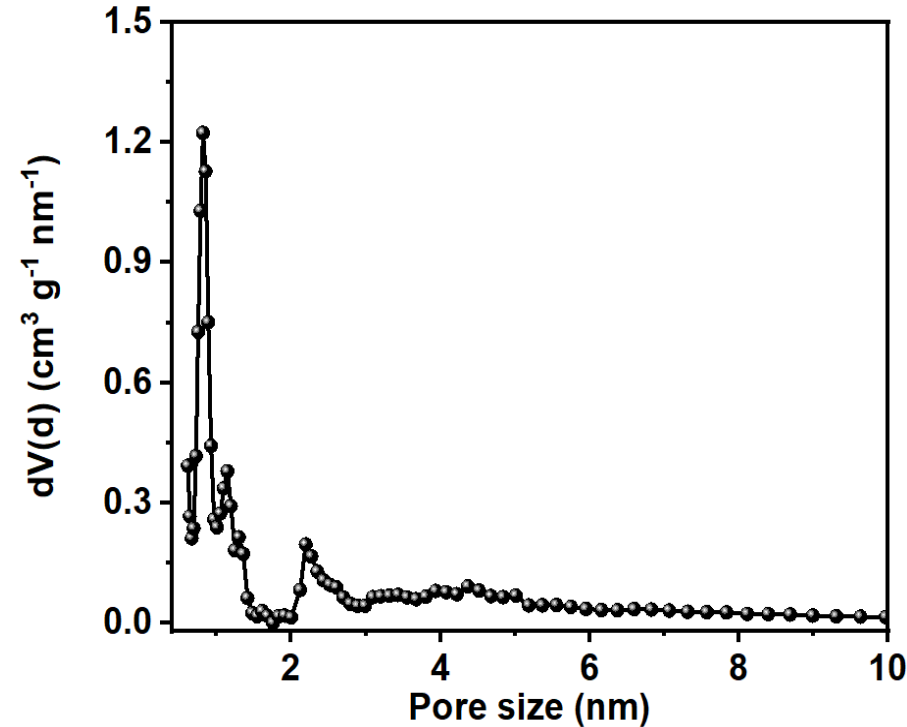
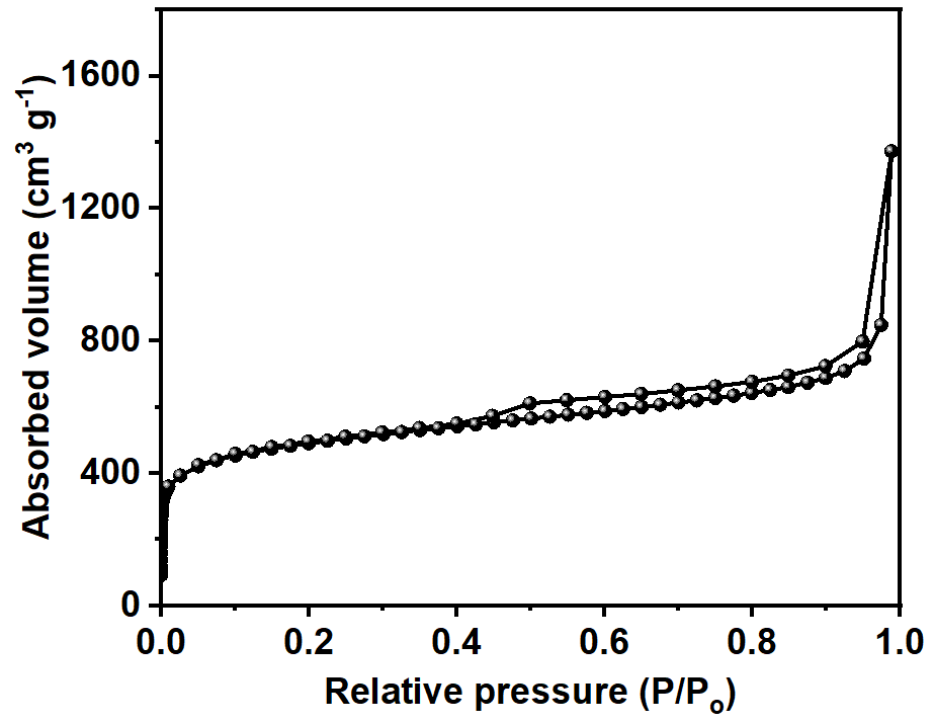
Characteristics of turbostratic graphene:

- High-intensity and narrow D, G, and 2D bands
- Symmetric 2D peak centered at 2,700 cm^{-1} with 532 nm excitation
- A good Lorentzian fit

Raman spectra and Lorentzian fitting (P2) of LLDPE-G.

