INCIRCULAR

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INtegrating Cybernated Innovation to Raise the scale of Circular Units Looping Allied Regions

Project INCIRCULAR (I3-2021-INV2a-MANU)
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Document History

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V.1	25.09.2025	First draft
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V.3	16.10.2025	Final Draft

R = Report, P = Prototype, D = Demonstrator, O = Other PU = Public PP = Restricted to other programme participants (including the Commission Services) RE = Restricted to a group specified by the consortium (including the Commission Services) CO = Confidential, only for members of the consortium (including the Commission Services) Restraint UE = Classified with the classification level "Restraint UE" according to Commission Decision 2001/844 and amendments Confidential UE = Classified with the mention of the classification level "Confidential UE" according to Commission Decision 2001/844 and amendments Secret UE = Classified with the mention of the classification level "Secret UE" according to Commission Decision 2001/844 and amendments

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EXECUTIVE SUMMARY

This deliverable presents the outcomes of activities focused on the development and optimisation of recycled fibre-reinforced polymer materials within the INCIRCULAR project. The work highlights the growing importance of recycling in the plastics industry, where circular economy principles drive the transition from linear material use towards sustainable, resource-efficient manufacturing. Recycling of post-consumer and post-industrial plastic waste represents a crucial step in reducing environmental impact, conserving resources, and enabling new material value chains. A key approach to improving the mechanical and functional properties of recycled plastics is the integration of fibre reinforcement. Fibre-reinforced recycled polymers combine the environmental benefits of recycling with enhanced stiffness, strength, and dimensional stability. Within this context, natural and recovered fibres play a significant role in balancing performance and sustainability, offering viable alternatives to traditional glass or carbon fibre reinforcement.

This report provides an overview of the optimized InCircular composite formulations based on recycled polypropylene (rPP) sourced from GORENJE's secondary waste stream, reinforced with 20 wt.% ZZC-500 fibres derived from post-consumer waste and 10 wt.% B600 cellulose fibres from virgin sources. The formulations were systematically designed and evaluated to achieve high-performance, processable, and circular composite materials suitable for industrial applications. A comprehensive description of the optimized formulations, processing parameters, and up-scaling strategies is included. Special emphasis was placed on processing optimisation for extrusion and injection moulding, ensuring material consistency and reproducibility at larger scales. Furthermore, additional analyses were performed on the final selected formulations, including capillary rheometry, Pressure-Volume-Temperature (PVT) analysis, and Dynamic Mechanical Analysis (DMA). These tests provided deeper insight into the rheological behaviour, thermal-mechanical properties, and processing performance of the developed composites.

The findings presented in this deliverable contribute to the establishment of a robust framework for the industrial adaptation of recycled fibre-reinforced materials, promoting sustainable material design and improved end-of-life valorisation pathways within the plastics industry.

ACTION DESCRIPTION

Deliverable ***D3.4 "Report on adaptation of recycled fibre-reinforced materials"*** is directly linked to Milestone ***MS14 "Recycled fibre-reinforced materials selection and production"*** and Milestone ***MS15 "First demonstrators produced and validated"***, achieved by M15 (December 2024) and M24 (September 2025), respectively. These milestones mark the successful completion of the material development phase and the initial validation of demonstrator components produced from the optimized recycled fibre-reinforced formulations.

Deliverable D3.4 consolidates the results of activities carried out under Tasks 3.1, 3.2 and 3.5, focusing on the adaptation, optimisation, and validation of recycled polypropylene (rPP)-based composite formulations reinforced with post-consumer ZZC-500 fibres (20 wt.%) or virgin B600 cellulose fibres (10 wt.%). The work aimed to enhance the mechanical performance, processability, and circularity of recycled plastics while maintaining cost-effectiveness and industrial scalability.

The deliverable provides a comprehensive overview of the optimized composite formulations, detailing the material composition, compounding parameters, and processing conditions for upscaling using extrusion and injection moulding technologies. In addition, it reports on advanced testing campaigns performed on the selected formulations, including capillary rheometry, Pressure-Volume-Temperature (PVT) analysis, and Dynamic Mechanical Analysis (DMA), which offered valuable insights into the rheological, thermal, and viscoelastic behaviour of the developed composites for simulation modelling in T3.2.

The results confirm that the adapted recycled fibre-reinforced materials exhibit improved mechanical integrity and thermal stability suitable for industrial implementation. The findings also serve as the technical foundation for the subsequent validation and integration of these materials into GORENJE demonstrator components, thereby completing the first stage of product-level evaluation within T3.5.

1. INTRODUCTION

1.1. Importance of Recycling in Plastics Industry

The plastics industry is undergoing a profound transformation driven by global environmental, economic, and regulatory pressures. As plastic production continues to increase—surpassing 400 million tonnes annually—the need to reduce waste generation, greenhouse gas emissions, and dependence on fossil-based raw materials has never been more urgent. Recycling plays a central role in this transition, enabling the shift from a linear “take–make–waste” model toward a circular and resource-efficient plastics economy. Recycling in the plastics sector contributes to significant environmental benefits, including reduced landfill disposal, lower incineration-related emissions, and decreased extraction of virgin fossil feedstocks. By closing material loops, recycling minimises the environmental footprint associated with plastic production, particularly in terms of energy consumption and carbon emissions. Secondary plastic materials typically require 70–90% less energy to produce compared to virgin polymers, making recycling one of the most effective measures for mitigating climate impacts within the polymer value chain. From an industrial perspective, the use of recycled polymers represents an increasingly strategic and economically viable solution. The volatility of fossil resource prices, combined with the growing demand for sustainable materials, encourages manufacturers to incorporate recycled feedstocks into their products. For many sectors—including packaging, automotive, consumer goods, construction, and electronics—recycled materials offer opportunities to enhance supply chain resilience, diversify material sources, and meet the rising expectations of end-users and markets.

Regulatory frameworks at both European and global levels are accelerating this shift. Legislation such as the EU Circular Economy Action Plan, Plastics Strategy, Packaging and Packaging Waste Regulation (PPWR), and Eco-design requirements set ambitious targets for recycled content, recyclability, and waste reduction. These measures create strong incentives for companies to adopt recycled materials and develop new technologies that improve recyclate quality and functionality. As a result, the plastics industry is witnessing rapid advancements in mechanical, chemical, and biological recycling, as well as the emergence of hybrid recycling methods that broaden the range of materials that can be effectively recovered. Despite these opportunities, recycled plastics often face challenges related to material quality, property variation, contamination, and degraded mechanical performance. This is particularly critical in applications where structural integrity, dimensional stability, or thermal resistance are required. To overcome these limitations, innovative approaches are being developed—such as fibre reinforcement, compatibilization, advanced sorting, and improved processing techniques—to enhance the performance of recycled polymers and extend their applicability to more technically demanding sectors. Within this context, fibre reinforcement—whether based on natural, synthetic, or hybrid fibre systems—has proven to be a transformative solution. Integrating recovered or bio-based fibres into recycled polymer matrices enables significant improvements in stiffness, strength, impact resistance, and thermal stability, making high-value applications achievable even with secondary materials. This approach supports the principles of the circular economy not only by valorising plastic waste but also by creating pathways for the utilisation of agricultural residues, cellulose-based fibres, and other low-impact reinforcement materials.

Figure 1: *Single Screw Extrusion Line for Recycling and Pelletizing of Rigid Plastic Scrap*

In summary, recycling is a cornerstone of the modern plastics industry, enabling environmental protection, regulatory compliance, and sustainable economic growth. Its importance continues to expand as industries and policymakers adopt circularity-oriented strategies. The development of high-performance recycled fibre-reinforced composites, such as those investigated in the INCIRCULAR project, represents a critical step toward achieving durable, scalable, and environmentally responsible material solutions for the future.

1.2. Role of Fiber Reinforcement in Recycled Polymers

Recycled polymers often suffer from reduced mechanical performance compared to their virgin counterparts due to thermal degradation, chain scission, contamination, and the presence of mixed polymer fractions introduced during the recycling process. As a result, recycled plastics typically exhibit lower tensile strength, impact resistance, and thermal stability, which limits their applicability in demanding technical applications. Fibre reinforcement has emerged as one of the most effective strategies for enhancing the structural and functional properties of recycled polymers and expanding their industrial use. Fibre reinforcement compensates for the inherent deficiencies of recycled polymers by introducing high-aspect-ratio reinforcing elements that improve stiffness, strength, and dimensional stability. Natural fibres (e.g., cellulose, hemp, flax, sisal, banana fibres), synthetic fibres (e.g., glass or carbon fibres), and hybrid fibre systems can all be incorporated into recycled matrices, creating versatile composite structures tailored to specific performance requirements. These reinforcements help transfer mechanical loads more efficiently, reduce deformation under stress, and significantly improve rigidity without excessive increases in material density.

Figure 2: *Natural Fibre examples*

For sustainable and circular material development, natural fibres offer additional advantages such as low weight, reduced environmental footprint, biodegradability, and lower production energy requirements. When combined with recycled plastics, they create hybrid circular materials that maximise resource efficiency and reduce reliance on virgin petroleum-based reinforcements. Post-consumer fibre streams (e.g., recovered cellulose fibres) further enhance circularity by valorising waste into functional composite components. The effectiveness of fibre reinforcement depends on several key factors, including fibre–matrix interfacial adhesion, fibre length and aspect ratio, dispersion uniformity, fibre surface chemistry, and overall composite morphology. Compatibilizers—such as maleic-anhydride-grafted polymers—are often necessary to strengthen bonding between hydrophilic natural fibres and hydrophobic recycled polymer matrices such as PP or PE. Improved interfacial adhesion enables efficient load transfer, resulting in better tensile and flexural performance and enhanced resistance to crack propagation. In addition to mechanical improvements, fibre reinforcement can also contribute to better thermal behaviour, including increased heat deflection temperature (HDT), reduced thermal expansion, and improved dimensional stability during processing and application. These enhancements are particularly relevant for industrial sectors such as household appliances, automotive components, packaging systems, construction elements, and consumer goods, where recycled polymers alone may not meet performance requirements. By enabling recycled plastics to achieve mechanical and thermal properties closer to, or in some cases exceeding, those of virgin materials, fibre reinforcement significantly broadens the potential market applications for secondary raw materials. This approach supports the development of higher-value recycled composites, promoting greater material retention within the circular economy and reducing the need for virgin resources.

Overall, fibre reinforcement plays a critical role in unlocking the full potential of recycled polymers, enabling sustainable, high-performance materials that align with environmental, regulatory, and industrial expectations. In the context of the INCIRCULAR project, these principles are fundamental to the development of advanced recycled fibre-reinforced composites capable of meeting the requirements of real industrial applications and demonstrator validation.

