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Multifunctional Hybrid Composites with Gradient Interphase

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Composite Approaches

Micro-Reinforcement "*Traditional*" Approach

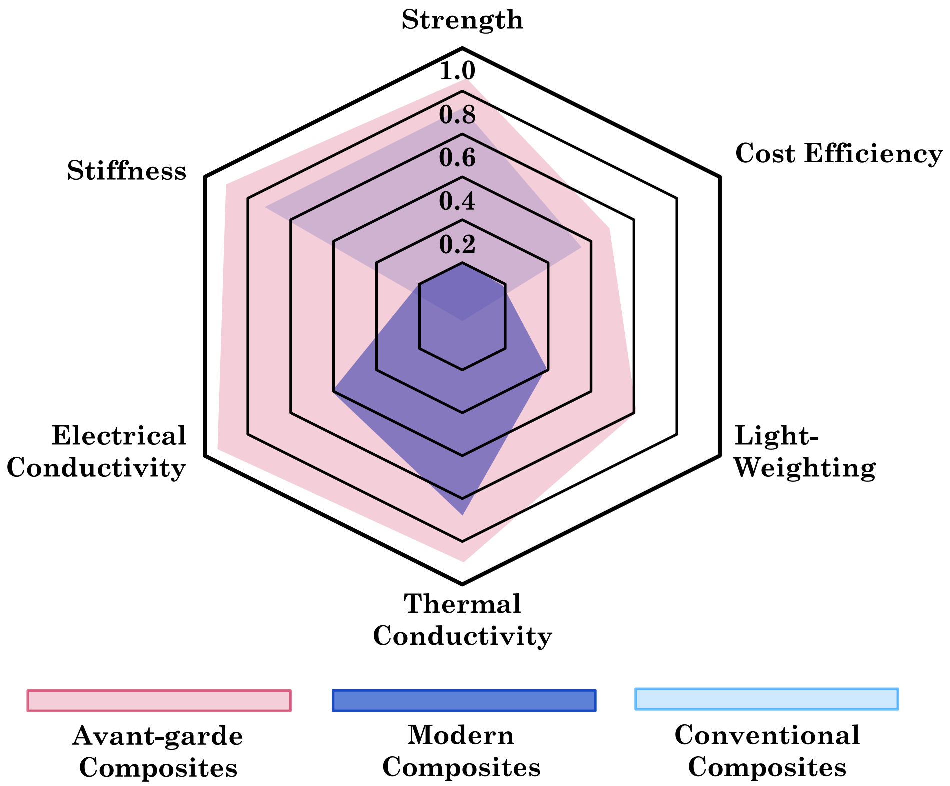
- **GF-reinforced PP-based Composites** are widely used in structural applications in the **automotive industry.**
- Challenges: **High Fiber Loadings (30 – 60 wt.%),** increase in density, decrease in melt flow rate (MFR), increase in brittleness, and reduced processability.

Nano-Reinforcement "*Modern*" Approach

- **Graphene Nanoplatelets (GnPs)** have exceptional mechanical, electrical, and thermal properties, allowing them to be used in a variety of industries.
- Challenges: **Agglomerated GnPs** create areas of **high-stress concentration** that **initiate composite failure.**

Hybrid-Reinforcement "*Avant-garde*" Approach

- **Hybrid composites** have become increasingly popular for a variety of engineering applications, due to their **enhanced properties** compared to traditional biphasic composites.
- Significant Advantage: Originates from the synergistic effect.



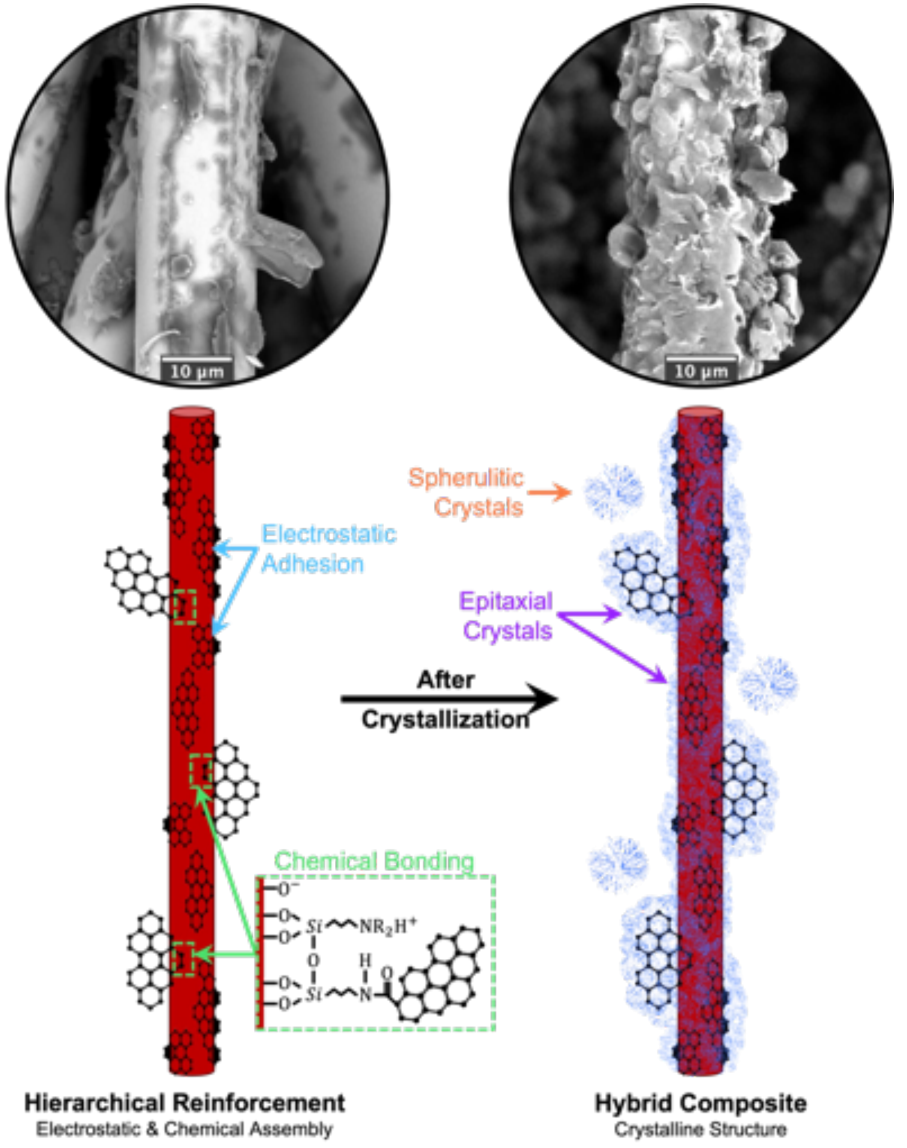
Thomason (2005), Composites Part A: Applied Science and Manufacturing
Liang et al. (2016), Composites Part B: Engineering
Szeluga et al. (2015), Composites Part A: Applied Science and Manufacturing