Upcycling PE Waste into Turbostratic Graphene

Physical Properties of LLDPE-Derived Graphene

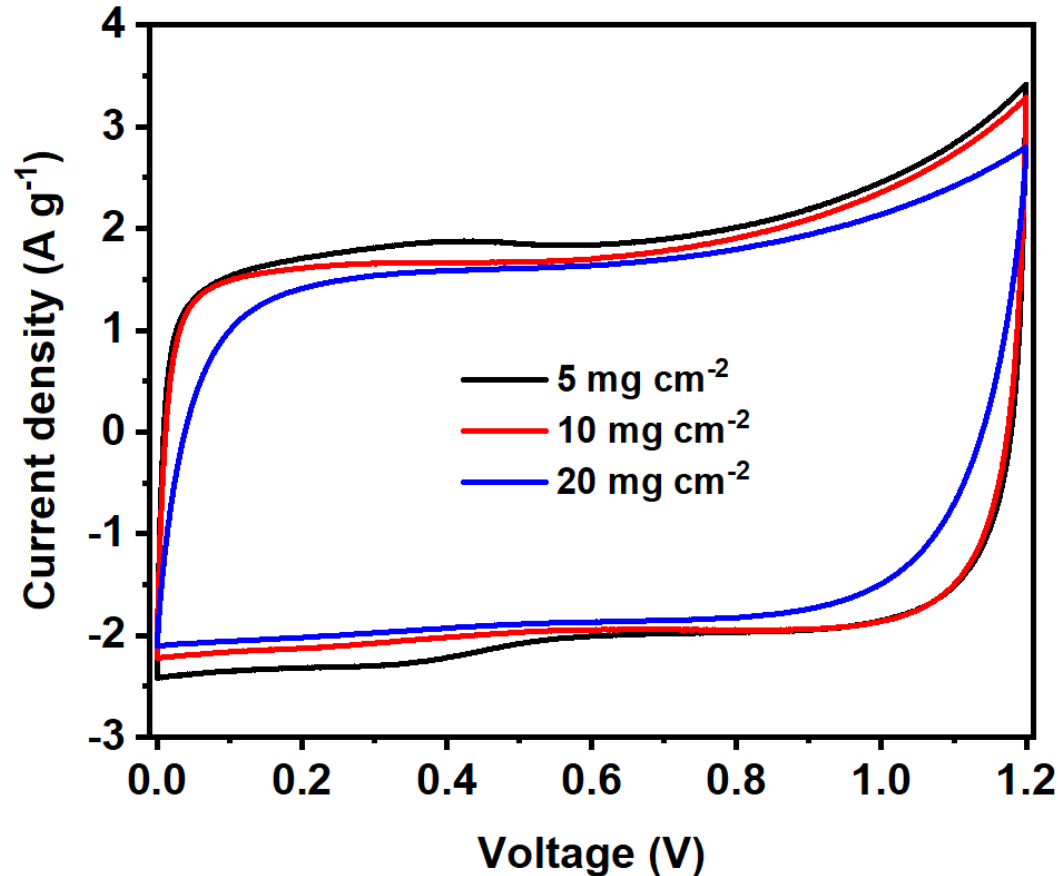


N₂ adsorption – desorption isotherm and QSDFT pore size distribution of LLDPE-G.