1.3. Overview of the InCircular Optimized Formulations

Within the framework of the INCIRCULAR project, the development and optimization of recycled fibre-reinforced polypropylene (rPP) formulations represented a central activity aimed at creating high-performance, circular composite materials suitable for industrial use. Building on the material selection criteria established in D3.1 and D3.2 the processing and characterization activities conducted in D3.3, several candidate formulations were produced, compounded, and subjected to extensive mechanical, thermal, rheological, and morphological evaluation. The optimization process considered key performance indicators relevant to real industrial environments, including processability (extrusion and injection moulding suitability), dimensional stability, mechanical strength and stiffness, thermal behaviour, moisture sensitivity, and surface quality. Fibre–matrix compatibility was also a critical parameter, as effective interfacial adhesion directly determines the reinforcement efficiency and overall composite performance.

Following a systematic assessment and characterization results of natural and recycled cellulose fibre types, fibre loadings, compatibilizers, and processing conditions, two composite formulations emerged as the most promising candidates:

- **ZZC500-20:** recycled PP reinforced with 20 wt.% of ZZC500 recycled cellulose fibres obtained from post-consumer waste streams.
- **B600-10:** recycled PP reinforced with 10 wt.% of B600 bleached virgin cellulose fibres.

Both formulations demonstrated a favourable balance between mechanical performance, thermal stability, and production scalability, making them suitable for further development within the project. The ZZC500-20 composite showed excellent reinforcement potential, attributed to improved interfacial adhesion between the rPP matrix and the recycled cellulose fibres. This synergy enabled enhanced tensile and flexural properties and increased stiffness, supporting the suitability of this formulation for structural components where material robustness and circularity are essential. Despite the higher fibre content, processability remained stable, facilitated by optimized compounding parameters and controlled moisture management strategies. The B600-10 formulation exhibited superior surface quality, lower viscosity, and highly stable processing behaviour, making it particularly suitable for applications requiring both aesthetic and mechanical integrity. The virgin B600 cellulose fibres provided uniform dispersion, consistent fibre-matrix bonding, and improved dimensional stability during injection moulding. These characteristics are especially relevant for demonstrators where visual appearance and precision are critical. Together, the two optimized formulations provide complementary performance advantages: ZZC500-20 aligns strongly with high-circularity objectives and structural reinforcement scenarios, while B600-10 ensures high process reliability and superior final-part quality. These materials form the technical foundation for subsequent activities in WP3, including digital simulation, moulding trials, demonstrator fabrication, and validation under industrial conditions. The formulation insights and test results presented in this section confirm the feasibility of producing reliable, high-quality recycled fibre-reinforced composites from both post-consumer and virgin fibre sources, supporting the broader objectives of the INCIRCULAR project to advance sustainable material innovation and industrial circularity.

2. MATERIALS OVERVIEW

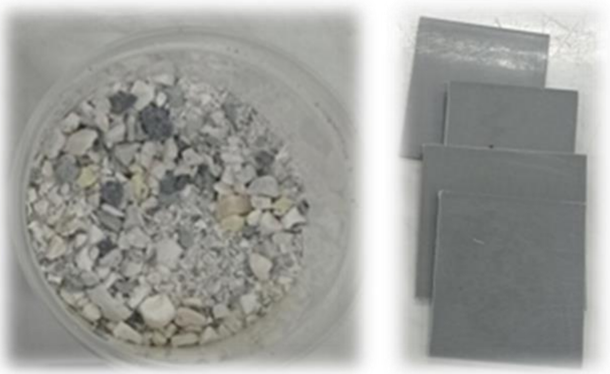
2.1 Recycled Polypropylene (rPP): Sources and Properties

The white goods industry, which includes large household appliances such as refrigerators, washing machines, and dishwashers, generates substantial quantities of polypropylene (PP) waste at the end of the products' life cycle. This waste stream represents a valuable secondary raw material; however, its heterogeneity poses a challenge for advanced material valorisation. The recycled polypropylene (rPP) obtained from such appliances typically consists of a diverse blend of PP grades and filler systems originally used to meet specific functional and mechanical performance requirements of the appliance components. In the case of GORENJE IPC, the rPP stream reflects the company's broad use of PP in various applications—ranging from structural housings and internal mechanical parts to decorative or non-visible elements. As a result, the recycled material encompasses a mixture of PP homopolymers, random and impact copolymers, and a variety of mineral- and fibre-reinforced compounds. The presence of these materials provides both opportunities and constraints in the development of recycled fibre-reinforced composites.

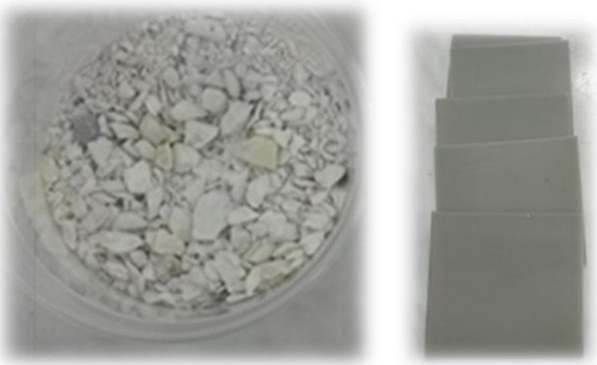
2.1.1 Composition of rPP from GORENJE

Two major rPP material groups were identified during the on-site inventory:

1. Mixed PP with mineral fillers, consisting of:
    - PP with talc
    - PP with calcium carbonate ($CaCO_3$)
    - Unfilled PP with total filler contents reaching up to 40 wt.%.



2. Glass-fibre-reinforced PP (PP-GF) containing 35–50 wt.% glass fibres.



Such diversity arises from the functional requirements of appliance components. Mineral fillers such as talc and $CaCO_3$ are used to enhance stiffness, dimensional stability, and heat resistance, while glass fibres increase strength, impact resistance, and thermal stability. These fillers remain embedded in the recycled material and significantly influence the performance and reusability of the resulting rPP matrix.

2.1.2 Material Recovery and Pre-Processing

The rPP originating from GORENJE is typically collected in the form of irregular plastic lumps, which require systematic pre-processing to obtain a homogeneous and processable material stream. Within Task 3.1, a dedicated sorting and collection methodology was established at the start of the project. On 16 February 2024, a total of 1,570 kg of separated PP-based material was delivered to OMAPLAST for further treatment.

At OMAPLAST, the material underwent a two-stage size reduction protocol using industrial shredding and grinding technologies:

- Stage 1: Shredding with a 150 mm screen (Lindner system), followed by ferrous metal extraction.
- Stage 2: Fine grinding through a 10 mm mesh (Sorema system), producing a clean and uniform regrind fraction.

The resulting output comprised 1,365 kg of reprocessed rPP, divided into:

- 1,141 kg of mineral-filled rPP (talc and $CaCO_3$ blend)
- 224 kg of glass-fibre-reinforced rPP

This material was subsequently packaged and delivered to TECOS for detailed characterization and compounding.

2.1.3 Characterization of Recovered Materials

Laboratory analyses confirmed the suitability of the recovered materials as polymer matrix candidates for further composite development under the INCIRCULAR project. Key findings included:

- **Spectroscopy (FTIR):** confirmed PP purity with no detectable presence of foreign polymers.
- **Burn Test:** verified the inclusion of mineral fillers and glass fibres.
- **Density Measurements:** indicated the presence of both filled and unfilled polymer fractions.
- **Visual Inspection:** identified localized burn marks, degradation spots, and minor impurities typical of post-consumer appliance waste streams.

Quantitative analyses revealed filler contents of **31.4 wt.%** in the talc/$CaCO_3$-filled rPP fraction and **35.4 wt.%** in the glass-fibre-filled rPP fraction.

2.2 Post-Consumer Waste Fibres: Origin and Characteristics

The characterization of post-consumer waste fibres was conducted using a comprehensive set of analytical techniques to evaluate their suitability as reinforcement materials in recycled polymer composites. These fibres—primarily sourced from post-consumer cellulose waste streams—were assessed for their structural, chemical, thermal, and physio-mechanical properties, providing a complete understanding of their performance potential within the INCIRCULAR material formulations.

The analyses performed include elemental composition, moisture and ash content, chemical composition, thermal behaviour (TGA/DSC), functional group identification (FTIR), water and oil retention capacity, dimensional properties (length, thickness, aspect ratio), and physico-chemical characteristics (bulk density, pH, zeta potential). Additionally, SEM imaging provided high-resolution insights into fibre morphology, enabling precise evaluation of fibre geometry and consistency. This multi-method approach ensures robust fibre selection and supports the development of high-performance fibre-reinforced recycled composites.

Figure 3: *Natural Fibres used for INCIRCULAR project*

2.2.1 Elemental Analysis

The fibres exhibit a high carbon (C) content (38–45%), consistent with cellulose-based materials. Recycled fibres show the highest overall elemental percentages (CNH-O), likely due to impurities or additives present from previous use or processing histories. These findings align with typical values reported in literature for recycled cellulose fibres.

2.2.2 Moisture and Ash Content

Moisture content across fibres ranged from 5–8%, which can negatively affect mechanical properties, fibre–matrix adhesion, and dimensional stability in composite applications. Fibres with higher water uptake—such as BC-300—may require pre-treatment or surface modification to enhance compatibility with hydrophobic polymer matrices. Ash content was notably higher in recycled fibres (16–23%), reflecting the presence of residual mineral impurities or additives originating from the prior lifecycle of the material.

2.2.3 Chemical Composition

Analysis of oxide composition revealed typical inorganic components found in both virgin and recycled cellulose fibre ash, including CaO, MgO, K_2O, Na_2O, SiO_2, and Al_2O_3. The higher inorganic fraction in recycled fibres correlates with their elevated ash content and signals prior chemical exposure or contamination.

2.2.4 Thermogravimetric (TGA) and Differential Scanning Calorimetry (DSC)

All fibres exhibited major mass loss (~80%) around 330–350 °C, corresponding to cellulose decomposition. Importantly, no thermal degradation occurred below the glass transition temperatures (Tg) of common polymers, confirming their suitability for use in thermoplastic composite processing.

2.2.5 Fourier Transform Infrared Spectroscopy (FTIR)

FTIR analysis identified key functional groups such as –OH, C–O–H, and C=O, which are responsible for fibre hydrophilicity and play a critical role in fibre–matrix interfacial adhesion. These groups support mechanical anchoring when paired with maleic-anhydride-grafted compatibilizers.