HIERARCHICAL HYBRID APPROACH

- A **hierarchically-structured hybrid composite**, consist of two reinforcing materials of **different length scales** (i.e., micro-sized and nano-sized) within one matrix material.

- The interfacial interactions between the two reinforcing materials within the hybrid composites can consist of:
  1. Physical Interactions
  2. Electrostatic Interactions
  3. Chemical Interactions

- Generally, **hierarchically-structured fibrous reinforcements**, that are **chemically bonded** (or **grafted**) and/or **electrostatically attached**, are known to **provide greater mechanical properties** and **functionalities,** compared to those that only possess physical interactions.
  - **Tailoring Mechanical Performance**
  - Tailoring Electrical/Thermal Conductivity Performance (EMI Shielding)

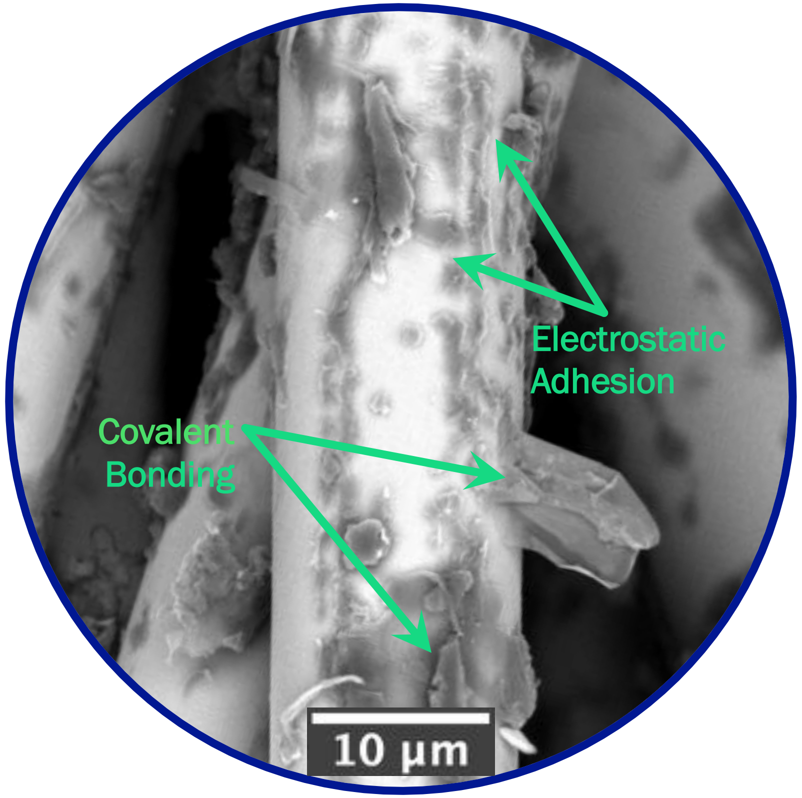


Sansone et al. (2022), ACS Applied Materials & Interfaces
Salari et al. (2023), Chemical Engineering Journal

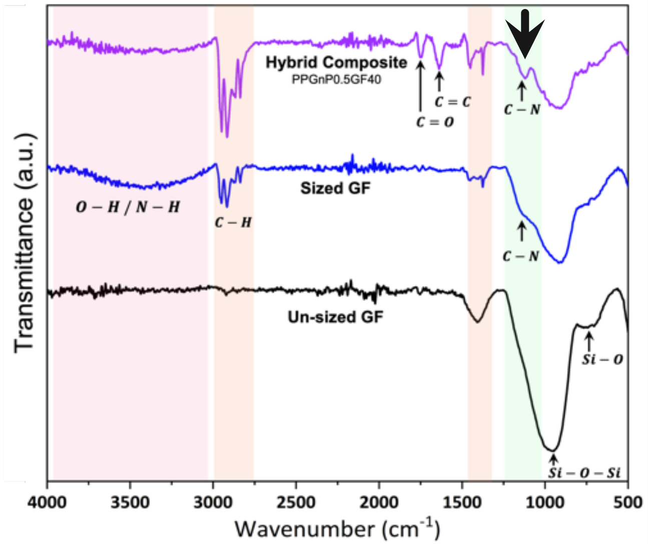
HIERARCHICAL STRUCTURE ASSEMBLY

How can we confirm Self-Assembly?

