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

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***MS&T24***

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# Authors and Contact Information


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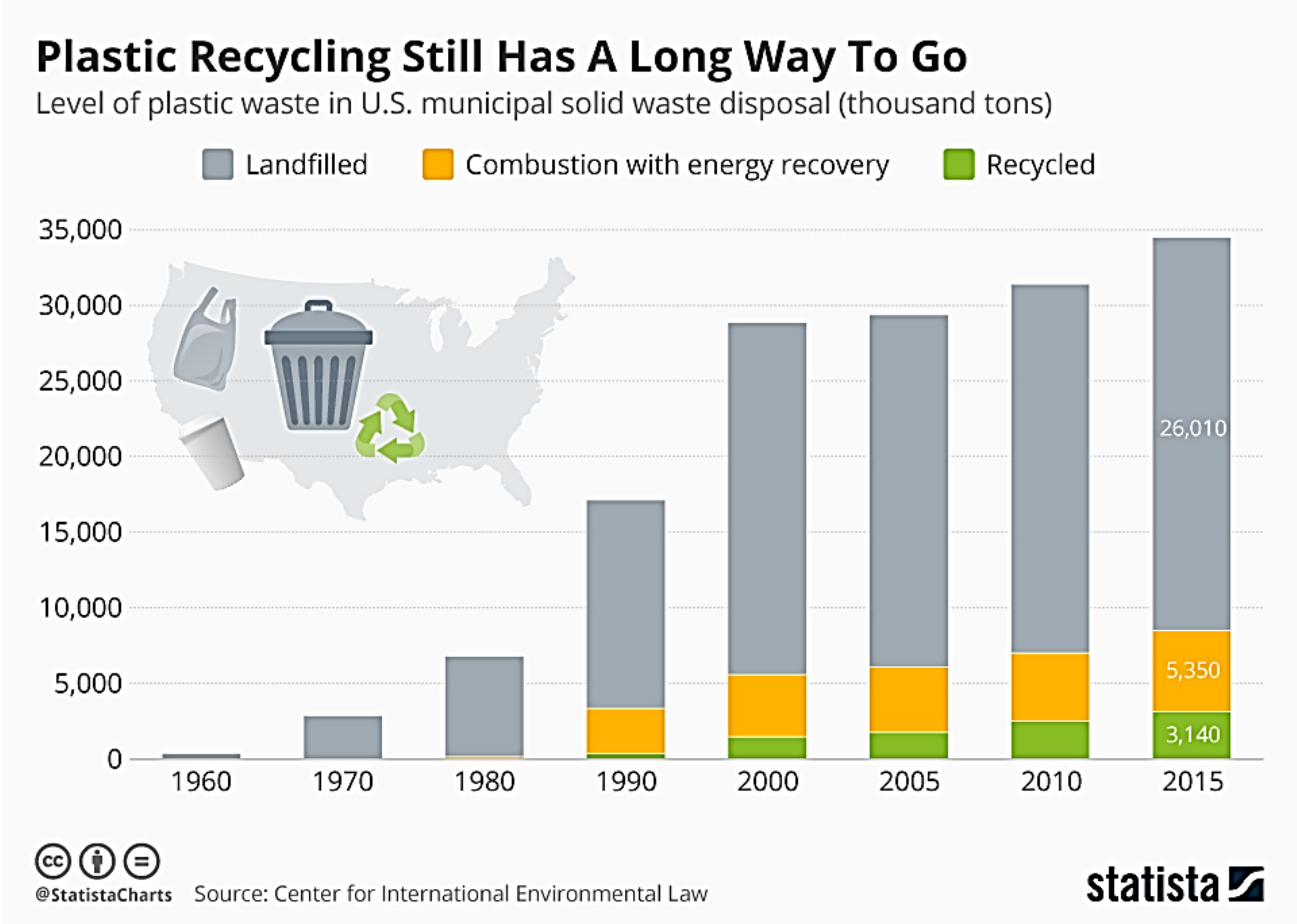
# Background-Plastic Recycling and Upcycling




- 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.
- Polyethylene (PE) is difficult to recycle/reuse; decomposes during thermal processing.