2.2.6 Dimensional Analysis: Length, Thickness, and Aspect Ratio

Fibre geometry varied significantly across samples. Aspect ratio—an essential parameter affecting reinforcement efficiency—differs according to fibre type. SEM imaging confirmed wide morphological variability, especially in SIS-250 fibres, which exhibited the largest discrepancies between reported and measured dimensions.

2.2.7 Bulk Density

Laboratory-determined bulk density values for BC-300 and B-600 were slightly higher than those in the technical datasheets, while SIS-250 exhibited notably lower density. PWC-500 and ZZC-500 values fell within expected ranges.

2.2.8 pH and Zeta Potential

All fibres demonstrated pH values within acceptable ranges specified by their datasheets. Zeta potential measurements, although not provided in technical documentation, revealed negative surface charges consistent with cellulose chemistry. These values suggest similar fibre–surface behaviour across fibre types.

2.2.9 SEM Morphological Assessment

SEM imaging revealed variability in fibre lengths and thicknesses, reinforcing the need for controlled dispersion and compounding conditions. Recycled fibres often showed mixed morphologies, a common characteristic of post-consumer inputs.

2.2.10 Summary and Conclusions

The analysed post-consumer waste fibres display a diverse range of chemical and physical characteristics that influence their performance in recycled polymer composites. Recycled fibres tend to exhibit:

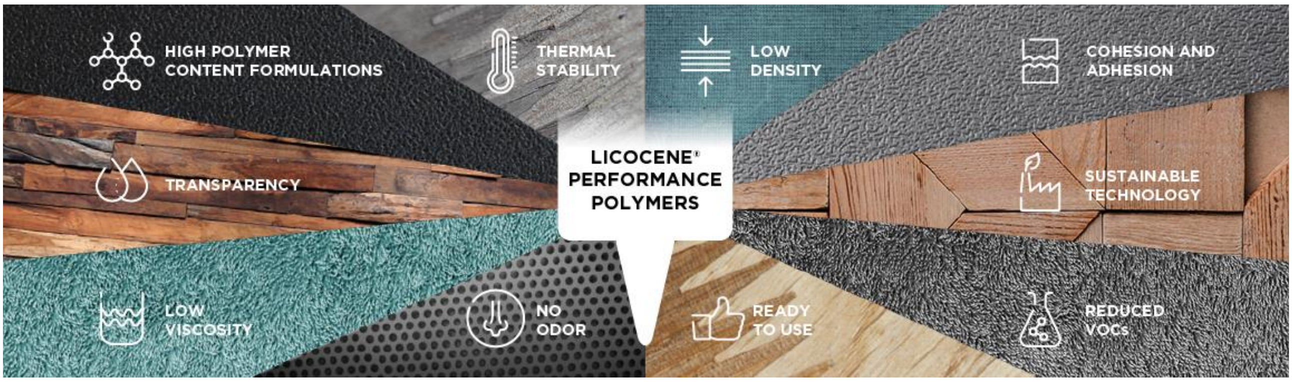
- Higher ash content due to impurities
- Broader dimensional variability
- Strong thermal stability suitable for thermoplastic processing
- Functional groups beneficial for compatibilization
- Sufficient structural integrity for reinforcement

These findings confirm the technical feasibility of using post-consumer fibres in rPP composites, provided that appropriate processing strategies are implemented (e.g., drying, compatibilization, controlled dosing). The comprehensive characterization supports informed fibre selection and ensures that the chosen materials offer a balance of sustainability, processability, and mechanical performance for composite development within the INCIRCULAR framework.

2.3 Additives and Coupling Agents (MA-g-PP and Compatibilizers)

The incorporation of additives and coupling agents is essential for enhancing the performance of polymer composites, particularly when combining materials with intrinsically different chemical structures, polarities, or surface energies. In fibre-reinforced recycled polypropylene (rPP) systems, achieving strong interfacial bonding between hydrophobic PP and hydrophilic lignocellulosic fibres is critical to ensuring mechanical integrity, processing stability, and long-term durability. State-of-the-art composite engineering strategies commonly employ reactive compatibilizers—most notably maleic anhydride-grafted polypropylene (MA-g-PP)—to overcome these chemical incompatibilities and to promote efficient stress transfer across the fibre–matrix interface.

MA-g-PP is broadly recognised in academic and industrial literature as the most effective and widely used coupling agent for PP-based composites reinforced with natural fibres, mineral fillers, or engineering polymers. Numerous studies demonstrate that its use significantly improves composite stiffness, tensile strength, and moisture resistance while enabling higher fibre loadings and mitigating phase separation in multiphase blends. Thus, in the context of circular materials engineering, compatibilizers such as MA-g-PP play a crucial role in enabling the valorisation of recycled fibres within high-quality polymer matrices.



2.3.1 *Composition and Functionality*

MA-g-PP is produced by grafting maleic anhydride onto polypropylene chains via a free-radical mechanism, typically initiated by organic peroxides. This process creates reactive maleic anhydride groups on an otherwise non-polar, hydrophobic PP backbone.

These polar grafts introduce chemical functionality capable of forming:

- Hydrogen bonds with hydroxyl groups on cellulose,
- Covalent or ester linkages under appropriate processing conditions,
- Dipolar interactions with oxygen-containing surface groups.

These interactions significantly enhance compatibility between PP and hydrophilic fibres. In composite systems, the result is improved wetting of fibre surfaces, reduced interfacial voids, and stronger adhesion at the fibre–matrix interface.

2.3.2 *Key Applications of MA-g-PP in Composite Systems*

- **Natural Fibre-Reinforced Composites**

  MA-g-PP is indispensable when reinforcing PP with cellulosic fibres (wood, hemp, jute, sisal). It compensates for differences in polarity, resulting in improved tensile strength, flexural stiffness, and impact performance, as well as reduced water uptake.

- **Filler-Modified PP ($CaCO_3$, talc, silica)**

  The compatibilizer enhances filler dispersion and reduces agglomeration, enabling efficient reinforcement even at higher filler loadings.

- **PP Blends with Engineering Polymers (PA, PET)**

  In multiphase polymer blends, MA-g-PP reduces interfacial tension between incompatible phases, improving toughness, processability, and overall blend homogeneity.

- **Recycled and Mixed Waste Polymer Streams**

  Recycled PP typically contains impurities and trace amounts of polar polymers. MA-g-PP stabilizes these heterogeneities, increasing consistency, melt strength, and final part quality.

2.3.3 Benefits of Using MA-g-PP

- **Enhanced Interfacial Bonding:** Stronger fibre–matrix adhesion promotes efficient load transfer and reduces microvoids.
- **Improved Filler/Fibre Dispersion:** Leads to uniform morphology, better mechanical performance, and reduced processing defects.
- **Increased Mechanical Performance:** MA-g-PP-modified composites consistently outperform uncompatibilized systems in tensile, flexural, and impact properties.
- **Improved Thermal Behaviour:** The compatibilizer stabilizes the interface and maintains composite integrity during processing and elevated service temperatures.
- **Greater Sustainability in Recycled Systems:** Enables effective reuse of post-consumer fibres by compensating for their surface heterogeneity.

2.3.4 Challenges and Considerations

- **Cost Implications:** The use of MA-g-PP increases material costs, though its performance benefits often justify its use in high-value applications.
- **Optimal Grafting Level:** Insufficient grafting produces weak interfacial bonding, while excessive grafting can embrittle the composite due to over-crosslinking or oxidative degradation.
- **Processing Sensitivity:** High shear or overheating during extrusion can degrade both PP and maleic anhydride groups, reducing compatibilization efficiency. Careful optimisation of graft level, dosage (typically 2–5 wt.% in natural fibre composites), and compounding conditions is therefore required.

2.3.5 Functional Role in This Project

In the INCIRCULAR formulations, 3 wt.% Licocene PP MA 7452 GR was used as the coupling agent across all fibre types and loadings. Its functions in the tested composites include:

- Improving compatibility between hydrophobic recycled PP and hydrophilic cellulose fibres (virgin or post-consumer).
- Enhancing fibre surface wetting, enabling uniform dispersion during extrusion compounding.
- Stabilising the interface during injection moulding, reducing porosity and improving dimensional stability.
- Supporting mechanical integrity by enhancing interfacial adhesion—particularly critical for the high-performing ZZC500-20 formulation.

The overall results confirm that MA-g-PP was essential for achieving the favourable mechanical, thermal, and morphological characteristics observed in the optimal composite batches.

Figure 4: *Coupling agent - Licocene PP MA 7452 GR used for INCIRCULAR project*

2.3.6 Summary

MA-g-PP plays a decisive role in the success of PP-based composites reinforced with natural or recycled fibres. By bridging the interface between hydrophilic cellulose and hydrophobic PP, it enhances mechanical strength, improves dispersion, stabilises the composite structure, and enables the integration of post-consumer waste fibres into high-performance materials. Its use aligns with circular economy principles by facilitating the development of durable, recyclable, and resource-efficient composite products.

2.4 Selection Rationale for Fibre–Matrix Combination

A broad screening of cellulose-based fibres was conducted to identify the most suitable reinforcement candidates for recycled polypropylene (rPP) matrices originating from GORENJE. The fibre selection included materials from different origins and purity levels to evaluate their compatibility, reinforcement efficiency, and processing behaviour within recycled polymer streams. Five fibre families were examined:

- **B600 and BC300** – virgin bleached hardwood cellulose fibres
- **PWC500** – pre-consumer recycled cellulose fibres
- **ZZC500** – post-consumer recycled cellulose fibres
- **SIS250** – recycled sisal fibres

To enhance interfacial adhesion between the hydrophilic fibres and the hydrophobic PP matrix, 3 wt.% of a maleic-anhydride-grafted PP compatibilizer (Licocene PP MA 7452 GR) was consistently added. Fibre loadings of 10%, 20%, and 30% were processed following a structured compounding protocol that included manual pre-mixing of fibres, compatibilizer, and milled rPP to ensure uniform dispersion and stable dosing during extrusion.

2.4.1 Processing and Performance Evaluation

The fibres were assessed based on their processing behaviour during compounding (dosing stability, pelletizing performance, moisture behaviour), as well as their mechanical, thermal, and physicochemical effects on the final composites.

- **ZZC500 fibres** showed very good processability at 10% and 20% loadings, with only moderate challenges at 30%. They provided consistent drying behaviour, stable dosing, and good pelletizing characteristics, even at higher fibre contents.
- **B600 fibres**, while offering excellent surface quality and strong reinforcement at lower loadings, presented significant processing difficulties at 30%, requiring discontinuous manual feeding and causing clogging at the hopper.
- **Other fibre families** (PWC500, BC300, SIS250) showed balanced but less optimal combinations of crystallinity, dimensional stability, and thermal conductivity compared to ZZC500 and B600.