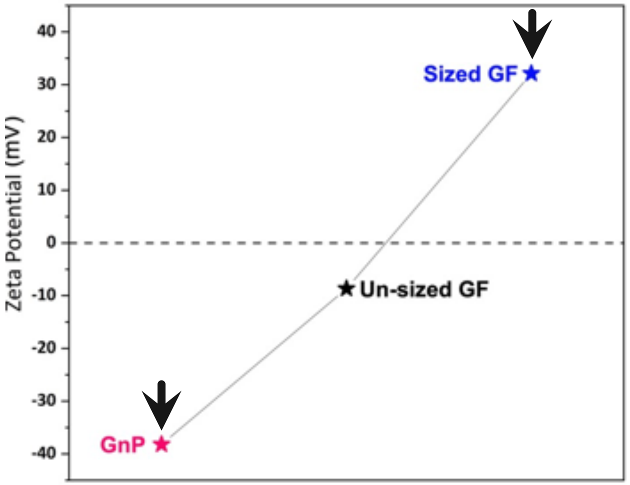
- **Electrostatic Interactions:**
  - Zeta Potential Measurements
- **Chemical Interactions:**
  - Fourier-Transform Infrared Spectroscopy (FTIR)
  - X-Ray Photoelectron Spectroscopy (XPS)