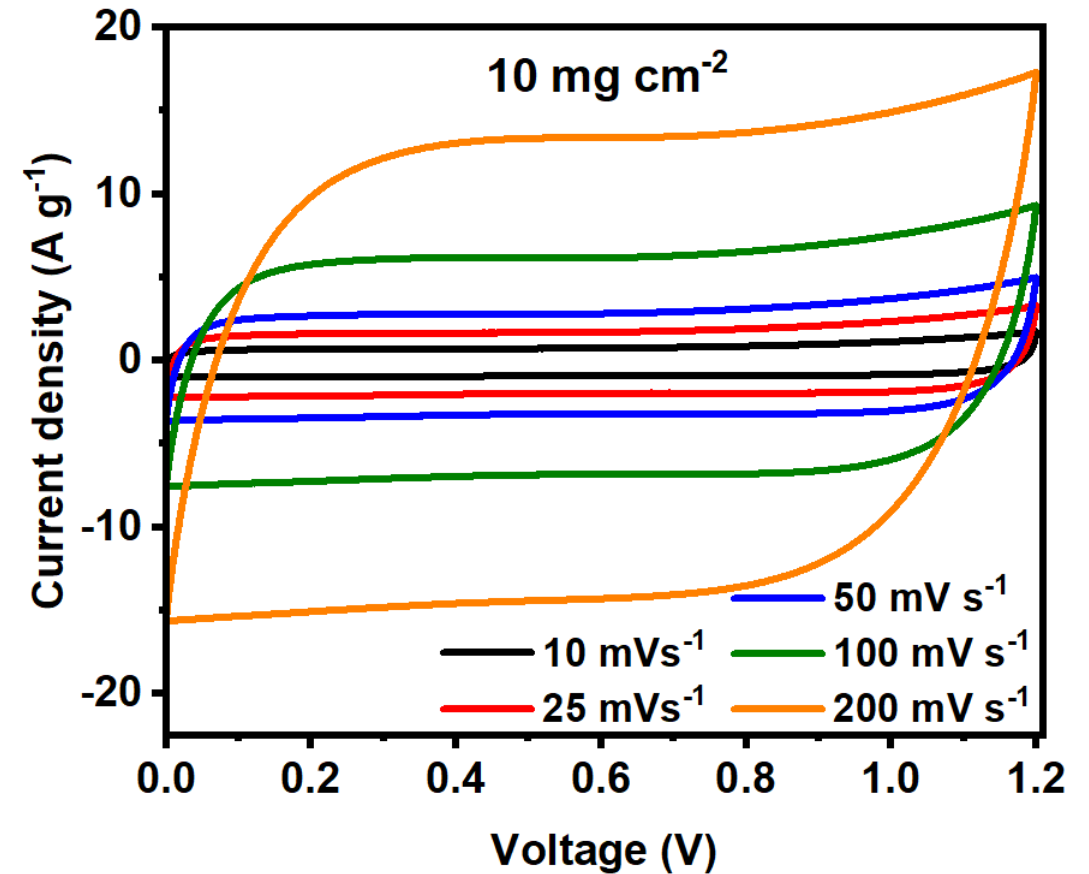
- Specific surface area: 1,800 m²/g
- Total pore volume: 1.16 cm³/g
- Pore structure: Hierarchical pore size distribution

High Mass Loading Supercapacitor Application

Electrochemical Capacitive Properties



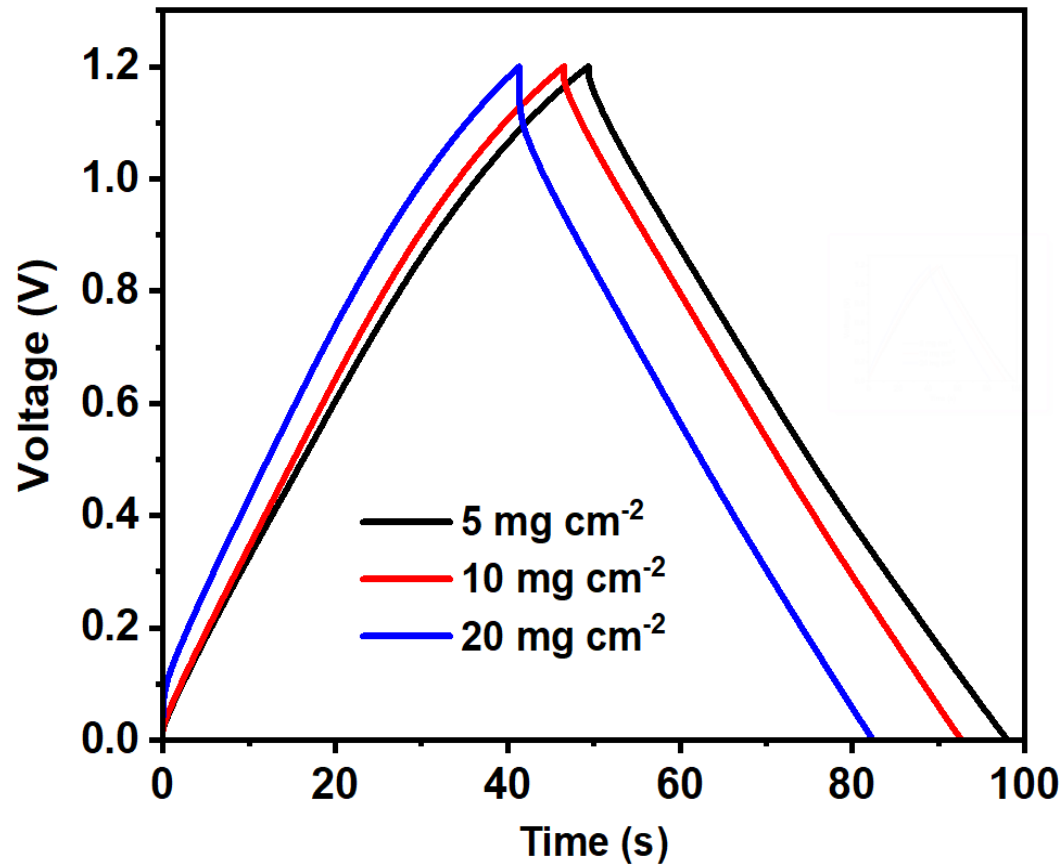
Cyclic voltammetry at scan rate of 25 mV/s .