# Background-Supercapacitor’s Application

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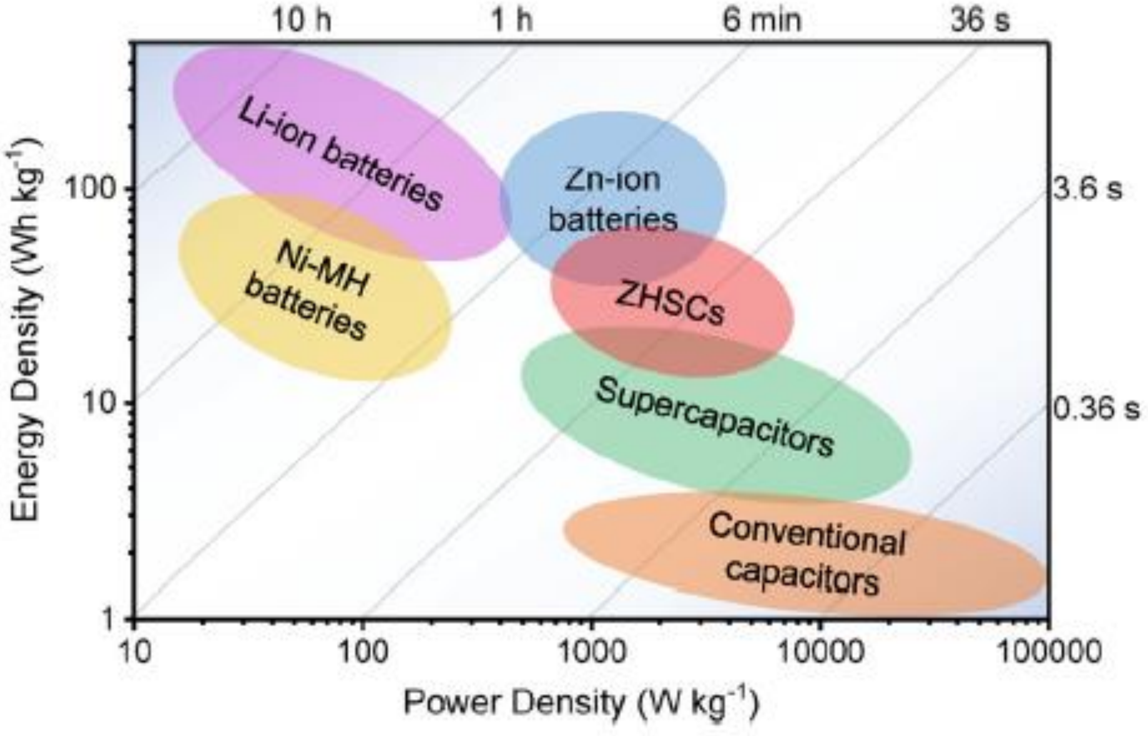
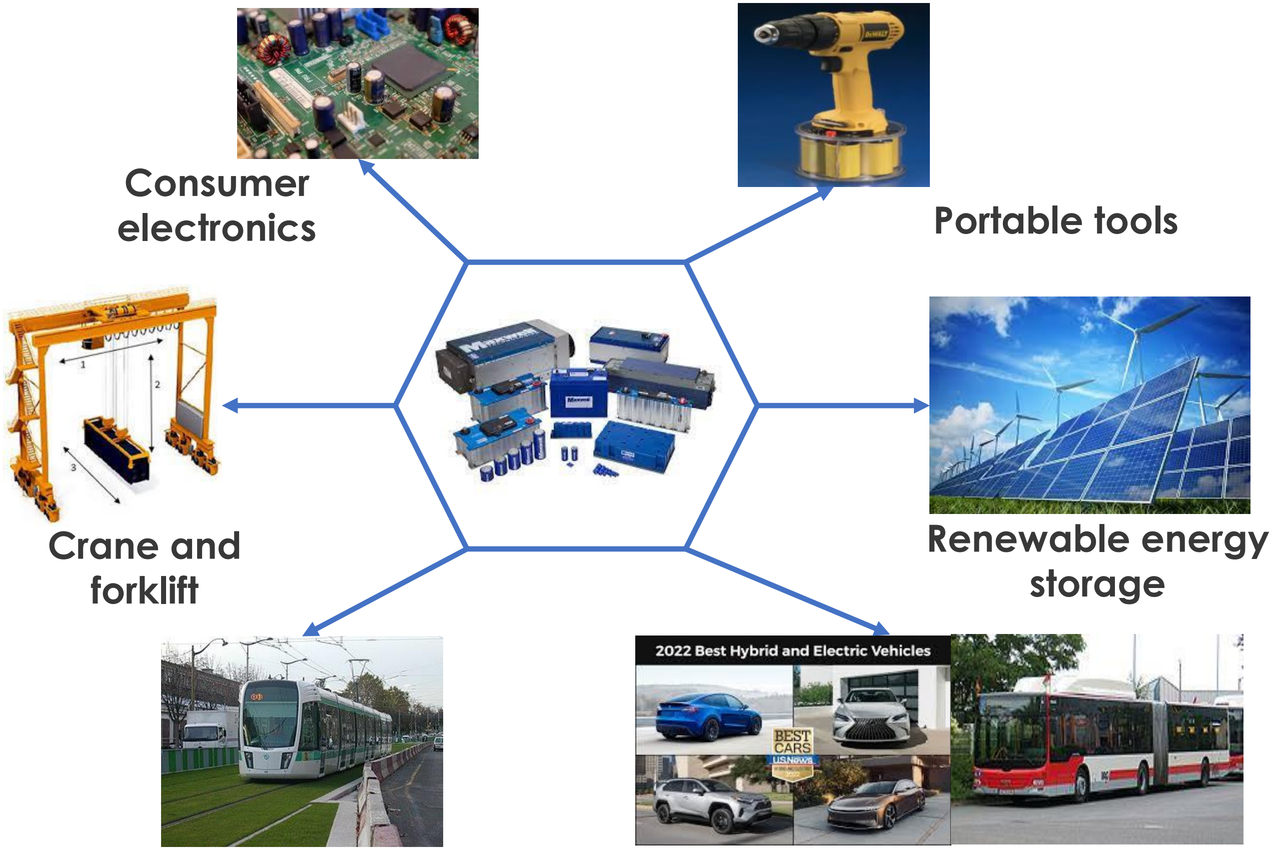
## Supercapacitors vs. other electrochemical storage devices