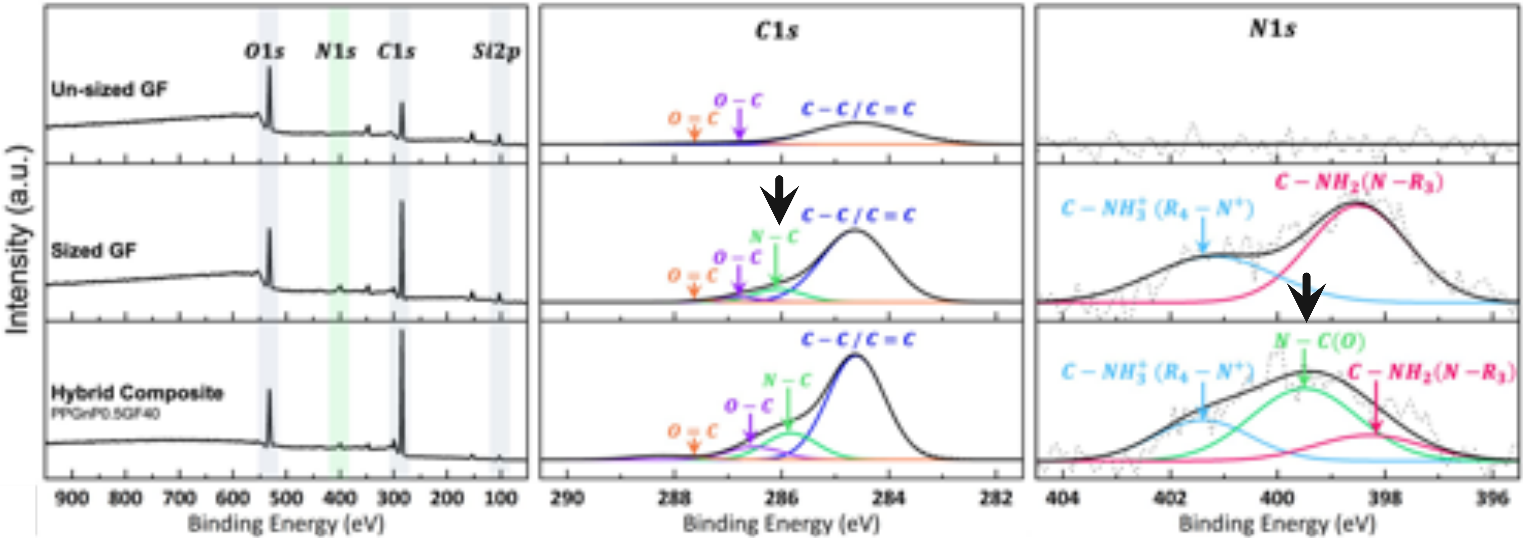


Fourier-Transform Infrared Spectroscopy



Zeta-Potential





X-Ray Photoelectron Spectroscopy

Sansone et al. (2022), ACS Applied Materials & Interfaces

Hierarchical Crystalline Microstructure

Effect of β-Crystal Formation

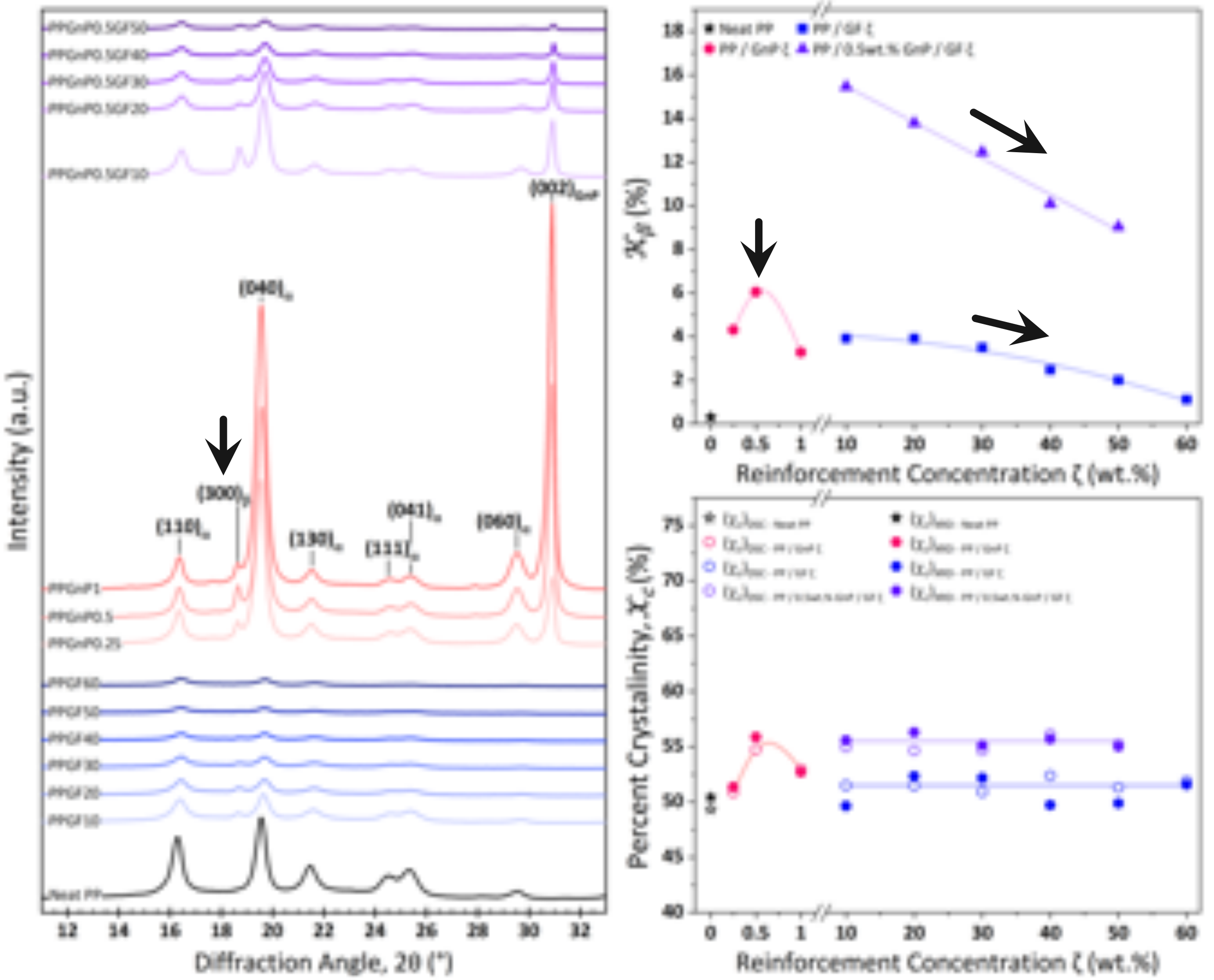
- The $(300)_\beta$ crystallographic plane is indicative of ***β*-crystals** within the crystalline microstructure.

- β-crystals provide **superior mechanical properties:** toughness, tensile strength, elongation at break, and impact strength.

- **Turner-Jones** et al. **Method:**

$$\mathcal{K}_\beta = \frac{H_{\beta(300)}}{H_{\beta(300)} + H_{\alpha(110)} + H_{\alpha(040)} + H_{\alpha(130)}} \times 100\%$$

- **Biphasic PP/GF Composites**: $\mathcal{K}_\beta$ is constant at **~ 4%**, with < 30 wt.% GF. Composites with > 30 wt.% GF decreased with increasing concentration.
- **Biphasic PP/GnP Composites**: optimum formation of $\mathcal{K}_\beta$ found in PPGnP0.5, maximum of 6%.
- **Hybrid PP/GnP/GF Composites**: $\mathcal{K}_\beta$ decreases from ~ 16% as the concentration of GF increases to ~ 9%.



