Extensional rheology of CNT/redox-active polymer inks for critical element recovery

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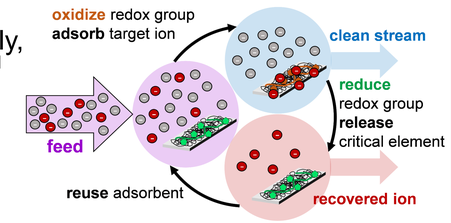
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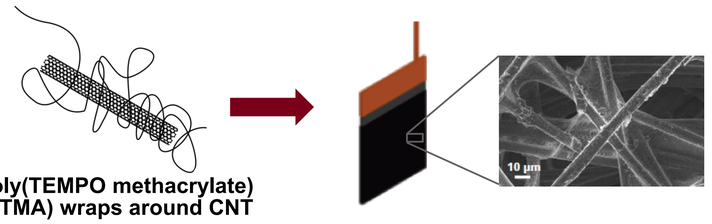
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Motivation

- Traditional critical element recovery methods are costly, inefficient, and irreversible[1]
- PTMA/CNT porous networks enable highly selective recovery via redox-active adsorption

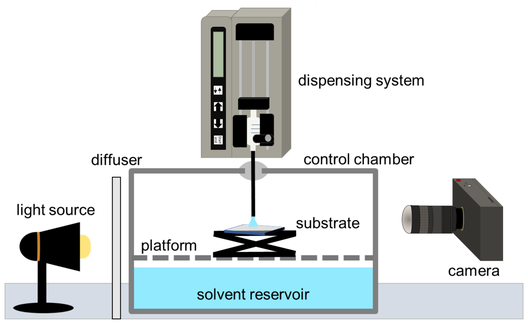


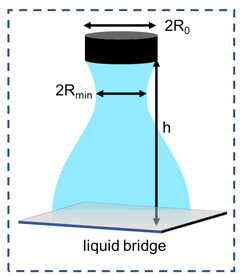
- Conventional PTMA synthesis is bottlenecked at $M_n < 200\ kDa$, leading to polymer leaching and limiting fiber spinnability
- Extensional rheology (more sensitive to chain stretch than shear) → probe dispersion stability, deformations, suitability for scalable coating (inkjet printing, spray coating, roll-to-roll)[2]



EC DoS instrument design

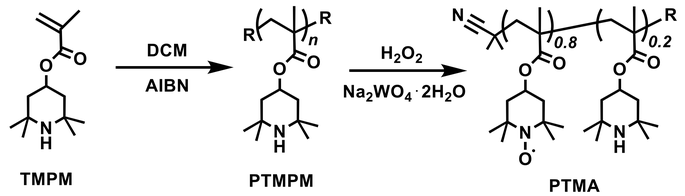
- Dripping-onto-substrate → minimal volume, pre-deformation
- DoS environmental control chamber with sacrificial solvent reservoir → minimal evaporation[3]





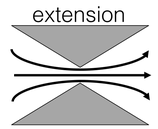
UHMW PTMA synthesis

- **First synthesis** of UHMW PTMA ($M_n > 2.5\ MDa$) by optimizing free radical polymerization



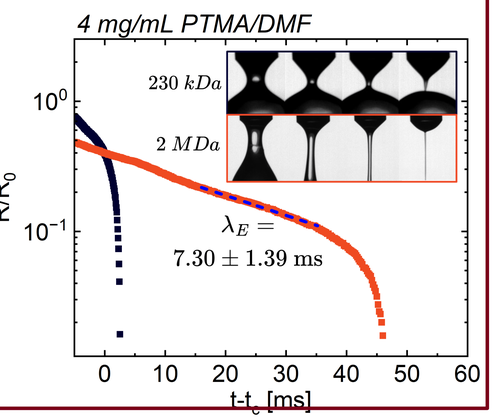
MW effect on ext. flow behavior

Inertio-capillary (IC) thinning	Elasto-capillary (EC) thinning
• Low MW PTMA	• UHMW PTMA
$\frac{R(t)}{R_0} = \alpha \cdot (t_b - t)^{\frac{2}{3}}$	$\frac{R(t)}{R_0} = A \cdot \exp(-\frac{t-t_c}{3\lambda_E})$
• Cone-shaped liquid bridge	• Cylindrical filament
• Rapid breakup time t_b	• Slow exponential decay