Thermal characterization confirmed that the ZZC500 family demonstrates one of the best overall balances, combining high thermal stability, stable crystallinity progression, controlled thermal expansion, and strong thermal conductivity improvements—despite being a fully recycled post-consumer resource.

2.4.2 Final Fibre Selection

Based on a comprehensive evaluation of all material families, including mechanical performance, processability, thermal stability, and morphology**,** two formulations were selected for further validation and simulation activities in subsequent work packages:

- **ZZC500-20**: rPP reinforced with **20 wt.%** post-consumer recycled cellulose fibres
- **B600-10**: rPP reinforced with **10 wt.%** virgin bleached hardwood cellulose fibres

Both materials demonstrated a strong balance between processing stability and performance. However, the ZZC500-20 formulation was ultimately selected for up-scaling within the project due to:

1. **Clear environmental advantages**, as it utilizes post-consumer recycled cellulose, directly supporting circular economy objectives.
2. **Excellent reinforcement performance**, with strong interfacial adhesion and significantly improved crystallinity, mechanical strength, and thermal stability.
3. **Reliable processing**, especially at 20% fibre loading, where dosing, drying, and pelletizing remained stable and industrially feasible.

2.4.3 Conclusion

The fibre selection process identified ZZC500-20 as the most promising composite system for industrial-scale production, offering the best combination of sustainability, processing efficiency, and overall performance. This formulation will therefore serve as the primary material for further development, scaling, and demonstrator manufacturing in the following stages of the INCIRCULAR project.

3. UPSCALED EXTRUSION COMPOUNDING PROCESS

3.1 Compounding Process and Upscaling Strategy

The transition from laboratory-scale composite development to industrial-scale production is a critical step in ensuring the technical feasibility, economic viability, and replicability of newly developed materials. In the INCIRCULAR project, compounding served as the central processing stage for converting recycled polypropylene (rPP), post-consumer cellulose fibres, virgin fibres, and additives into homogeneous, high-performance composite granulates suitable for injection moulding applications. This chapter provides an overview of the compounding principles, describes the specific challenges associated with upscaling fibre-reinforced recycled polymer systems, and presents the successful implementation and validation of the compounding process for the selected INCIRCULAR formulations.

Figure 5: *USEON Lab30 Compounding and Granulating Production Line*

3.1.1 Fundamentals of Twin-Screw Extrusion for Composite Compounding

Twin-screw extrusion is the industry-standard technology for producing polymer composites, especially when precise dispersion of fibres, fillers, and additives is required. The co-rotating twin-screw design ensures intensive mixing through a combination of shear, elongational flow, and controlled residence time.

Key advantages of twin-screw extrusion for composites:

- **High dispersion efficiency:** Ensures uniform distribution of natural fibres, fillers, and compatibilizers within the polymer matrix.

- **Precise temperature control:** Essential for preventing thermal degradation of cellulose fibres and recycled polymers.
- **Modular screw configuration:** Allows tailoring of kneading blocks and conveying zones to improve fibre wetting and minimize fibre breakage.
- **Scalable process parameters:** Laboratory-scale settings can be transferred to industrial extruders while preserving material quality.

Critical process parameters for fibre-filled rPP systems:

- **Temperature profile:** Must be optimized to ensure melt homogeneity without overheating cellulose fibres (typically < 210 °C).
- **Screw speed:** Influences shear forces, fibre breakage, and dispersion uniformity.
- **Feeding stability:** Natural fibres, especially recycled ones, may have variable moisture and density, requiring careful dosing.
- **Pelletizing:** Cutting speed must match melt flow characteristics to obtain uniform granules.

The INCIRCULAR compounding workflow followed these principles, adapting them to the specific challenges of processing recycled PP with both virgin and post-consumer cellulose fibres.

Figure 6: *Hoppers (main and side extrusion hoppers) with INCIRCULAR material components*

Figure 7: *USEON Lab30 Compounder - Output: ~50 kg/h (depends on formula and operation conditions)*

Screw Diameter: 30mm; Max speed: 600 rpm; L/D = 32/64; Motor: 18.5 kW

3.2 Compounding Challenges in Upscaling Recycled Fibre-Reinforced rPP

Upscaling composite production from pilot scale to industrial scale introduces several challenges:

- **Variability of recycled feedstock**

  rPP originating from the white goods sector contains inherent heterogeneity in polymer grade, filler content, and thermal history. This variability can affect melt rheology, fibre wetting, and final properties.

- **Fibre handling and dosing issues**

  Post-consumer fibres as ZZC500 exhibit fluctuating bulk density and moisture content, leading to:

  - dosing instability,
  - occasional hopper clogging,
  - increased drying requirements.

- **Thermal sensitivity of cellulose fibres**

  Excessive residence time or elevated temperature can cause:

  - fibre degradation,
  - discoloration,
  - odour release,
  - decrease in mechanical properties.

- **Maintaining dispersion at high fibre loadings**

  20–30% fibre content increases viscosity, which may result in:

  - screw overload at high throughput rates,
  - higher torque,
  - fibre agglomerates if mixing is insufficient.

- **Achieving consistent pellet quality**

  Uniform pellet geometry is critical for stable injection moulding. High fibre content increases risk of:

  - strand breakage,
  - uneven pellet size,
  - void formation during cooling.

These challenges guided the optimization of compounding parameters used for the INCIRCULAR formulations.

3.3 Optimized Processing Parameters for INCIRCULAR Formulations

Following iterative testing and parameter refinement, a robust and scalable compounding protocol was established. The optimized processing window was defined as follows:

Temperature profile: 160 – 170 – 185 – 190 – 195 – 200 °C

This profile ensured stable PP melt flow while preventing fibre degradation.

Screw rotation speed: 150–200 rpm

Balanced shear intensity and fibre retention while enabling efficient dispersion.

Pelletizing speed: ~200 rpm

Provided consistent pellet morphology suitable for downstream injection moulding.

Drying conditions: 24–48 hours at 60 °C

Ensured moisture removal from fibres and reduced the risk of voids in the extruded composite.

These parameters resulted in:

- stable extrusion with minimal fluctuations in torque and pressure,
- homogeneous dispersion of fibres within the rPP matrix,
- high pellet quality with repeatable rheological behaviour.

3.4 Upscaling Production: From Pilot Scale to Industrial Trials

Following successful laboratory development, TECOS implemented the optimized compounding parameters to produce larger material batches for industrial validation.

Production volumes achieved:

Material Type	Quantity	Destination / Purpose
INCIRCULAR composite	**235 kg**	Overall pilot-scale production
ZZC500-20	Included in total	For demonstrators and stakeholder testing
B600-10	Included in total	For demonstrators and stakeholder testing
PP-GF recycled composite	**50 kg**	IPC trials for washing machine components
INCIRCULAR composite	**110 kg**	IPC demonstrator production – industrial box
INCIRCULAR composite	**25 kg**	Ecocastulum replication demonstration
ZZC500-20	**50 kg**	TECOS + Slovenian stakeholder collaborations

This distribution enabled full evaluation of materials in multiple real-world industrial contexts.

Industrial demonstrator implementations:

1. **IPC industrial boxes:**

   Assessment of mechanical integrity, surface quality, injection moulding stability, and cycle time optimization.
2. **Washing machine components (PP-GF):**

   Testing for dimensional accuracy, stiffness, thermal resistance, and assembly compatibility.
3. **Replication demonstrators (Ecocastulum):**

   Validation in external stakeholder environments.

These upscaling activities confirmed that the optimized compounding process delivers high-quality pellets suitable for industrial-scale moulding.

3.5 Selection of Optimal Composite Formulations for Full Upscaling

Based on mechanical, thermal, rheological, morphological, physiological, and processing analyses, two formulations were selected as the optimal INCIRCULAR composites:

3.5.1 B600-10

Recycled PP reinforced with 10% virgin B600 cellulose fibres

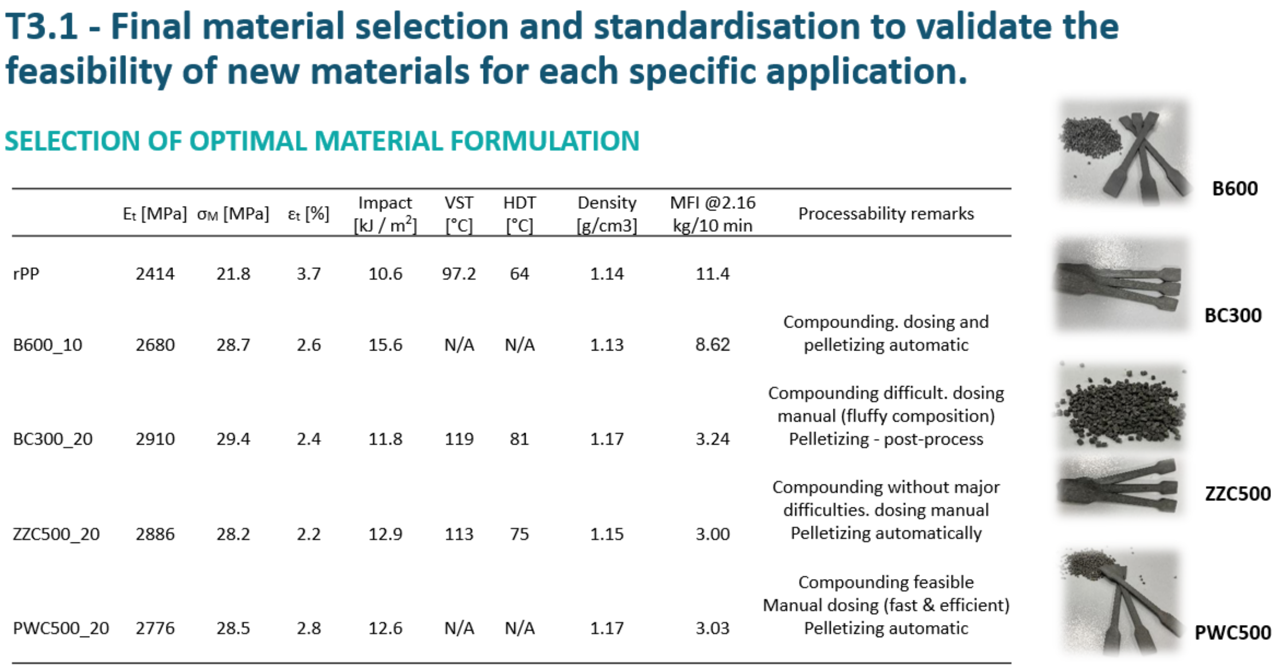
- Excellent processing stability
- Smooth surface finish
- Balanced mechanical and thermal performance

3.5.2 ZZC500-20

Recycled PP reinforced with 20% post-consumer ZZC500 cellulose fibres

- Strong environmental benefit (post-consumer source)
- Very good dimensional and thermal stability
- Highly favourable mechanical properties
- Efficient processing despite higher fibre loading

Both materials are already and will be further produced in significantly larger quantities for demonstrator trials (T3.3) on the new Gorenje INCIRCULAR recycling and compound extrusion line.