J. Varga (2002), Journal of Macromolecular Science
Sansone et al. (2022), ACS Applied Materials & Interfaces

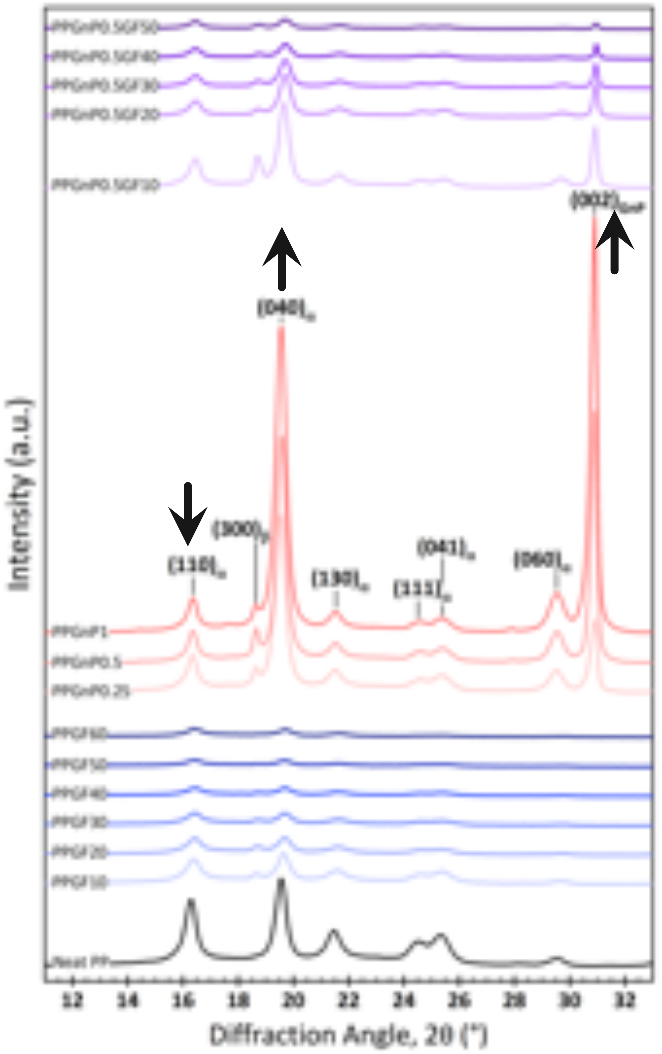
Hierarchical Crystalline Microstructure

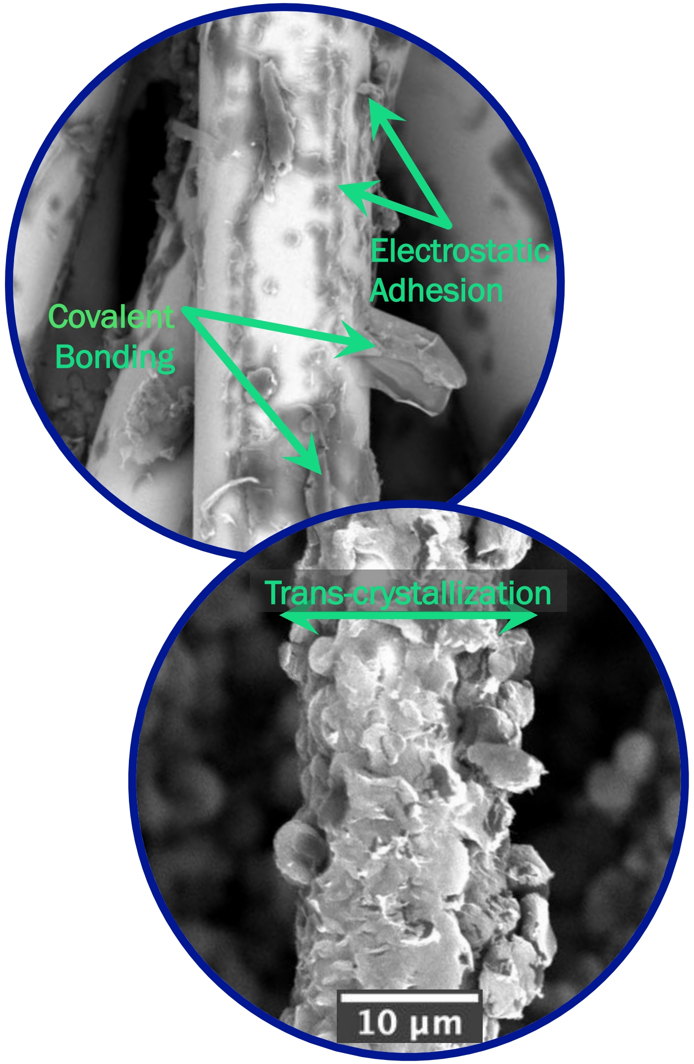
Effect of Trans-crystallization

- Crystallographic plane $(002)_{GnP}$ **increases exponentially**, as the **GnP content increases**, along with:
  - **Increase** in $(040)_{\alpha}$
  - **Decrease** in $(110)_{\alpha}$

- The promotion of trans-crystallization (i.e., the effect of the reinforcements to induce epitaxial growth) is quantified by: $I_{(040)_\alpha}/I_{(110)_\alpha}$
  - **Neat PP:** 1.28
  - **PPGnP0.5:** 7.92
  - **PPGF10:** 1.25
  - **PPGnP0.5GF10:** 3.18