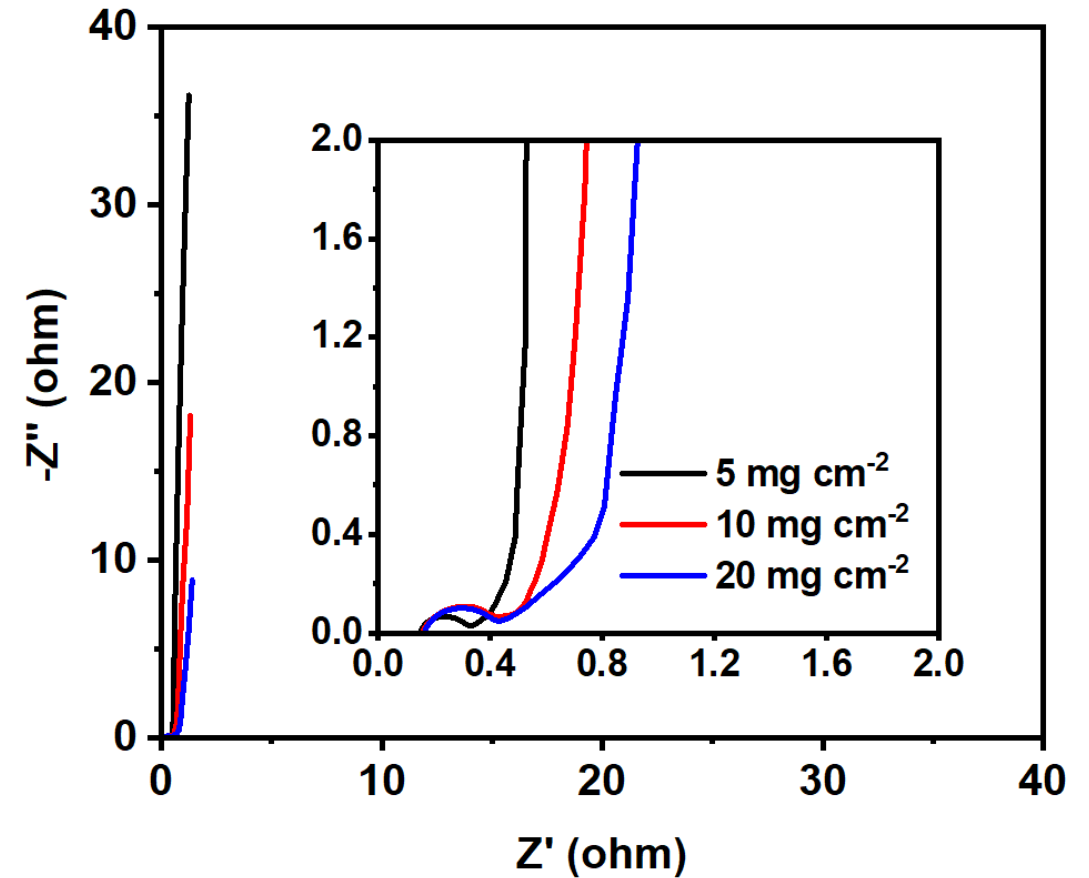
Cyclic voltammetry at various scan rate.