T3.1 - Final material selection and standardisation to validate the feasibility of new materials for each specific application.

SELECTION OF OPTIMAL MATERIAL FORMULATION

	E_t [MPa]	σ_M [MPa]	ε_t [%]	Impact [kJ / m^2]	VST [°C]	HDT [°C]	Density [g/cm3]	MFI @2.16 kg/10 min	Processability remarks
rPP	2414	21.8	3.7	10.6	97.2	64	1.14	11.4	
B600_10	2680	28.7	2.6	15.6	N/A	N/A	1.13	8.62	Compounding. dosing and pelletizing automatic
BC300_20	2910	29.4	2.4	11.8	119	81	1.17	3.24	Compounding difficult. dosing manual (fluffy composition) Pelletizing - post-process
ZZC500_20	2886	28.2	2.2	12.9	113	75	1.15	3.00	Compounding without major difficulties. dosing manual Pelletizing automatically
PWC500_20	2776	28.5	2.8	12.6	N/A	N/A	1.17	3.03	Compounding feasible Manual dosing (fast & efficient) Pelletizing automatic

Figure 8: *INCIRCULAR compound ready for GORENJE trials*

- The extrusion compounding process completed:

rPP	ZZCC500 - 20%	CA	TOTAL
38,5 kg	10 kg	1,5 kg	50 kg
77 kg	20 kg	3 kg	100 kg

⇨ 70 kg Dried & packed (@ TECOS)

rPP	B600 - 10%	CA	TOTAL
43,5 kg	5 kg	1,5 kg	50 kg
87 kg	10 kg	3 kg	100 kg

⇨ 30 kg Dried & packed (@ TECOS)

3.6 Recommendations for Future Research

To further advance composite development and support large-scale industrial implementation, future research should focus on:

- **Advanced compatibilizers and fibre surface treatments**

Exploring silane-based, enzymatic, or nano-modified surface treatments to improve adhesion and reduce moisture sensitivity.

- **Development of innovative fibre systems**

  Hybrid natural–synthetic fibres or recycled fibres with controlled morphology could offer improved mechanical performance with enhanced sustainability.

- **Rheology–process modelling**

  Using simulation to optimise:

  - screw configuration,
  - residence time,
  - thermal gradients,
  - fibre dispersion profiles.

- **Long-term durability studies**

  Including:

  - fatigue,
  - wear,
  - ageing under humidity and temperature cycling,
  - UV stability.

- **Sustainable production strategies**

  Investigating:

  - bio-based PP alternatives,
  - green additives,
  - energy-efficient process routes.

These directions will support the broader industrial adoption of fibre-reinforced recycled PP composites developed under INCIRCULAR.

4. INJECTION MOULDING PROCESS DEVELOPMENT

4.1 Injection Processing and First Validation of INCIRCULAR Demonstrators

The injection moulding validation of INCIRCULAR composite materials represents a crucial stage in confirming material processability under industrial conditions and assessing end-product performance. In alignment with Milestone 15 (First demonstrators produced and validated), the newly developed rPP–cellulose composites (ZZC500-20 and B600-10) were processed into full-scale modular storage boxes ("grmadnik") designed by Gorenje. This section summarises the injection moulding parameters, upscaling behaviour, structure–property relationships, dimensional stability characteristics, and processing efficiency.

The demonstrator was supported by complete CAD design, advanced simulation workflows (shrinkage, warpage, core deflection), and a smart moulding setup enabled by collaborations between Gorenje, TECOS, SISE, and SIMCON. These preparatory steps ensured a high degree of process robustness when transitioning to industrial production.

Figure 9: *INCIRCULAR injection line at GORENJE*

4.1.1 Injection Moulding Parameters

The injection trials were carried out on an **ENGEL DUO 1300-ton 8160 injection moulding machine**, which provided the necessary clamping force and injection capacity for the large-format storage box demonstrator.

Final validated processing parameters were:

- **Hydraulic injection pressure:** 20 bar
- **Cycle time:** 28 s
- **Cooling time:** 15.5 s
- **Plasticizing cushion:** 64.36 mm
- **Material:** INCIRCULAR composite (ZZC500-20 for main validation; B600-10 for comparative evaluation)
- **Feeding system:** Full thermal and runner system supported by rheological monitoring
- **Drying of pellets:** 24–48 h at 60 °C prior to injection

These parameters were optimized through iterative trials and confirmed to ensure stable melt flow behaviour, complete cavity fill, and controlled shrinkage, even with cellulose fibre reinforcements that increase melt viscosity.

Figure 10: *ENGEL DUO 1300-ton 8160 injection moulding machine*

Supportive technologies included:

- **Rheological nozzle (SISE)** for real-time viscosity monitoring
- **FEM-based mould and part simulations (SIMCON)**
- **Runner and venting system optimization (TECOS)**

This integrated approach ensured high process reliability and reduced the risk of defects such as incomplete filling, sink marks, fibre agglomerates, and local warpage.

4.1.2 Scaling Effects on Part Quality and Reproducibility

Upscaling from laboratory injection tests to full demonstrator production required addressing several scaling effects typical of fibre-reinforced recycled materials:

- **Melt rheology adjustments**

The higher shear stresses in industrial-scale injection units improved the dispersion of fibres and lowered the apparent viscosity of the rPP composites, positively affecting cavity filling.

- **Flow length and fibre alignment**

The storage box features long flow paths and thin-thick wall transitions, which can induce orientation effects:

- Fibre alignment was observed along primary flow directions, enhancing stiffness where needed.
- Local variations in fibre concentration were minimized through optimized runner balancing.

- **Reproducibility and cycle stability**

Across multiple cycles, the process demonstrated:

- stable pressure curves,
- repeatable cushion values,
- consistent demoulding behaviour,
- negligible burn marks or weld-line weaknesses.

This confirmed that the composite formulations were fully compatible with high-throughput industrial injection moulding.

4.1.3 Process–Structure–Property Relationships

The interactions between injection moulding conditions, fibre morphology, and the resulting material properties were carefully evaluated.

Key relationships observed:

- **Fibre Dispersion and Orientation**

Higher shear rates during injection improved fibre distribution and wetting, resulting in:

- enhanced tensile stiffness,
- improved impact resistance compared to unreinforced rPP,
- directional mechanical behaviour aligned with flow paths.

- **Cooling Rate and Crystallinity**

The moderate cooling time of **15.5 s** supported controlled crystallization, reducing internal stresses and improving dimensional stability of the large part.

- **Warpage Behaviour**

Residual warpage was successfully predicted and mitigated:

- TECOS simulations fed deformation maps into mould compensation adjustments.
- Optimized cooling channel layout and material card improved FEM accuracy.
- Final parts exhibited warpage levels within acceptable design tolerances.

These findings validate the strong synergy between CAE simulations, material characterization, and injection moulding practice.

4.1.4 Dimensional Stability and Shrinkage Control

Dimensional control was a major focus due to the large size and functional requirements of the modular storage box (stackability, fitment tolerances, geometric stability).

Tools and strategies applied:

- **Full 3D warpage and shrinkage simulation** (TECOS + SIMCON)
- **Mould geometry correction** using compensation factors
- **Fibre-orientation modelling** to predict anisotropic shrinkage
- **Monitoring of cooling asymmetry** across cavity surfaces

Results:

- Shrinkage remained stable and within predicted tolerance ranges.
- Warpage was significantly reduced after mould compensation adjustments.
- Structural stiffness was improved due to fibre reinforcement, preventing sagging in large flat surfaces.
- No significant post-processing deformation was observed after conditioning.

Thus, the INCIRCULAR composites demonstrated reliable dimensional performance for large technical applications.

4.1.5 Energy and Processing Efficiency Considerations

An important objective of the INCIRCULAR demonstrator validation was assessing processing efficiency, especially considering the sustainability goals of the project.

Key outcomes:

- **Short cycle time (28 s)**

Comparable to standard PP cycles despite fibre content, demonstrating excellent processability.

Low hydraulic pressure requirement (20 bar)

Indicates favourable melt flow and low resistance during cavity filling, contributing to:

- reduced energy consumption,
- lower mechanical wear of the injection unit.

- **Efficient cooling (15.5 s)**

Fibre-reinforced rPP dissipates heat faster than unfilled PP due to improved thermal conductivity, reducing overall cycle time.

- **Stable melt behaviour monitored by rheological nozzle**

Allowed fine-tuning of processing parameters, minimizing scrap rate and energy losses.

Together, these outcomes confirm that INCIRCULAR composites are not only technically feasible but also energetically efficient, supporting the broader objectives of resource-efficient and circular material solutions.

4.1.6 Conclusions

The injection moulding trials successfully demonstrated that the INCIRCULAR composite materials—particularly **ZZC500-20**, the chosen upscaled post-consumer fibre-reinforced formulation—can be processed into large structural components with high reproducibility and dimensional stability. The validated demonstrator (modular storage box) met all key functional requirements, confirming that the materials developed under WP3 are ready for continued industrial testing, full-scale validation, and replication within the INCIRCULAR ecosystem.

5. ADDITIONAL CHARACTERIZATION ANALYSES ON FINAL COMPOSITE FORMULATIONS NEEDED FOR SIMULATION MODELLING

5.1 Characterization Analyses done by UJA Team

5.1.1 INTRODUCTION

This technical report has been prepared within the framework of Task 3.1: Final Selection and Standardisation of Material, with the primary objective of validating feasibility and determining the optimal composite polymer formulation for its application in structural components. To this end, a comprehensive comparative characterisation of two key samples has been carried out: ZZC 500 20% and B600 10%, supplied by TECOS to the University of Jaén (UJA),

The characterisation focused on determining the essential thermodynamic and mechanical properties that govern the behaviour of the material during its processing and throughout its useful life:

- Pressure-Volume-Temperature (PVT) analysis: To establish the constitutive relationship between specific volume (v), temperature (T) and pressure (P). This information is critical for simulation and precise control of industrial processes such as injection moulding. The applicability of the Tait Model for predictive modelling was validated, observing that the ZZC 500 sample exhibits greater experimental consistency and a more defined phase transition.
- Dynamic Mechanical Analysis (DMA): To evaluate the viscoelastic response, determining the stiffness (Storage Modulus, E’) and the energy dissipation capacity (Damping Factor, Tan δ) as a function of temperature.