*(Carbon Energy, 2020, 2, 521–539)*

## Supercapacitor applications



# Background - Previous Work

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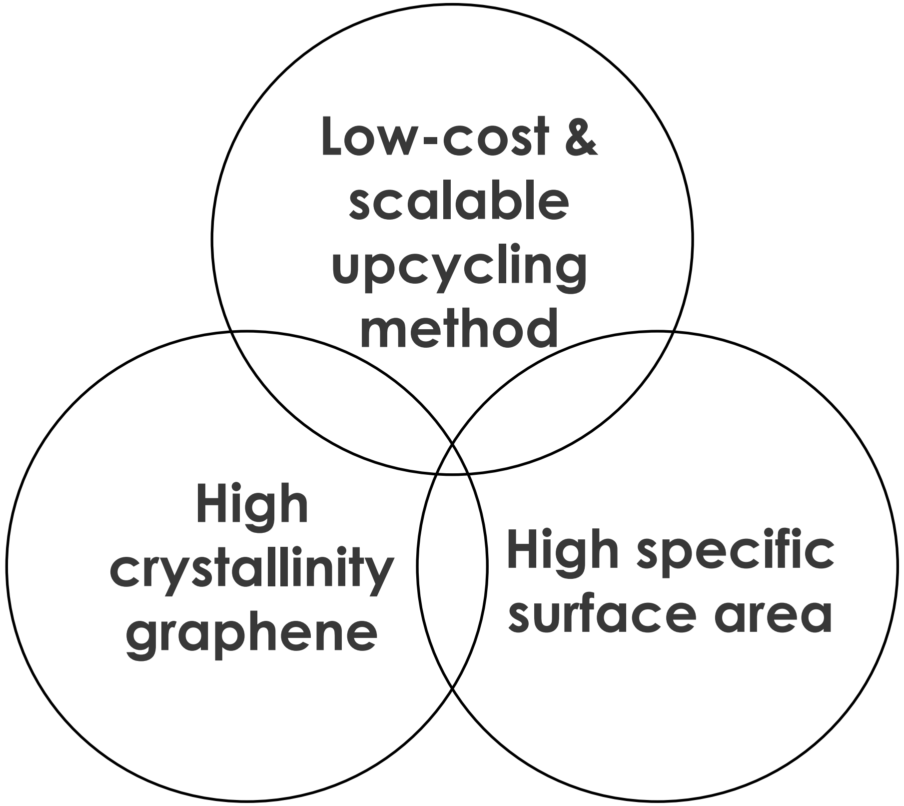
Summary of previous work on plastic upcycling into porous carbonaceous materials

| Plastic Waste | Product and Yield | BET SSA- ($m^2g^{-1}$) |
|---|---|---|
| PE or PP | Amorphous porous carbon | 750-2,200 |
| PVC, PEs, PP, PS, PET | Flash graphene | Not reported |
| Mixed plastic wastes | Holey graphene | ~900 |




**Upcycling plastic waste into high quality graphene with high crystallinity and high specific surface area is still very challenging.**