High Mass Loading Supercapacitor Application

Electrochemical Capacitive Properties



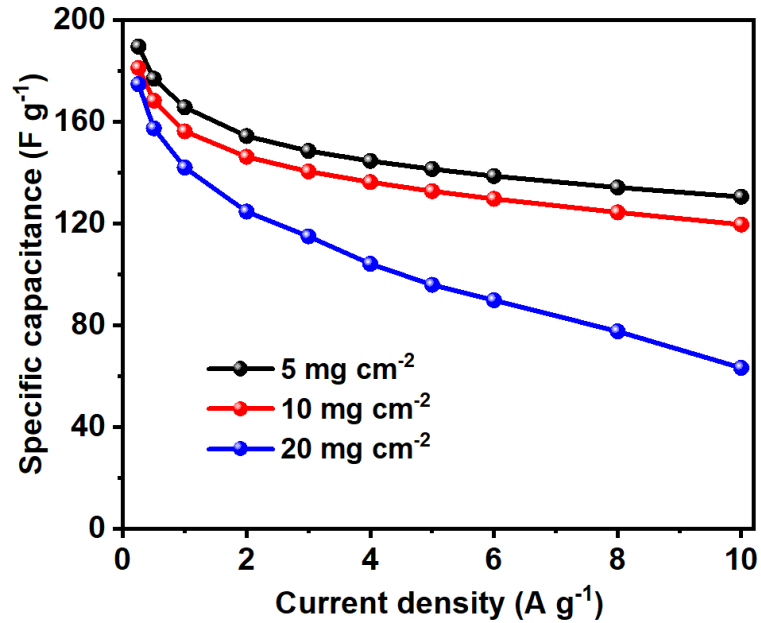
Charge-discharge curves at current density of 1.0 A/g.



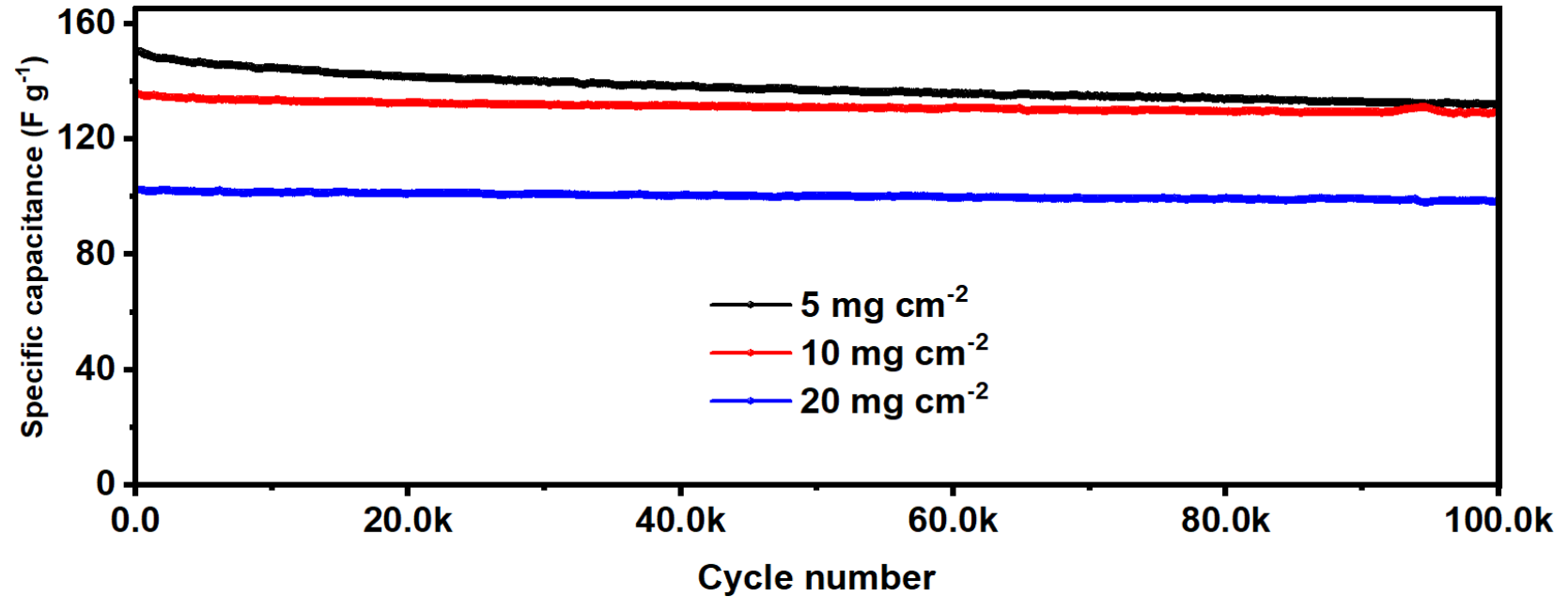
Nyquist plots.

High Mass Loading Supercapacitor Application

Electrochemical Capacitive Properties



Rate capability.



Cycling stability at current density of 4A/g.