The consolidated results confirm that both samples are semi-crystalline polymers with a similar operating range (given the coincidence in glass transition temperature, $T_g \approx 33-35$ °C). However, the ZZC 500 20% formulation demonstrated a higher Storage Modulus across the entire temperature range, positioning it as the most suitable material for structural applications that require greater resistance to deformation under the load.

The report details the analytical procedure and the implications of these results, including the fundamental consideration of the anisotropy generated by the moulding process, which is essential for the final validation of the material.

The tests and results presented in the report were carried out by the research team participating in the project at the University of Jaén and were conducted in the research laboratories of the Linares Polytechnic School and the Central Research Support Services (SCAI) of the University of Jaén.

5.1.2 RESEARCH TEAM:

Carmen Martínez García: Professor
Mª Teresa Cotes Palomino: Senior Lecturer
Ana B. López García: Senior Lecturer
Fco. Javier Iglesias Godino: Senior Lecturer
Manuel Valverde Ibáñez: Senior Lecture

5.1.3 OBJECTIVES

The main objectives of this report, within the framework of Task 3.1 are:

1. Establish the Material's Constitutive Relationship: Accurately determine and model the Pressure-Volume-Temperature (PVT) relationship of the ZZC 500 20% and B600 10% formulations, using the Tait Model, to obtain essential processing parameters that ensure accurate industrial simulation.
2. Quantify the thermo-mechanical behaviour: Evaluate the stiffness (E') and energy dissipation capacity (Tan δ) of both samples as a function of temperature using Dynamic Mechanical Analysis (DMA), identifying their respective glass transition temperatures (Tg) and safe operating range.
3. Select the Optimal Structural Formulation: Compare the properties obtained for ZZC 500 20% and B600 10% with the requirements of the final application in order to select the formulation that maximises stiffness and dimensional stability, positioning it as the final candidate for the manufacture of structural components.
4. Identify Anisotropy Risks: Point out the implications of anisotropy induced by the moulding flow on the mechanical properties and recommend characterisation in the perpendicular direction to avoid overestimating rigidity and ensure the validity of the material for standardisation.

5.1.4 MATERIALS AND METHODS

The tests carried out by **UJA** on samples well as the methodology used are shown in the following table:

Table 1. Samples characterization tests and standards

TEST	
TEST	**Technical/Equipment**
PRESSURE-VOLUME-TEMPERATURE ANALYSIS (PVT)	PVT 100 (Haake)
DYNAMIC MECHANICAL ANALYSIS (DMA)	TA Instrument DMA equipment, utilising film tension geometry

5.1.5 Pressure-volume-temperature analysis (PVT)

The PVT diagram was obtained in isobaric cooling mode at 5ºC/min in the temperature range from 250ºC to 50ºC and at different pressures (5 pressures: 200, 400, 800, 1200, 1600 bar). The data will be adjusted to the IKV and Tait equations.

A PVT 100 (Haake) device was used. The PVT diagram was obtained using the isobaric cooling mode in the pressure range from 200 to 1600 bar at a cooling rate of 5°C/min.

The specific volume at atmospheric pressure is calculated by extrapolation using the Tait equation included in the equipment software.

Figure 1. Samples analysed

The margin of error in the measurement for the parameters of this test is as follows:

Specific volume < 0.001 cm^3/g

Pressure: ±1% of actual pressure

Temperature: ±0.3% °C

Length: ±1 mm

5.1.6 Dynamic mechanical analysis (DMA)

The tests were carried out on TA Instrument DMA equipment, using the tension film geometry.

The test pieces were tested on this equipment in a direction parallel to the flow.

Figure 2. DMA de TA Instrument

5.1.7 RESULTS AND DISCUSSION

The following are the results obtained from the different tests carried out on the 2 samples received and described in the previous section.

❖ PRESSURE-VOLUME-TEMPERATURE ANALYSIS (PVT)

The results obtained are shown in the **PVT graph** (Specific Volume versus Temperature and Pressure). These results are crucial because they illustrate the constitutive relationship between the specific volume of the material (V), the temperature (T) and the pressure (P).

The graphs show the cooling curves at constant pressures (isobars), from 1 bar to 1600 bar, for two samples: ZZC 500 20% and B600 10%. The figures a) shows the corresponding average data and calculated deviations. The data at a pressure of 1 bar are extrapolated using Tait's equation. The figures b) shows the adjust to the Tait´s equation.

The Tait equation is described in terms of the state of the material: solid, molten, and transition zone.

$$\boxed{V(T,P) = V_0(T)\left[1 - C\ln\left(1 + \frac{P}{B(T)}\right)\right] + V_t(T,P)}$$

Description of the coefficients used: V (T,P) is the specific volume; V_0 is the specific volume at pressure 0; C is a universal constant such that C= 0.0894 and B accounts for the effect of pressure.

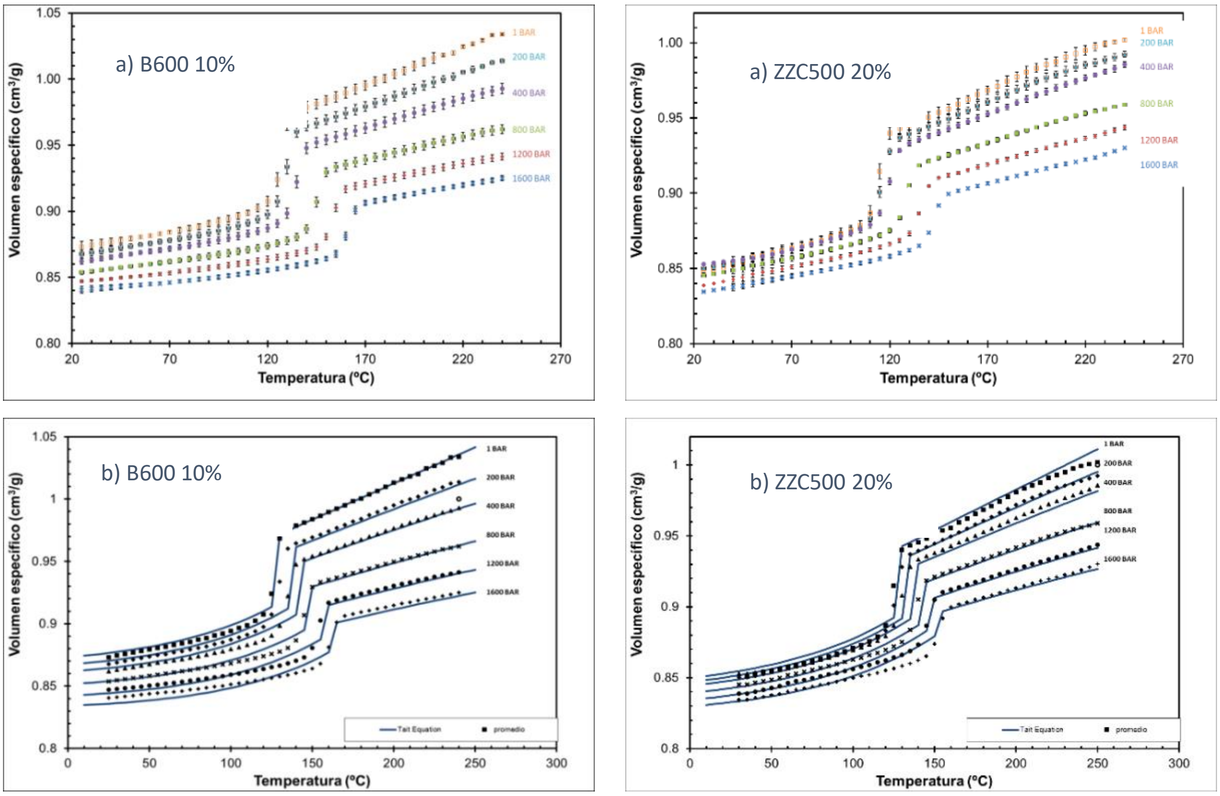


Figure 3. a) PVT diagrams. b) Adjust Tait´s equation. The experimental data have been determined at pressures of 200, 400, 800, 1200, and 1600 in the temperature range from T=250ºC to T=50ºC.

We observe several key points:

1. The specific volume decreases as the temperature decreases and the pressure increases. This confirms that both samples are compressible, a vital factor to consider in processes such as injection moulding or pressing.

2. The glass/crystalline transition zone shifts to higher temperatures as the pressure applied during moulding increases (800–1600 bar).

3. Both materials, as semi-crystalline polymers, show a sharp and marked change in specific volume around 130 °C. This is the melting temperature, and its existence implies that both materials will experience significant shrinkage upon solidification.

4. Below the melting temperature, both materials exhibit low compressibility, ensuring good dimensional stability once solidified.

Also, it is observed that the adjusted TAIT model correctly describes the PVT behaviour of both materials across the entire range of temperatures and pressures. This correlation is essential, as it allows for predictive modelling and accurate simulation of industrial processes.

The following table summarizes the key differences identified when comparing the two formulations, **B600 10 %** and **ZZC 500 20 %**.

Table 2. Key differences **B600 10 %** and **ZZC 500 20 %**.

CHARACTERISTIC	B600 10% (DISCONTINUOUS LINES)	ZZC 500 20% (CONTINUOUS LINES)	IMPLICATION
COMPRESSIBILITY IN MOLTEN PHASE		More pronounced (greater slope).	B600 is more pressure-sensitive in the molten state, requiring greater process adjustment.
PHASE CHANGE SHAPE	Slightly more rounded.	More abrupt and symmetrical.	ZZC 500 shows a sharper transition, useful for applications requiring thermal precision.
EXPERIMENTAL CONSISTENCY	Experimental curves show greater dispersion.	Experimental curves are more compact and well-aligned.	ZZC 500 exhibits lower experimental variability, suggesting greater homogeneity in its behaviour.

In summary, although the TAIT model is valid for both, the ZZC 500 demonstrates greater consistency and a more defined phase transition, which is advantageous for simulation and strict control of moulding parameters.

❖ *DYNAMIC MECHANICAL ANALYSIS (DMA)*

Two tests were conducted for each sample. The following figures present the temperature sweep curves, with the corresponding thermal transitions clearly identified.

The following graphs show the results obtained from the **viscoelastic characterization by Dynamic Mechanical Analysis (DMA)^(*)**. This technique is essential for determining key properties such as the **stiffness** and **energy dissipation capacity** of the material as a function of temperature.