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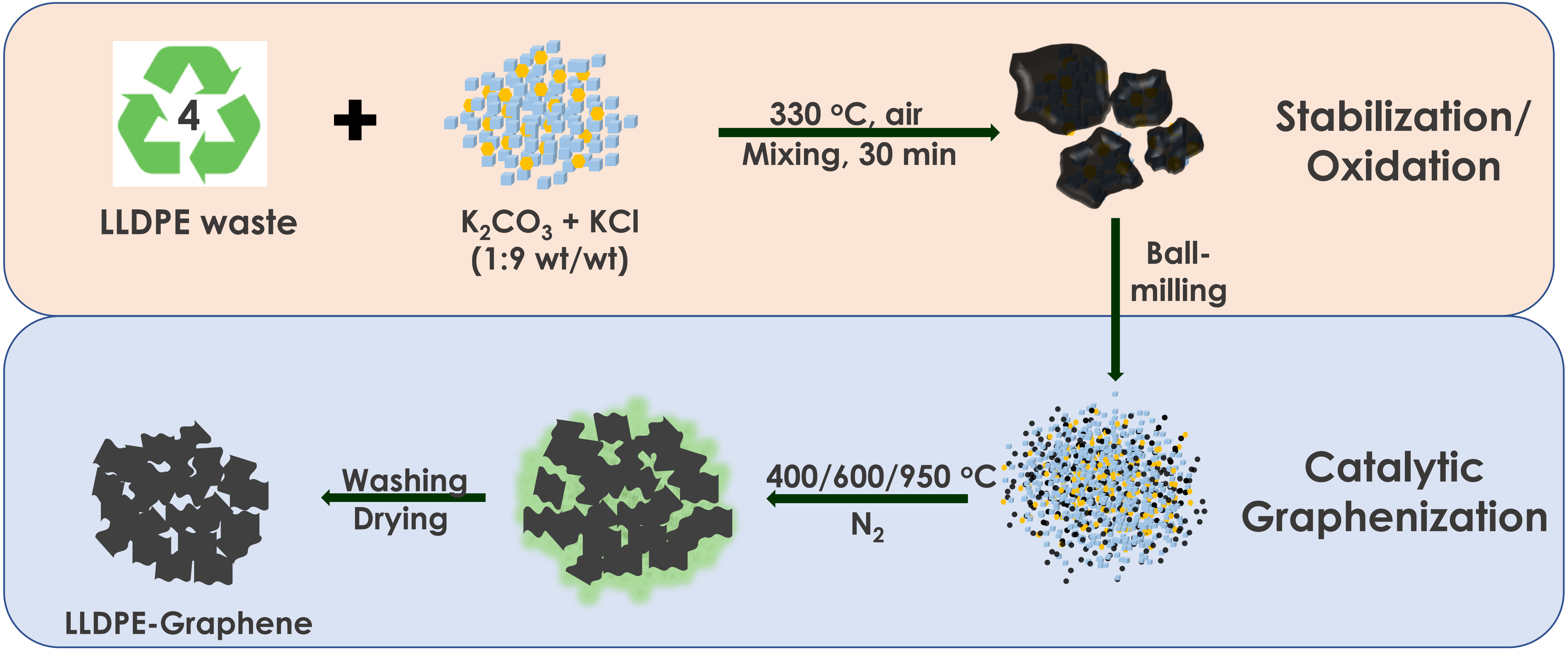
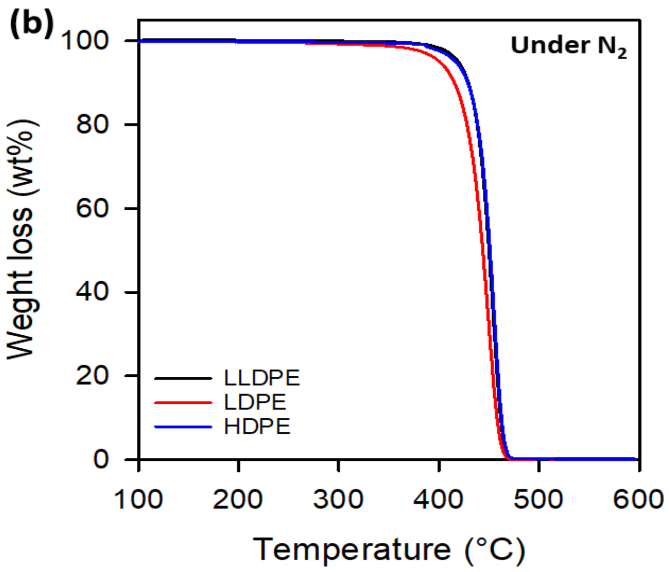
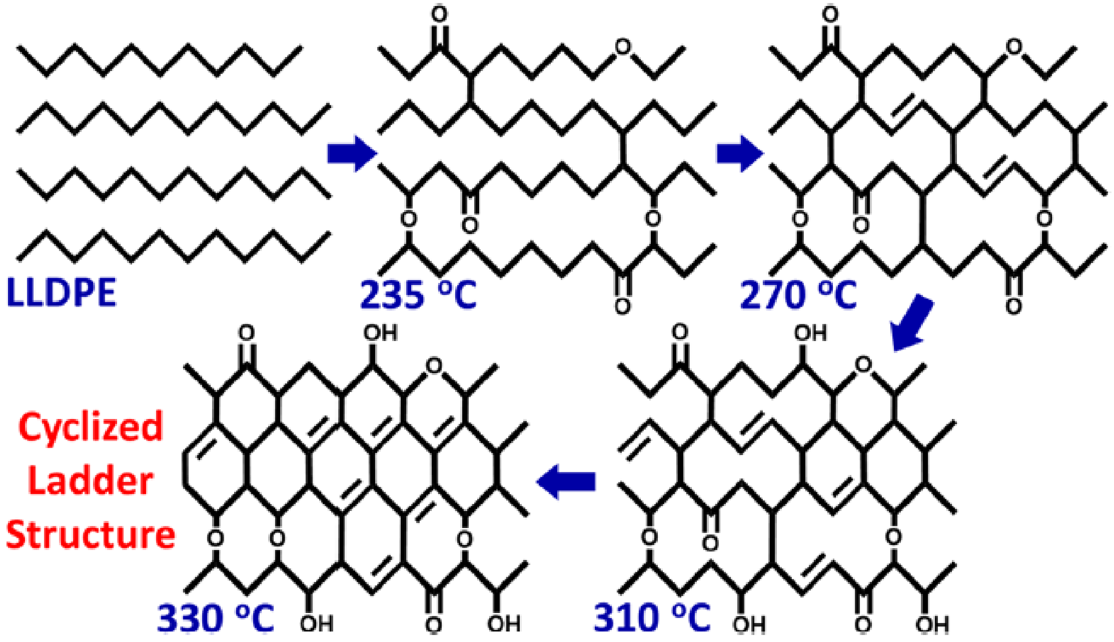
# PE Upcycling into Porous Graphene