Recycling Solid Additives

- KCl is very stable, not chemically changed during thermal treatment processes
- K_2CO_3 chemically changed during thermal treatment processes

During thermal treatment process:

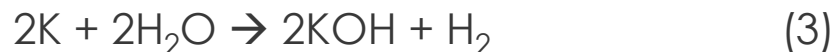
- A small quantity of K_2CO_3 is consumed during the activation redox reaction with carbonaceous feedstock above 600 °C



- Partial decomposition of K_2CO_3 also occurs above 800 °C



During washing process:



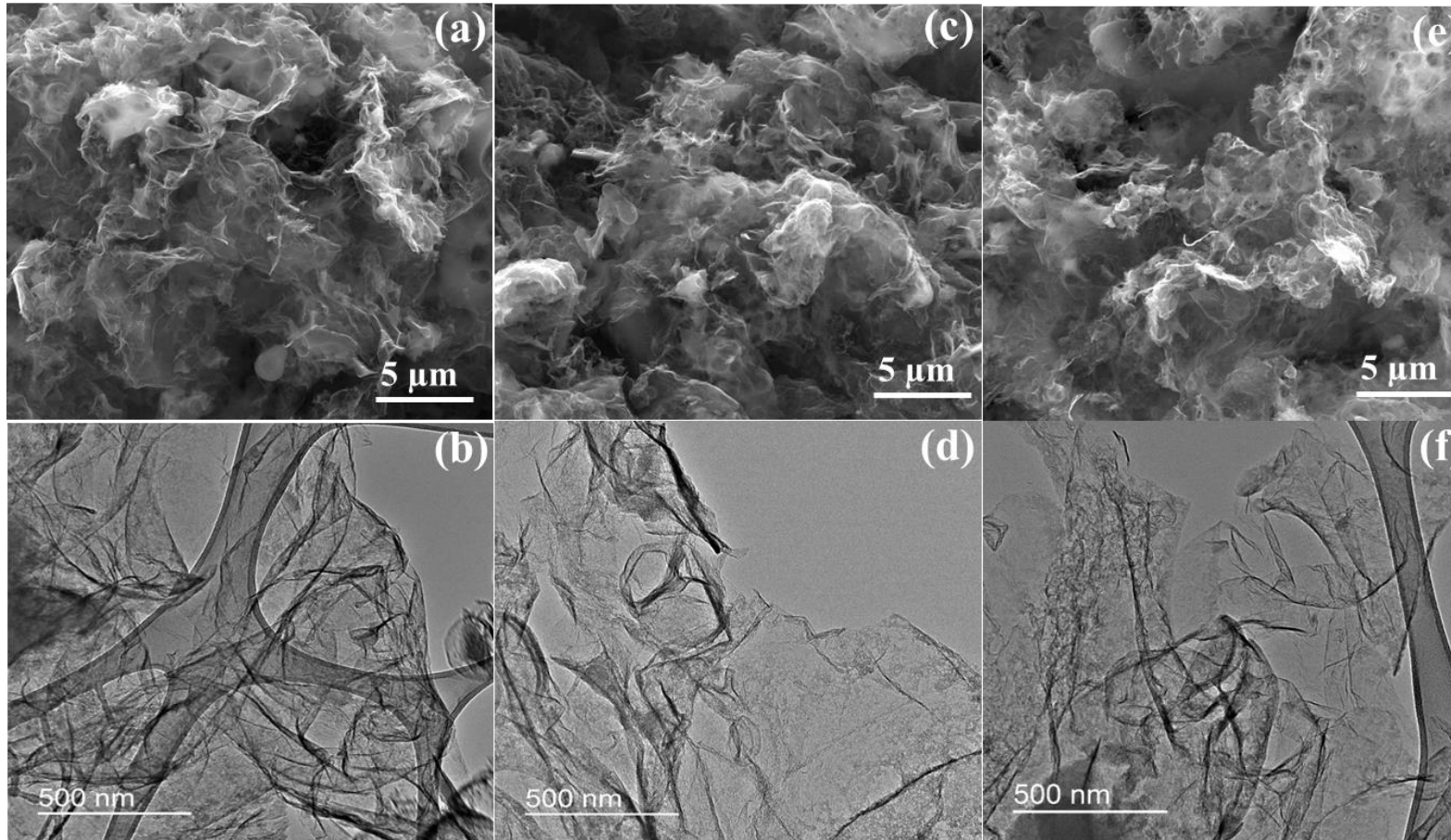
During recycling process: neutralize rinse solution with $KHCO_3$ until pH = 8



It is possible to recycle KCl and K_2CO_3 up to 95%.