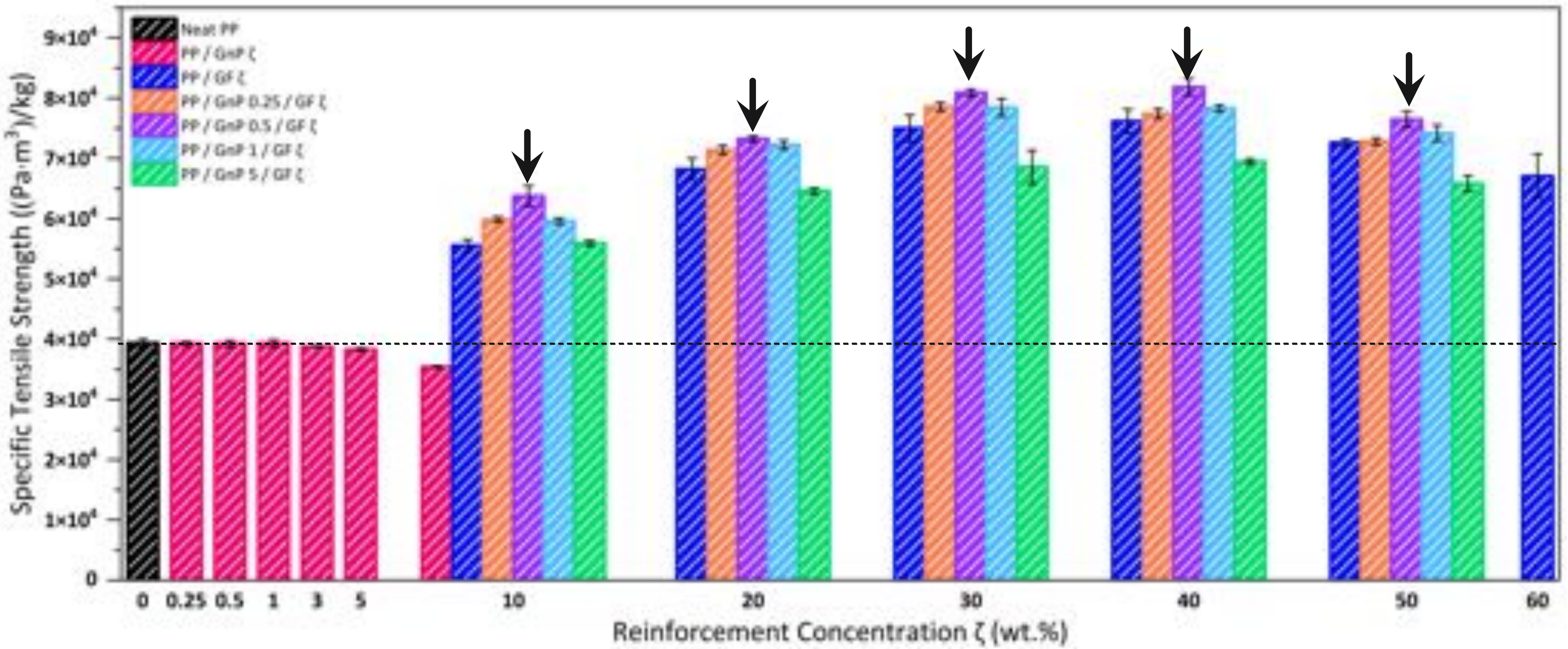
- **Therefore, the successful formation of trans-crystals within the hybrid composites promotes load transfer from the PP matrix to the hierarchically structured reinforcement system.**





Sansone et al. (2022), ACS Applied Materials & Interfaces

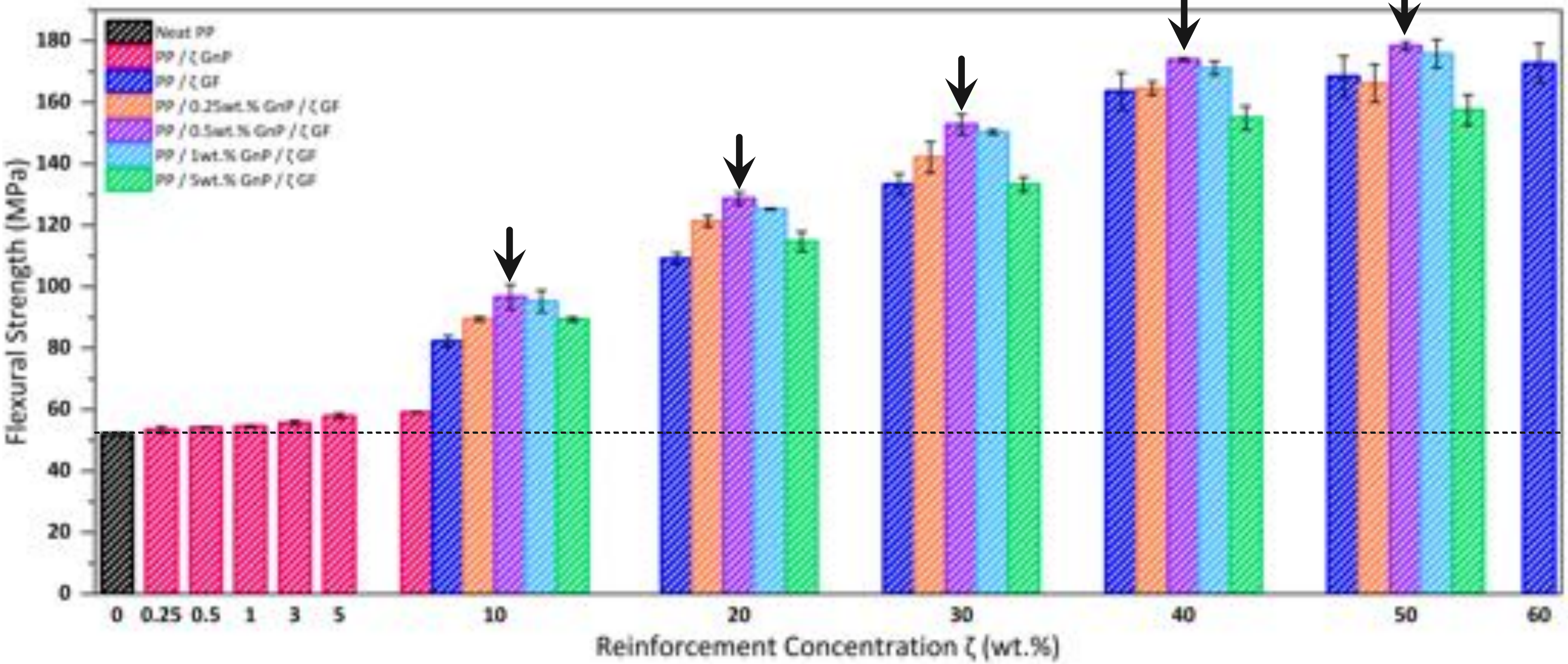
Mechanical Properties - Tensile Strength