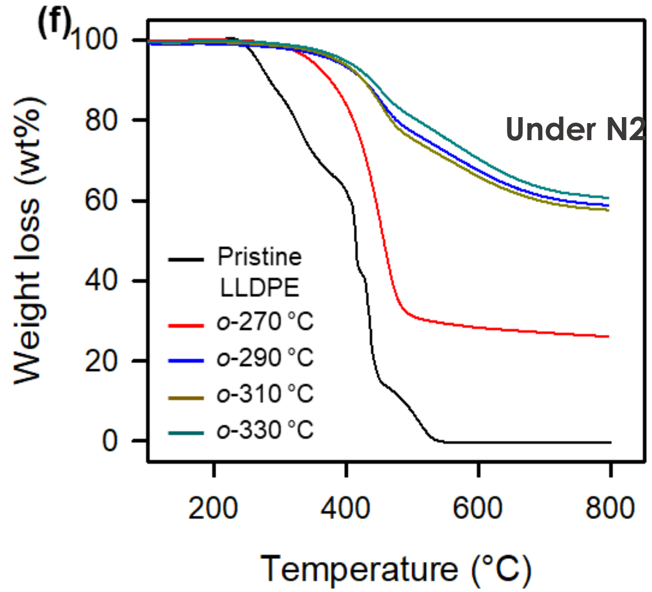

*(Y. Gao et al., Chemical Engineering Journal, accepted)*

# Why Do We Need Stabilization as a Pretreatment?



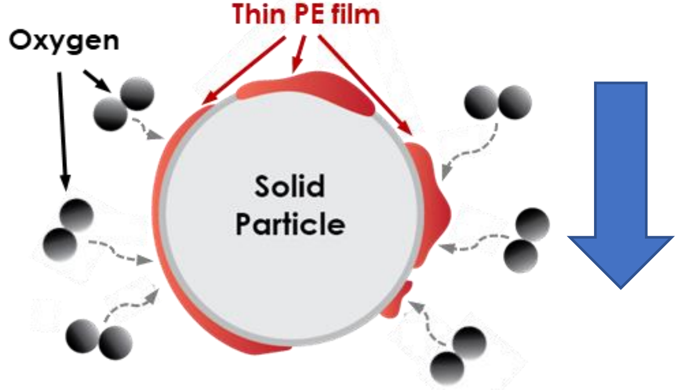

*Mechanism from: Choi et. al., Chem. Mater. 2017, 29, p 9518–9527*




*(K. Kim. et al,. ACS Sustainable Chemistry & Engineering, in review)*

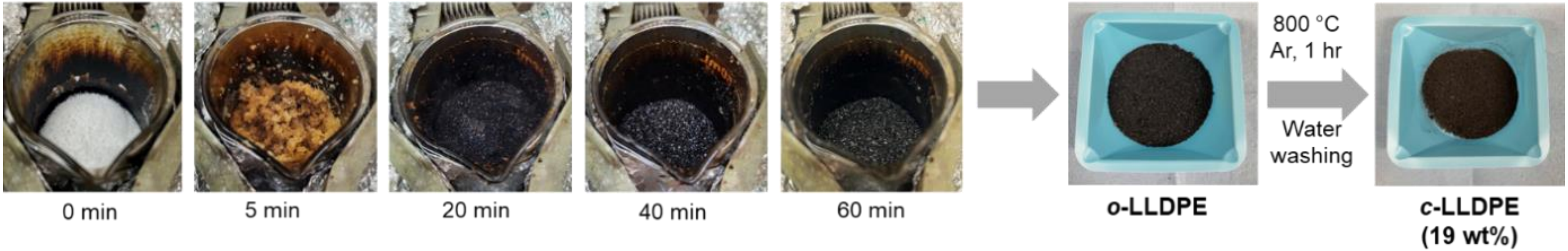
- Stabilization occurs from oxidation and formation of cyclized ladder structures which are more stable at high temperatures than original alkyl chain.
- Pre-treating between 290-330 °C stabilizes PE for high temperature processing under $N_2$.