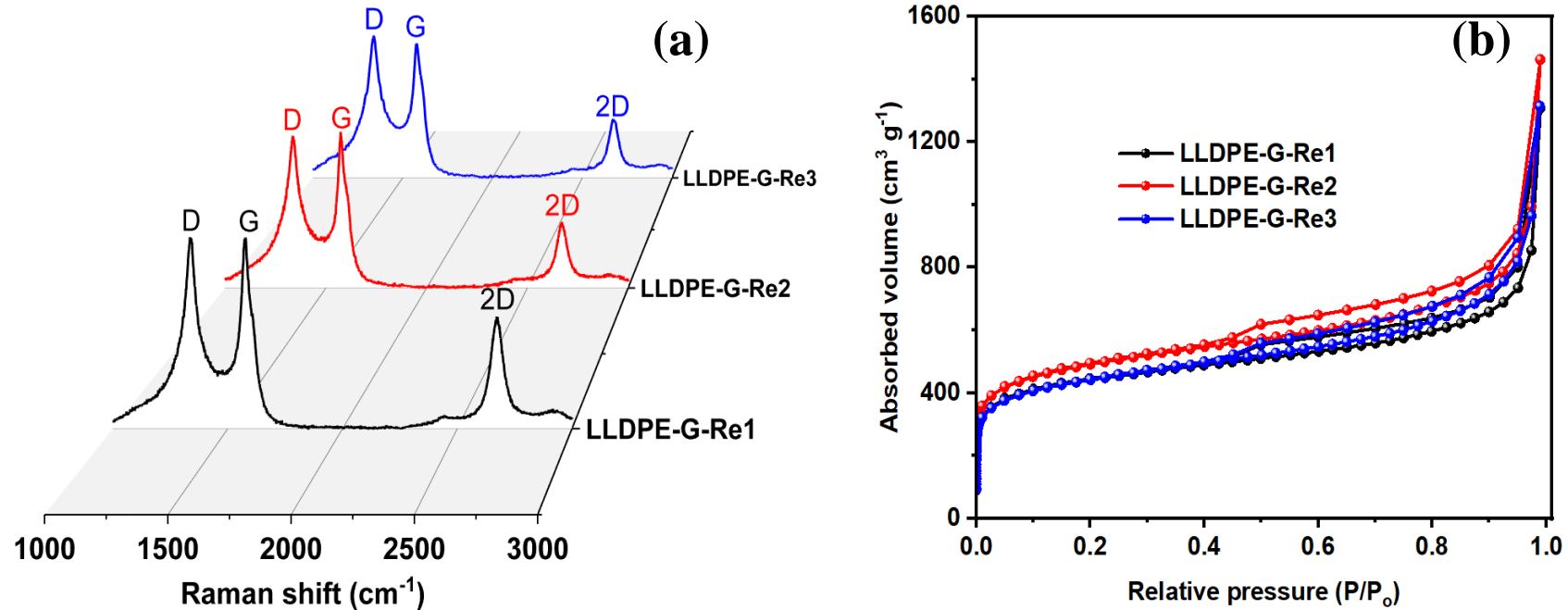
Recycling Solid Additives

Morphologies of LLDPE-G Using Recycled KCl and K_2CO_3



SEM and TEM images of (a & b) LLDPE-G-Re1, (c & d) LLDPE-Re-2, and (e & f) LLDPE-G-Re3.