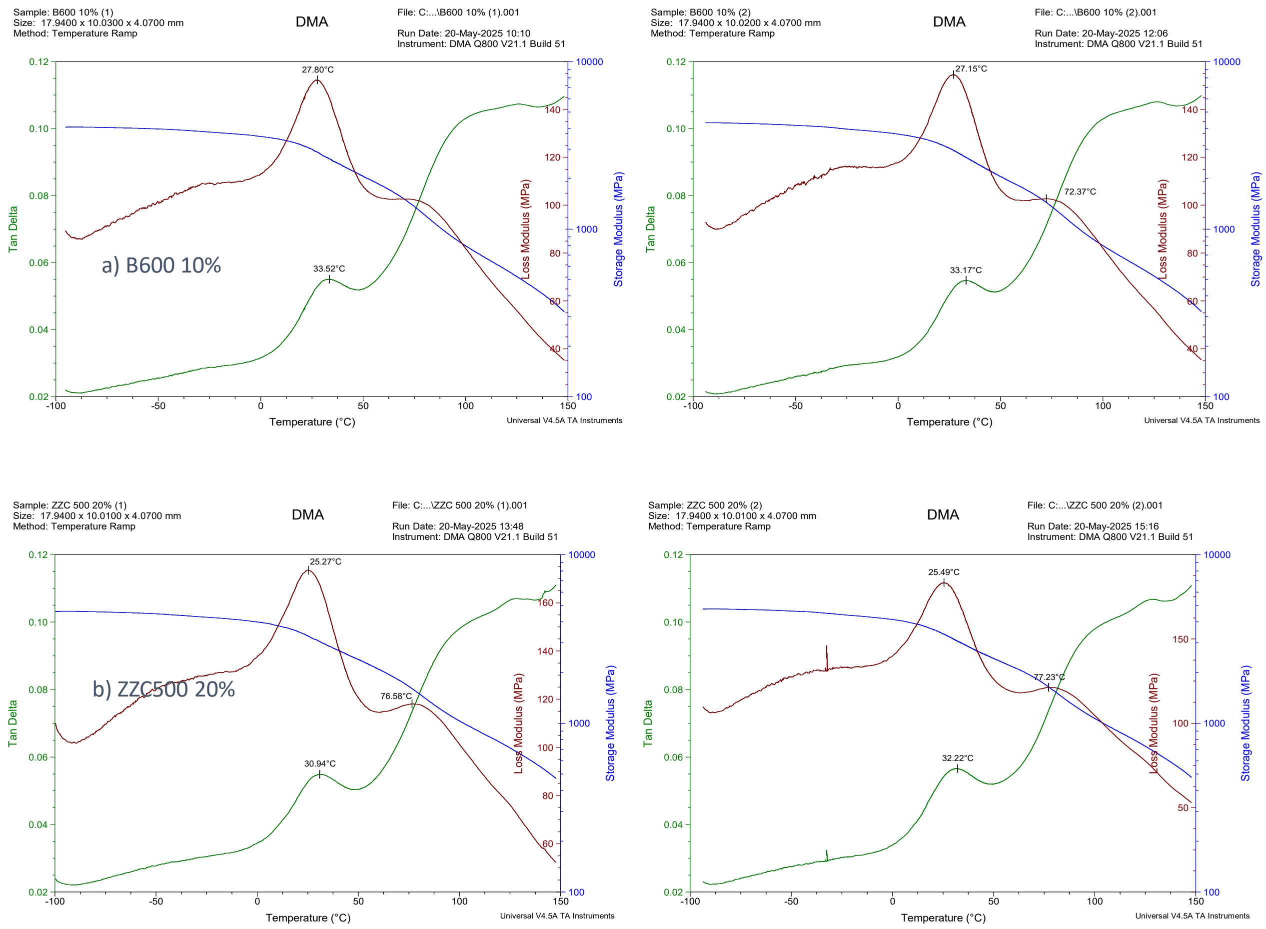


Figure 4. DMA analyses a) B600 10%; b) ZZC500 20%

Although both samples exhibit a **similar glass transition temperature (Tg)**, as shown in the table 3, the **ZZC 500 20%** formulation displays a **significantly higher Storage Modulus (E')**, indicating greater stiffness. Therefore, **ZZC 500 20%** can be considered the **more structurally robust material**.

Table 3. Comparison of samples **B600 10 %** and **ZZC 500 20 %**.

CHARACTERISTIC	B600	ZZC 500	COMPARISON
STORAGE MODULUS E'	Lower (range: 100–4200 MPa)	Highest (range: 150–4600 MPa)	ZZC 500 exhibits greater rigidity
TAN Δ (DAMPING)	Peak around 30–35 °C	Similar peak, slightly higher	**Similar viscoelastic properties**
GLASS TRANSITION T_G	~33 °C (Tan δ peak)	~33–35 °C (Tan δ peak)	Coincide, **same operating range**

(*) It is important to note that the **test specimens were analyzed in the direction parallel to the flow**, but it would be advisable to also perform tests **in the perpendicular direction**.

5.1.8 CONCLUSIONS

Based on the results of the characterization tests, we can conclude that:

1. **ZZC 500 is Structurally Superior (DMA).** The **ZZC 500 20%** formulation is considered the **more structural material**. This is because it exhibits a **significantly higher Storage Modulus** than B600 10% based on Dynamic Mechanical Analysis (DMA), which indicates **greater stiffness**.
2. **ZZC 500 Offers Greater Consistency and Process Control (PVT).** The **ZZC 500 20%** material demonstrates **greater consistency** and a **more defined phase transition**. Its experimental PVT curves are **more compact and well-aligned** (lower dispersion), which is advantageous for **simulation** and the **strict control of molding parameters**.
3. **Both Materials Require Management of Shrinkage and Pressure:** Both formulations are **semi-crystalline polymers** that exhibit a **sharp, marked change in specific volume** around 130 ºC (the melting temperature). This implies that both will undergo **significant shrinkage** upon solidification, and both are **compressible**, a vital factor for molding processes.
4. **B600 is More Pressure-Sensitive in the Molten Phase.** When comparing compressibility in the molten phase, **B600 10%** shows a **more pronounced sensitivity** to pressure (greater slope in the PVT isobars). This means B600 is **more pressure-sensitive in the molten state**, requiring **greater process adjustment**.

5. There is a **risk that the stiffness** may be **overestimated** and the energy dissipation capacity underestimated. This is because the DMA test specimens **were only analyzed in the direction parallel to the flow**, which tends to align the fibers and enhance the apparent stiffness.

5.1.9 REFERENCES

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5.2 Characterization Analyses done by ECO-CASTULUM

The aim of the report is to provide traceability of the capillary analysis laboratory work carried out by the company **ECOCASTULUM, S.L.** (hereinafter **ECOCASTULUM**) for the performance of characterization tests of two (2) samples called "RH-0.17%" and "RH-0.15%", facilitated by **TECOS** to **ECOCASTULUM**.

5.2.1 TECHNICAL STAFF

The following **ECOCASTULUM** team participated in the development of this experimental program that covers a laboratory testing phase for physical and chemical characterization tests and interpretation of the results:

Laboratory team:

- Elena Picazo Camilo – Chemical Engineering Department Manager at Ecocastulum. Industrial Chemical Engineer, MsC in Advanced Materials Engineering and pH. D student in Advanced Materials Engineering.

Data interpretation team:

- Juan Isidro Díaz García – Environmental Sustainability Advisor al Ecocastulum. Material Engineering, MsC in Materials Engineering and pH. D student in Research, Modelling and Analysis of Risk in the Environment.

Technical support team:

- Professor Francisco A. Corpas Iglesias - CTO (Chief Technology Officer) and Professor of Materials Engineering at the University of Jaén (Spain)

The Project Management Team:

- Mr. Ignacio López Anquela. CEO (Chief Executive Officer) of Ecocastulum
- Mr. Raúl Carrillo Beltrán - CRO (Chief Research Officer) at Ecocastulum

5.2.2 MATERIALS

TECOS supplied a total of two (2) samples: "RH-0.17%" and "RH-0.15%".

The composition of the samples is described below:

- Sample " RH-0.15%": rPP (Tac+$CaCO_3$)+ZZC500-20 wt.% (Figure 1a)
  AIMPLAS CODE: 25-1230-1

- Sample "RH-0.17%": rPP (Tac+$CaCO_3$)+B600-10 wt.% (Figure 1b)

  AIMPLAS CODE: 25-1230-2

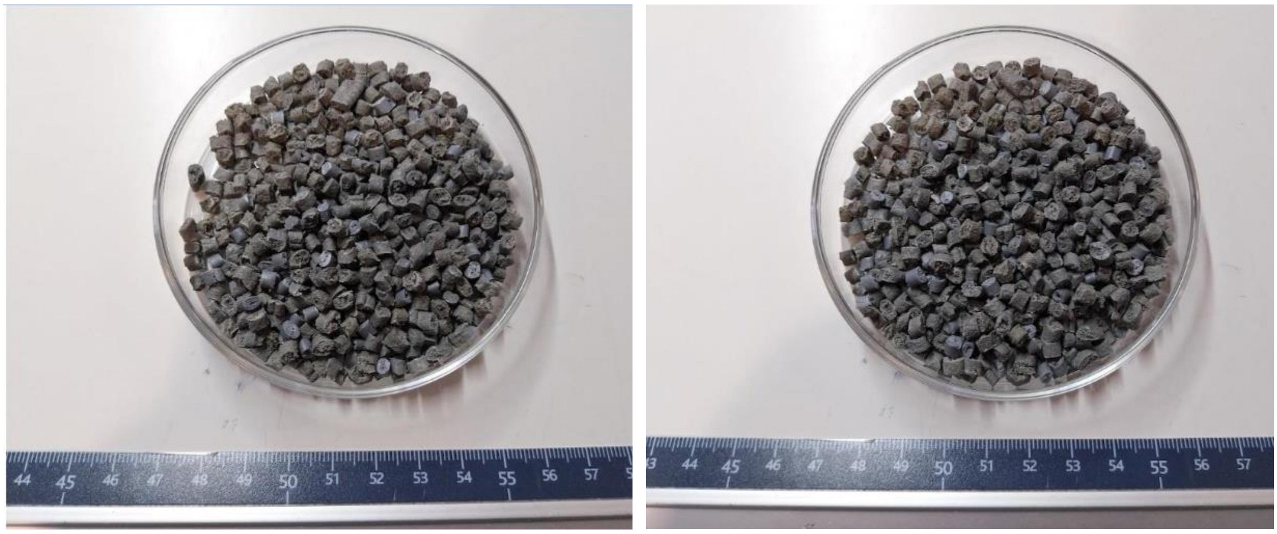

Figure 1. Samples: (a) RH-0.15% (25-1230-1) and (b) RH-0.17% (25-1230-2)

5.2.3 METODOLOGY

The aim of the applied methodology is the realization of a capillary analysis of samples ("RH-0.17%" and "RH-0.15%"). The determination of capillary analysis in the Göttfert Rheograph 25 Capillary Rheometer is performed under the test conditions described in Table 1.