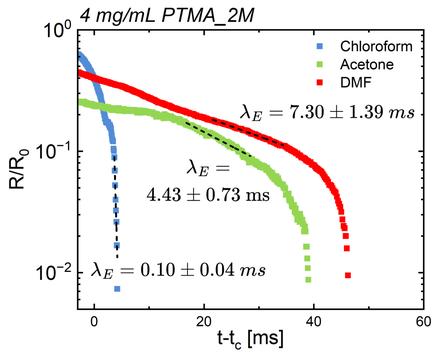


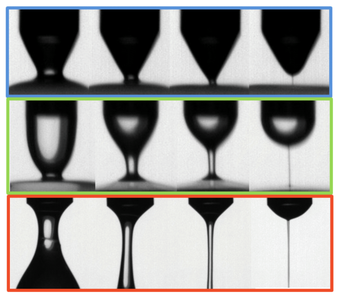
- M_w ↑ leads to: **elasticity** ↑, λ_E ↑, t_b ↑
- Low elasticity: prompt breakup for spraying, printing, roll coating[4]
- High elasticity: stabilize filament for continuous fiber spinning[5]



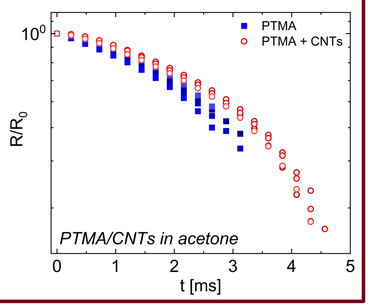
Solvent quality & CNT effects

- Solvent quality: poor (chloroform) → ϑ (acetone) → good (DMF)
- **Solvent quality** ↑ leads to: **elasticity** ↑, λ_E ↑, t_b ↑



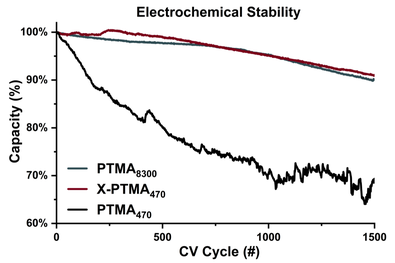


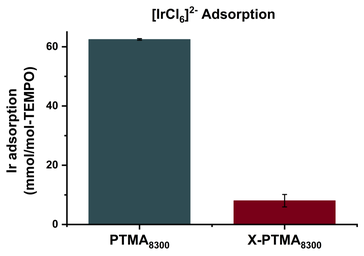
- **Adding CNTs**: η_E ↑, t_b ↑
- Short t_b & η_E → easily sprayed
- Long t_b → prevent misting, ensure clean ink deposition onto substrate



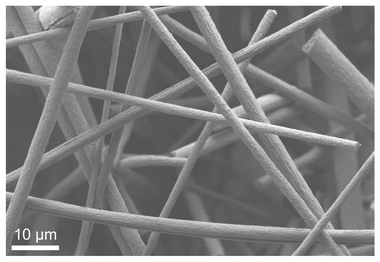
Conclusions & applications

- UHMW PTMA ($M_n > 2.5\ MDa$) can be synthesized easily through industrially-viable pathways
- **Tunable ink elasticity** (λ_E, t_b) can be achieved by controlling polymer M_w, solvent quality, and CNT addition
- Adding CNTs increases viscosity and breakup time





- UHMW PTMA = superior performance in organic electrochemical devices
- Electrodes with UHMW PTMA stable for **1000s** of cycles without requiring crosslinking & adsorb **>6x** more Ir than crosslinked equivalent
- UHMW PTMA also provides sufficient elasticity for fiber spinning
- Successful creation of PTMA/CNT fibers →



References

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Acknowledgements

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