Physical Properties LLDPE-G Using Recycled KCl and K₂CO₃

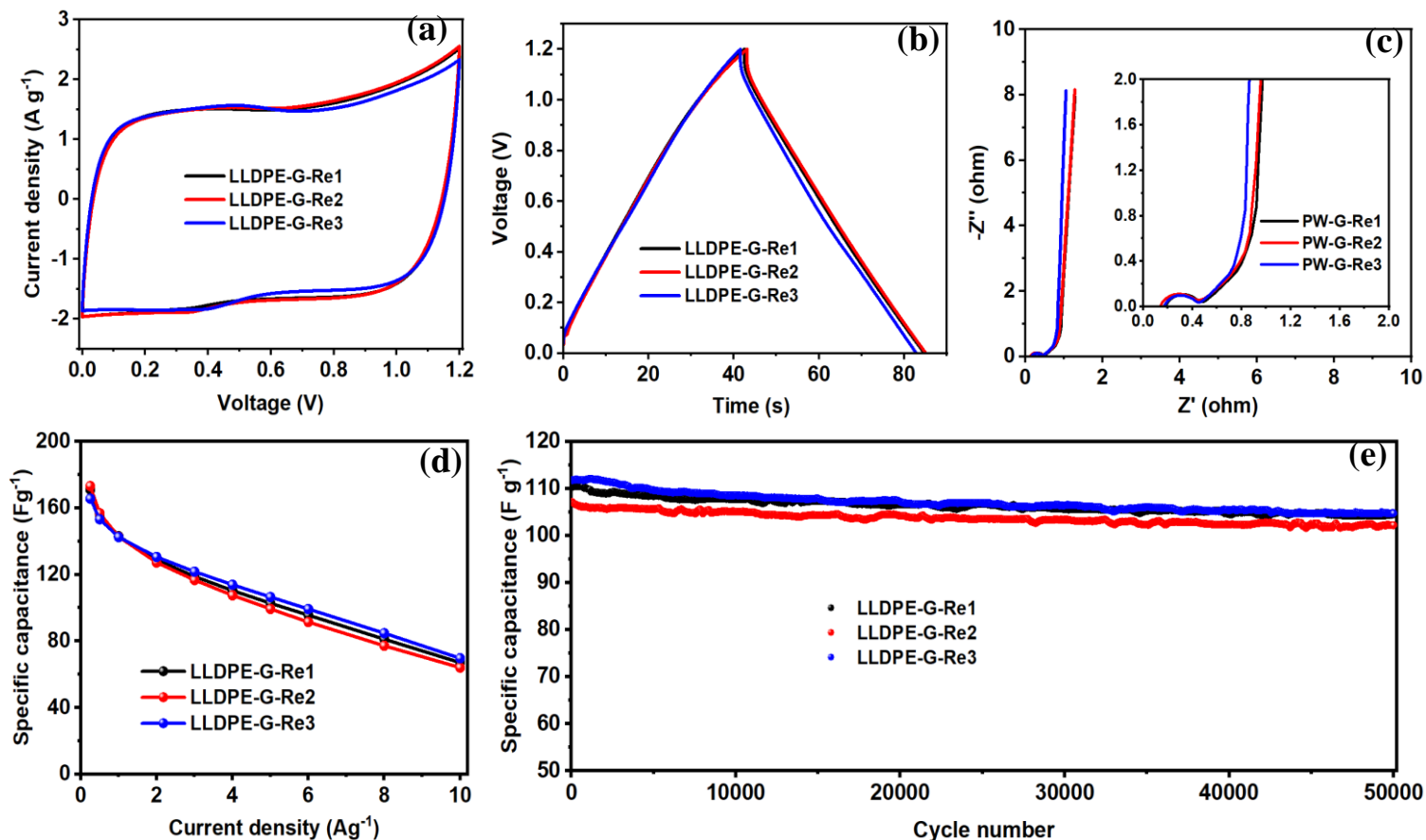


(a) Raman spectra and (b) N₂ adsorption-desorption isotherms of LLDPE-G-Rex.

- Raman spectra confirmed turbostratic graphene structure
- Specific surface area: 1,620– 1,790 m²/g
- Total pore volume: 1.15-1.30 cm³/g

Recycling Solid Additives

Electrochemical Capacitive Properties LLDPE-G Using Recycled KCl and K₂CO₃



Electrochemical capacitive properties of LLDPE-G-Rex supercapacitors with areal mass loading of 20.0 mg/cm².

- This study demonstrated a facile and scalable approach to upcycle LLDPE plastic waste into turbostratic graphene by pre-treatment thermal oxidation of LLDPE with air enabled by a mixture of chemically inert and catalytically active solid surface enhancer additives, followed by carbonization and catalytic graphenization.
- LLDPE-G possesses excellent properties for supercapacitor application with BET specific surface area up to 1,800 m²/g and total pore volume of 1.16 cm³/g with hierarchical micro-, meso-, and macro-porous structure.
- LLDPE-G demonstrates excellent electrochemical capacitive properties as a high mass loading (20 mg/cm²) supercapacitor electrode material with gravimetric specific capacitance of 175 F/g, areal specific capacitance of 3.5 F/cm², energy density of 9.45 Wh/kg, and cycling stability of 95.8 % capacitance retention after 100,000 cycles.
- Recycling KCl and K₂CO₃ for at least three times led to potentially low-cost and sustainable production of turbostratic graphene from plastic waste.

(Ref: V. H. Pham et al., Chemical Engineering Journal, 2024, 498, 155873)

Acknowledgments



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