# How Do We Do Stabilization in Bulk?

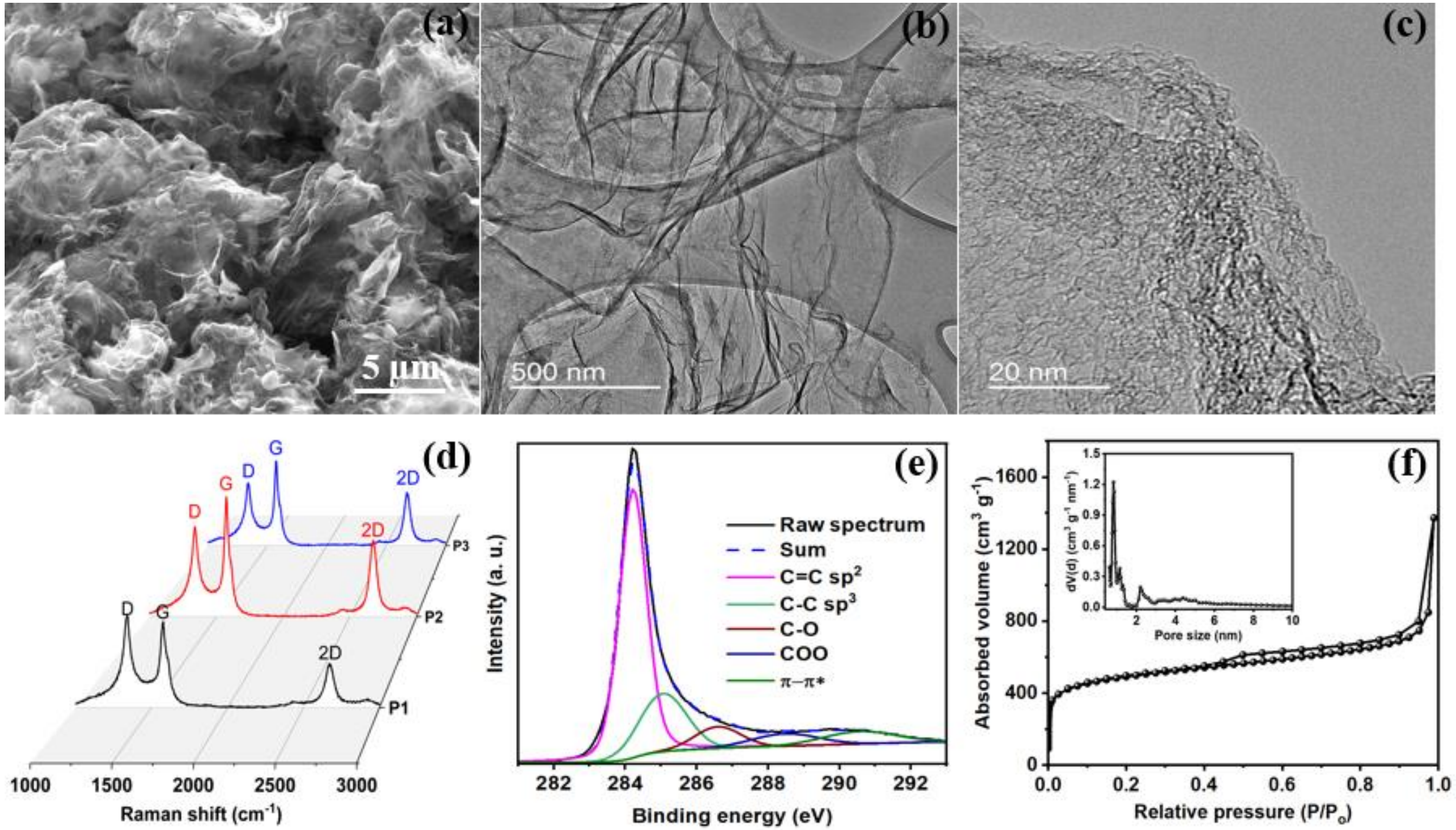




Adding Solid Additives

**KCl and $K_2CO_3$ mixture (ratio 9:1) is used as the solid additive for this application.**



# PE-Derived Porous Graphene




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

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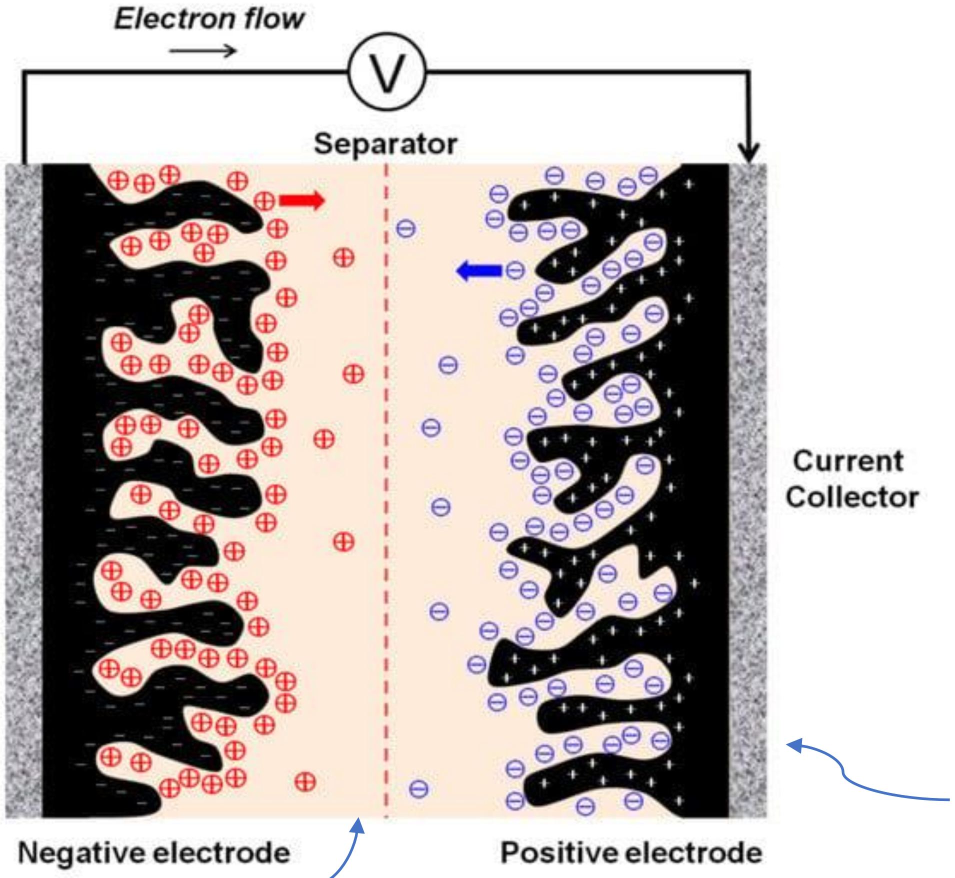
# Supercapacitor Test