- The Specific Tensile Strength of the hybrid PP/GnP/GF composites, containing $\leq 1\ wt.\%$ GnP, perform better than the corresponding biphasic composites with the same concentration of GF.
- An optimum concentration of GnP is observed in the hybrid composites with $0.5\ wt.\%$ GnP, yielding the best tensile strengths for all concentrations of GF.
- This behaviour is attributed to the synergistic effect generated between the two geometrically different fillers.

Sansone et al. (2022), ACS Applied Materials & Interfaces

Mechanical Properties - Flexural Strength



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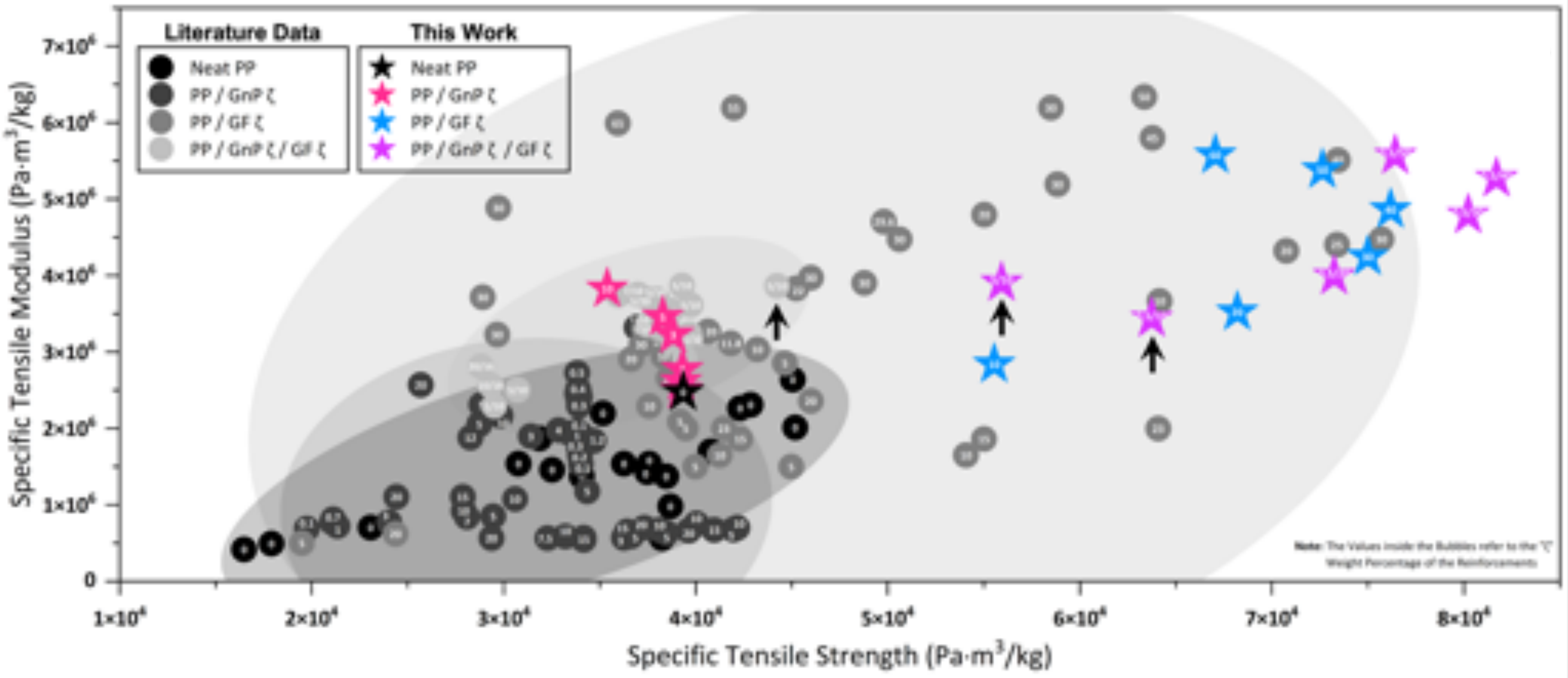
Tensile Mechanical Properties