	CAPILLARY ANALYSIS TEST
Equipment	Capillary Rheometer Göttfert Rheograph 25
Diameter of cylinder	15 mm
L/D	10/1; 30/1
Temperature	180°C
Shear rate	10 s^{-1} -10000 s^{-1}

Table 1. Test conditions

The tests have been carried out in the laboratories of AIMPLAS in Valencia. The interpretation has been carried out by ECOCASTULUM team and have been supervised by Professor Francisco Antonio Corpas Iglesias who has the following training: Dr. Mining and Energy Engineer; Chemical Engineer; Metallurgical Engineer and Industrial Engineer; Materials and metallurgical Expert, with different patents granted, numerous business and research projects and participation in books and countless publications.

5.2.4 RESULTS ANALYSIS

❖ *SAMPLE RH-0.15%*

Capillary analysis is used to determine the rheometric behavior of a thermoplastic. The results obtained, reported as mean and corrected values, are shown below. Bagley correction was applied to obtain the results of actual shear stress and actual shear velocities from the experimental results obtained from capillary theology. Experimental data under flow instability were omitted from the Bagley correction. Table 2 shows the corrected viscosity results at 180ºC.

Table 2. Viscosity - Experimental results corrected at 180°C

Shear rate (γ) s^{-1}	**Viscosity (η) Pa·s**
2.91E+01	2.25E+03
7.54E+01	1.25E+03
1.52E+02	7.83E+02
3.04E+02	4.90E+02
6.86E+02	2.35E+02
1.55E+03	1.67E+02
3.13E+03	1.06E+02
7.65E+03	5.57E+01

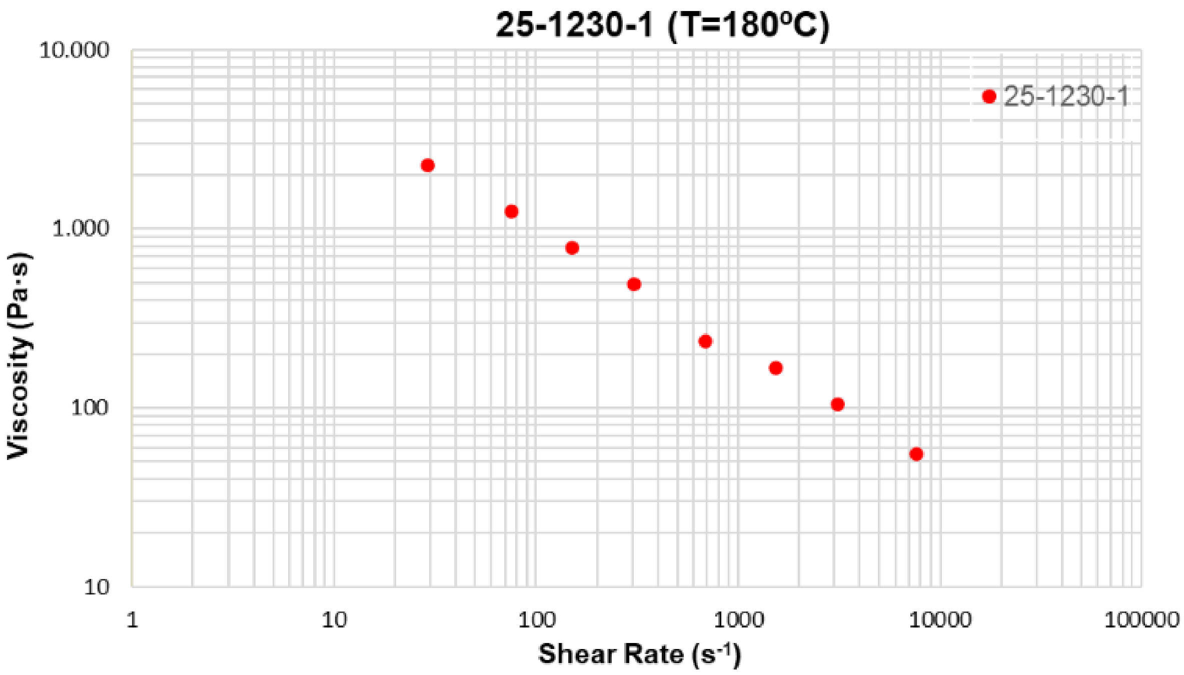

Figure 2. Viscosity curve of sample 25-1230-1 "RH-0.15%" at 180ºC

- *SAMPLE RH-0.17%*

Capillary analysis is used to determine the rheometric behavior of a thermoplastic. The results obtained, reported as mean and corrected values, are shown below. Bagley correction was applied to obtain the results of actual shear stress and actual shear velocities from the experimental results obtained from capillary theology. Experimental data under flow instability were omitted from the Bagley correction. Table 3 shows the corrected viscosity results at 180ºC.

Table 3. Viscosity - Experimental results corrected at 180ºC

Shear rate (γ) s^{-1}	Viscosity (η) Pa·s
1.281E+01	2.25E+03
2.71E+01	1.25E+03
7.08E+02	7.83E+02
1.43E+02	4.90E+02
2.85E+02	2.35E+02
7.19E+03	1.67E+02
1.44E+03	1.06E+02
2.89E+03	5.57E+01
7.30 E+03	5.51E+01
1.20 E+04	4.16E+01

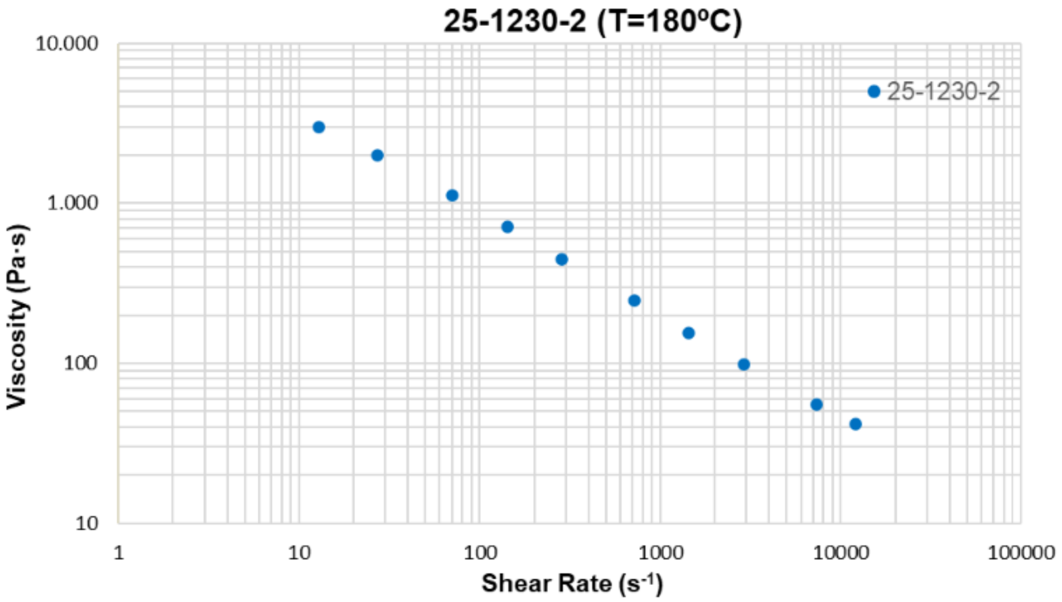

Figure 3. Viscosity curve of sample 25-1230-2 "RH-0.17%" at 180ºC

5.2.5 CONCLUSIONS

After performing capillary rheology tests on samples 25-1230-1 and 25-1230-2 at 180°C, it can be concluded that the two samples have similar behavior in viscosity values in the shear rate range 10 s-1 and 10000 s-1. Figure 4 reports the superposition of the two samples.

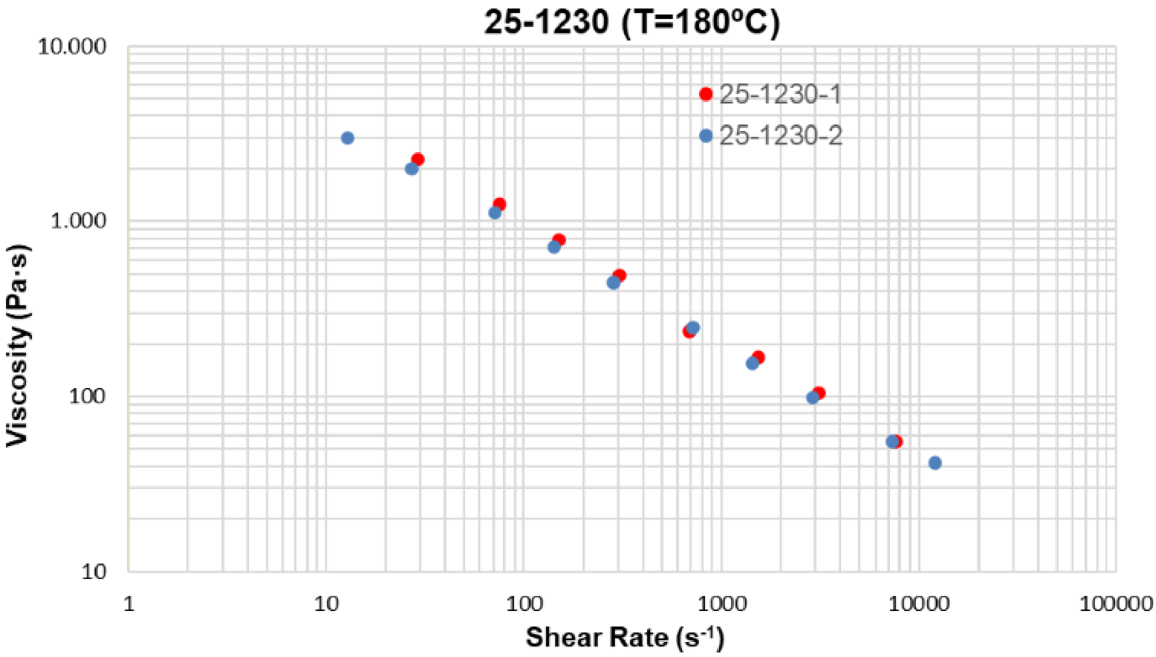

Figure 4. Overlapped shear viscosity curves of the samples 25-1230-1 and 25-1230-2 at 180ºC