PE-derived graphene electrode

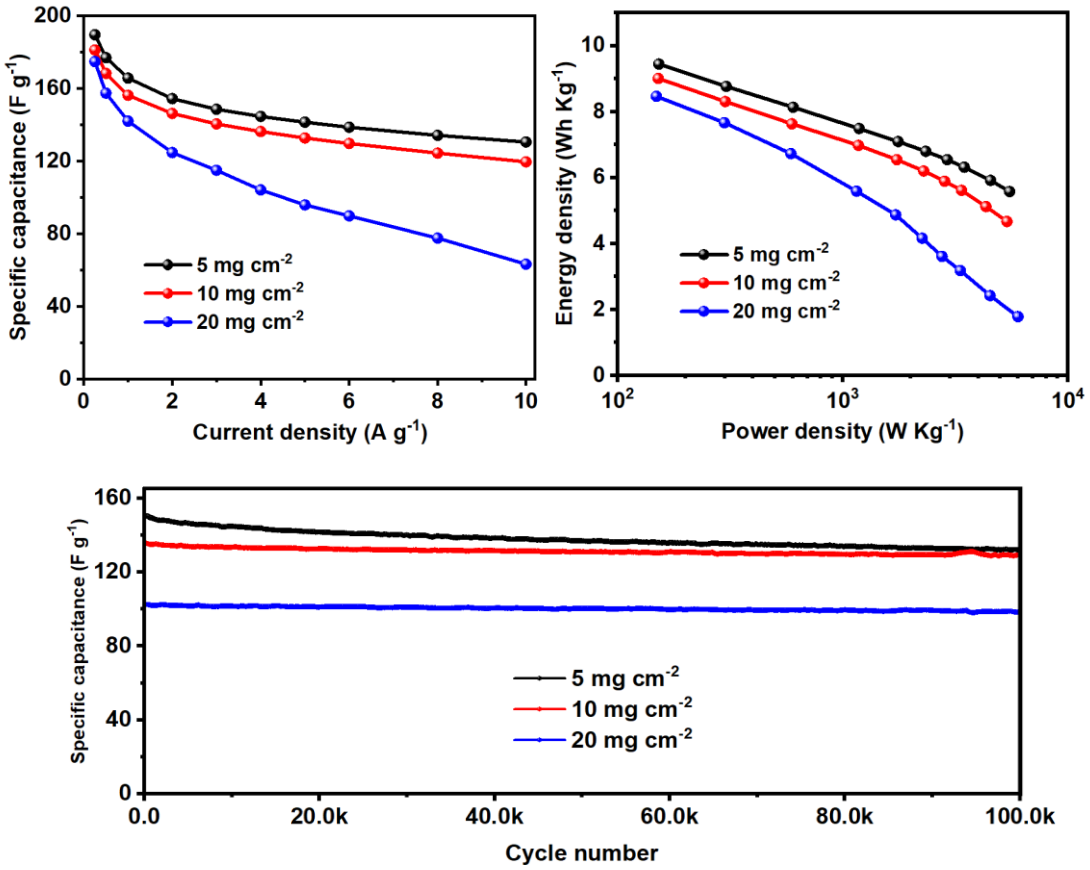
1M $H_2SO_4$ aqueous electrolyte

**High areal capacitance and high power density are desired.**

- Schematic illustration of the porous carbon-based supercapacitor (*Materials 2020, 13(18), 4215*)
- *V. H. Pham et al., Small Methods 2024, 2301426*

# Electrochemical Capacitive Properties






| | 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 (%) |
|---|---|---|---|---|
| PE-Derived Graphene | 1,800 | 175 | 7.86 | 95.8 |
| Commercial SOTAYP-50F | 1,760 | 150 | 6.27 | 88.5 |

**PE-derived graphene has comparable or even better electrochemical capacitive properties.**

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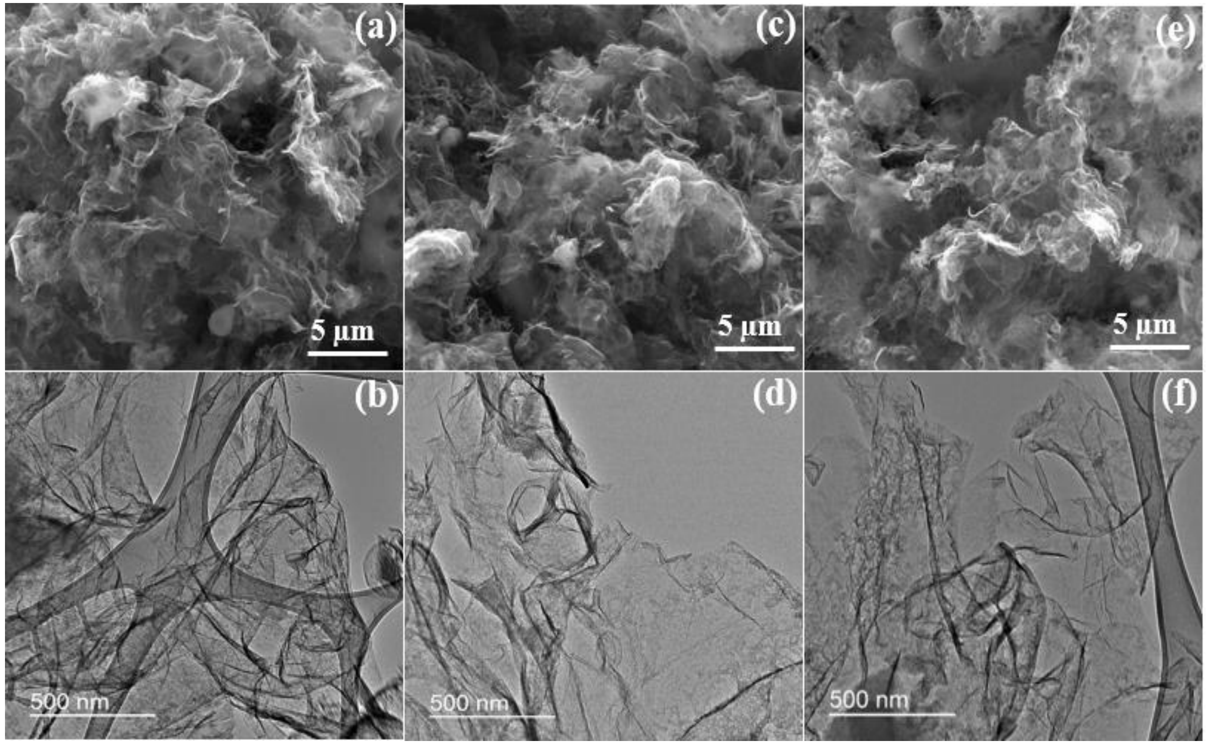
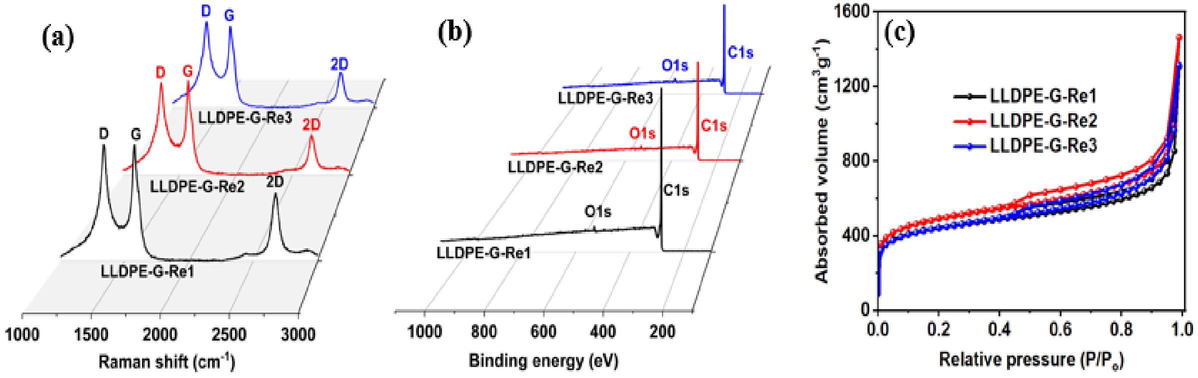
# Salt Recycling/Reuse