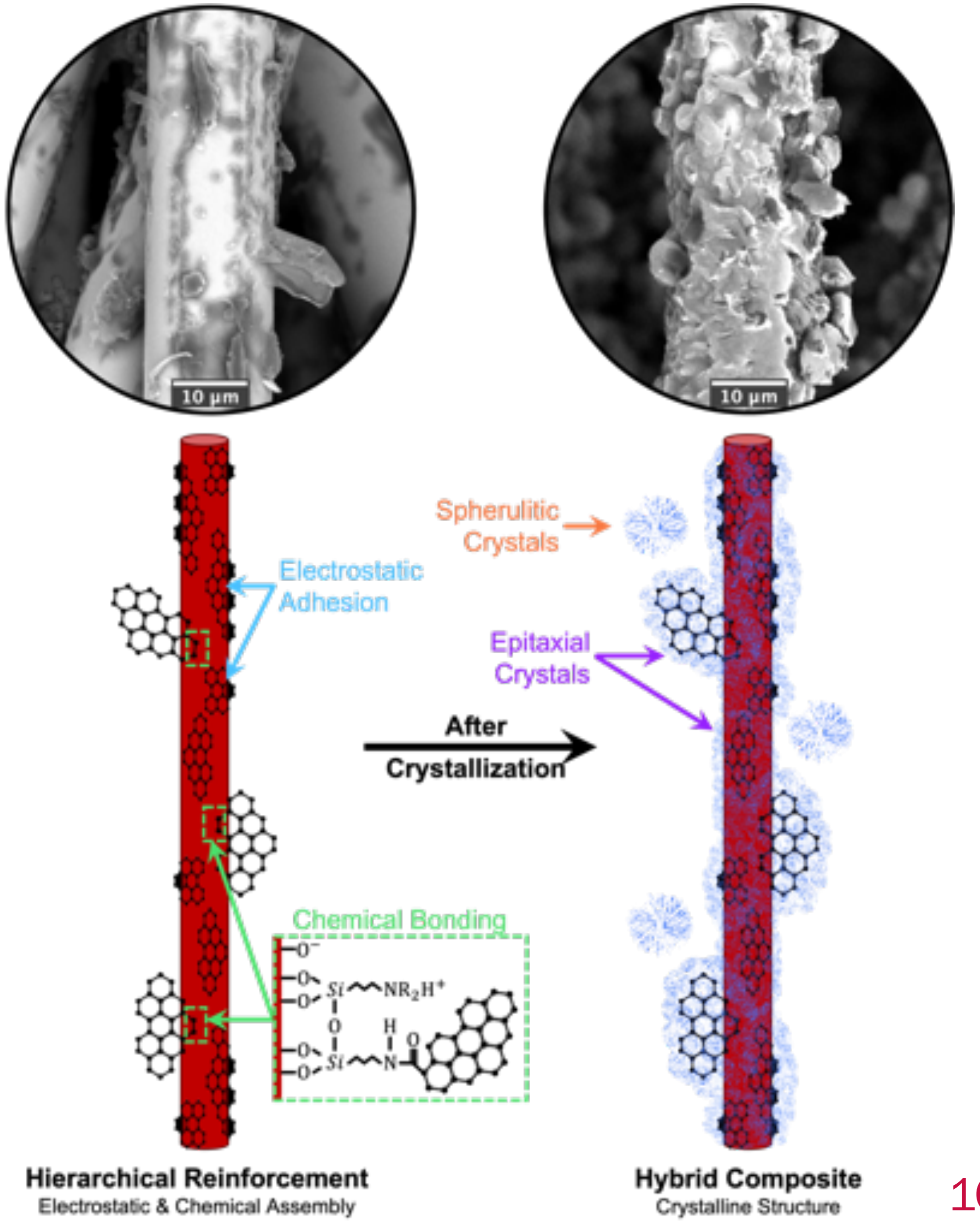
- This Material Selection Chart compares the ***Specific Tensile Modulus*** *versus* ***Specific Tensile Strength*** of PP composites published in literature, to those manufactured in our laboratory.
- The experimental hybrid PP/GnP/GF composites have advantageous stiffness-to-weight and strength-to-weight ratios, compared to their biphasic counterparts.
- The optimum performing hybrid composite is PPGnP0.5GF40, with the highest specific tensile strength.

Sansone et al. (2022), ACS Applied Materials & Interfaces

Conclusion

- The synergistic effect contributing to the enhanced mechanical properties has been attributed to:
  1. The Hierarchical Reinforcement induces a Gradient Interphase that Facilitates Load Transfer at the interface due to the Increased Degree of Trans-crystallization.
  2. The Superior Crystalline Microstructure with Increased Crystallinity and *β*-crystal Formation, enables the matrix to Absorb a Substantial Amount of Energy and Promote Stress Transfer to the reinforcements when exposed to strong mechanical forces.

- Primary Candidates:
  1. PPGnP0.5GF50 **exceeded the flexural strength** of PPGF60 by 3.3% while providing a 9% weight reduction.
  2. PPGnP0.5GF40 obtained the **same desirable flexural strength** as PPGF60, while providing an 18% weight reduction.



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