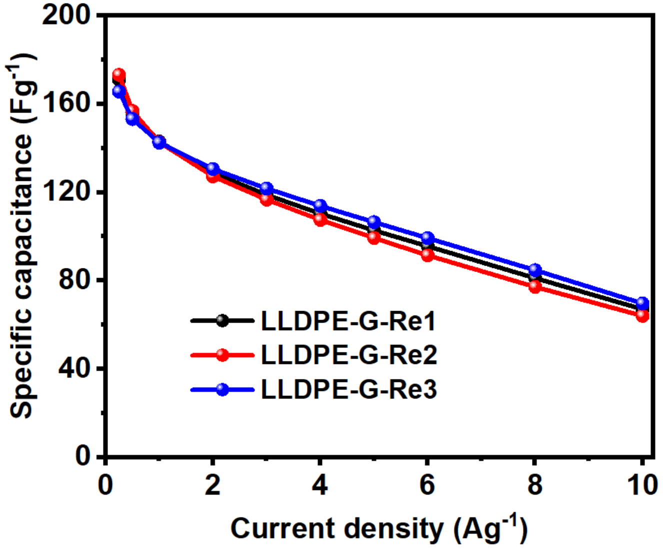
G-re1 G-re2 G-re3

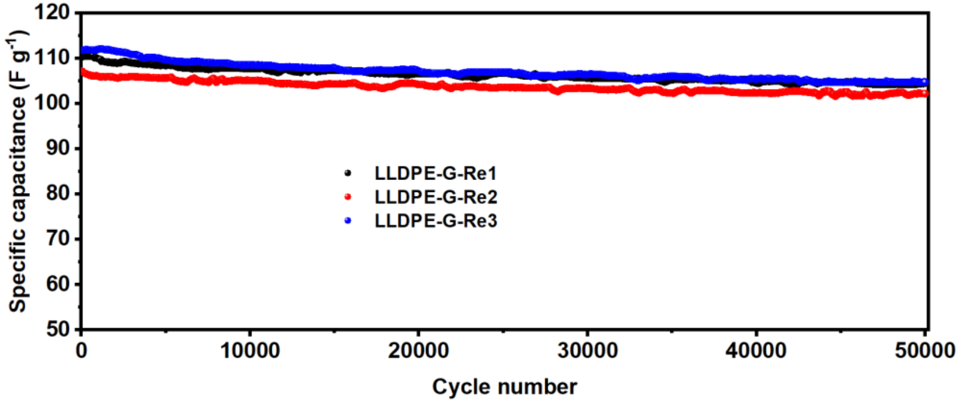



- 95% of salt mixture can be recovered and reused for subsequent synthesis.
- Salt mixtures are reused three times which produce similar quality porous graphene products as G-re1, re2, and re3.

# Salt Recycling/Reuse




| | BET Surface Area ($m^2$ $g^{-1}$) | Specific Capacity at 0.25 A/g (F $g^{-1}$) | Capacity Retention after 100,000 Cycles (%) |
|---|---|---|---|
| G-re0 | 1,800 | 175 | 95.8 |
| G-re1 | 1,622 | 171 | 94.7 |
| G-re2 | 1,792 | 173 | 95.6 |
| G-re3 | 1,615 | 166 | 93.6 |

**Subsequent graphene products have comparable electrochemical capacitive properties to re0, which indicates successful salt recycle and reuse.**

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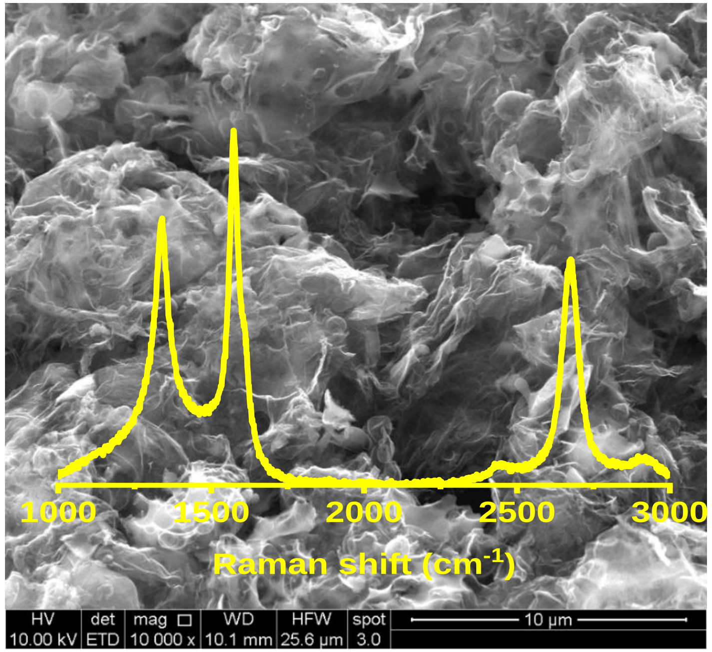
# Conclusion



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



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