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Challenges, trends and solutions in developing and processing of recycled polymer materials

BOOK OF ABSTRACT

Slovenj Gradec | 21-22 september 2023

FTPO
Faculty of Polymer Technology

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EDITORS
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September 2023

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Foreword to
1st IPPT_TWINN Conference

CHALLENGES, TRENDS AND SOLUTIONS IN DEVELOPING AND PROCESSING OF RECYCLED POLYMER MATERIALS

Dear reader,

The EU-funded project IPPT_TWINN Reinforcing the scientific excellence and innovation capacity in polymer processing technologies of the Faculty of Polymer Technology (FTPO) is one of the most important projects for the FTPO. The main objective of the project is to increase the knowledge of polymer processing and strengthen the cooperation between FTPO and partner institutions, as well as to improve/enhance the overall research excellence and innovation capacity on innovative polymer processing technologies at FTPO.

Recycling of polymer materials is one of them. Since we know that the amount of polymer materials processed will double by 2030 (up to 650 million tonnes per year), recycling and reuse of polymer materials are and will be of utmost importance. So it is no surprise that the 1st conference under the IPPT_TWINN project is devoted to recycling technologies.

We have succeeded in addressing the most important issues in the field of recycling technologies and attracting the key players in the world of recycling. The conference will therefore be an excellent opportunity to present and discuss the outstanding issues in recycling and to develop new solutions in this exciting field. The conference is divided into two parts. The first part is dedicated to the presentation of current material developments, industry case studies and the latest R&D results in the packaging, automotive, electrical and household appliance sectors.

This industry-oriented part will be enhanced by the panel discussion, which will allow a guided discussion between the participants. B2B meetings and training for young researchers on research funding opportunities will be organised to foster collaboration and cohesion among participants.

The second part of the conference will present the scientific results of recycling technologies, supported by 3 plenaries and 4 invited speakers, as well as 9 selected paper presentations. The added value of the conference is also the fact that all accepted papers not presented on the second day will be available as recordings.

The strong international participation (9 different European countries) in the conference is proof of its relevance and this is also the guarantee of its success.

Therefore, I would like to take this opportunity to thank all the contributors to the conference, the conference participants and especially the organising team who put a lot of effort into the preparation of this important and solid conference.

Last but not least, I would like to thank the European Commission for funding the IPPT_TWINN project. Without their support, this conference would not have happened.

Assoc. Prof. Dr. Blaž Nardin
Dean of the Faculty of Polymer Technology
Coordinator of IPPT_TWINN project

Scientific programme

FIRST DAY – 21st September 2023

8:30 - 9:00	Registration
9:00 - 9:15	Opening speech, Blaž Nardin - project leader and Dean of FTPO
9:15 - 10:45	**Plenary speakers** (4 × 15 minutes + 5 min questions) • **Plastics in circular economy – requirements and challenges** Daniel Schwendemann, IWK, Switzerland • **Recycling of plastic packaging waste - from product specifications to high-performance recyclates,** Joerg Fischer, JKU Linz, Austria • **State-of-the art and challenges at thermoset composite recycling** Marcello Caledani, Polytechnico Milano, Italy • **Industry-focused developments to introduce recycling and sustainability in plastic and composite materials,** Sonia Garcia, Technalia, Spain
10:45 - 11:15	**Recycled materials for packaging applications** (2 × 10 minutes + 5 min questions) • **Recycling of multilayer packaging film,** Michael Feuchter, University of Leoben, Austria • **Recycling packaging strategy of a chain of grocery stores** Samo Pergar, Lidl Slovenia d.o.o. k.d. Slovenia (on-line)
11:15 - 11:50	Coffee break: 20 min
11:45 - 12:15	**Recycled materials for automotive applications** (2 × 10 minutes + 5 min questions) • **From post-consumer polymer waste to technical parts,** Curdin Wick, IWK, Switzerland • **Recycled materials and the Automotive industry,** Aleš Adamlje, Hella Saturnus, Slovenia (on-line)
12:15 – 12:45	**Recycled materials for electro applications and home appliances (2 × 10 minutes + 5 min questions)** • **Recycling of plastics from WEEE,** Gunther Hoggerl, MGG, Austria • **Challenges and possible solutions for recycling of WEEE products** Marton Bredacs , Polymer Competence Center Leoben, Austria
12:45 - 13:30	**Recycling of cross-linked materials (thermosets and elastomers)** (3 × 10 minutes + 5 min questions) • **Strategies to enable the recyclability of thermoset composite materials,** Julio Vidal, AITIIP, Spain • **Recycling opportunities for vulcanized rubber goods,** Tamás BÁRÁNY, BME, Hungary • **Thermomechanical devulcanization of rubbers,** Zoltán Istán HALÁSZ-KUTAS, BME, Hungary
13:30 - 14:15	Lunch Break
14:15 – 15:15	**Panel discussion** • **Daniel Schwendemann,** IWK, Switzerland • **Joerg Fischer,** JKU Linz, Austria • **Julio Vidal,** AITIIP, Spain • **Marcello Colledani,** Italy • **Marton Bredacs,** Austria • **Sonia Garcia,** Tecnalia, Spain • **Blaž Nardin,** FTPO, Slovenia
15:15 – 15:30	Closing remarks of the 1st day

→

15:30 – 18:30	**B2B Meetings and FTPO lab tour**
15:30 – 18:30	**Open training on research funding opportunities,** AITIIP, Spain
19:00 - 21:00	**Dinner** at »Hiša Ančka«

SECOND DAY – 22nd September 2023

8:30 - 8:55	**Registration**
8:55 - 9:00	**Recal Day 1, Blaž Nardin** - Project leader and Dean of FTPO
9:00 - 9:30	INTRODUCTORY LECTURE **Myths and facts about the recyclability challenges of post-consumer lightweight plastics packag** Manica Ulčnik Krump, Interzero Plastics Innovations, Interzero Holding GmbH & Co., Germany
9:30 - 10:15	PLENARY LECTURE **A Journey from Processing to Recycling of Multilayer Waste Films:** **Main Challenges and Prospects with innovative approaches** Khalid Lamnawar, INSA Lyon, France
10:15 - 10:50	PLENARY LECTURE **Sustainability and chemical recycling of polymers** Christian Paulik, Johannes Kepler University in Linz, Institute for Chemical Technology of Organic Materials, Austria (on-line)
10:50 - 11:10	Coffee break: 20 min
11:10 – 11:55	PLENARY LECTURE **Chemical Technologies for Improved Circularity of Thermoset Polymer Composites** Stefano Turri, Polytechnico Milano, Italy
11:55 - 12:25	INVITED LECTURE **Chemical Recycling of Polymers** Ema Žagar, National Institute of Chemistry, Slovenia
12:25 - 12:45	INVITED LECTURE **Recycling of polyurethane thermosets – a recurring challenge for over 50 years** Hynek Beneš, Institute of Macromolecular Chemistry of the Czech Academy of Sciences, Czech Republik (on-line)
12:45 – 13:30	Lunch Break
13:30 – 14:50	INVITED LECTURE **Recycling of Polyamide 11 and 12 for 3D printing applications** Soeren Griessbach, GS-PRO GmbH, Germany LECTURE **Bio-based polyurethane foams with enhanced recyclability** Olga Gotkiewicz, Institute of Macromolecular Chemistry of the Czech Academy of Sciences, Czech Republik

→

13:30 – 14:50	LECTURE **Improving the quality of polypropylene recyclates** Jutta Geier, Polymer Competence Center Leoben, Austria LECTURE **Long-term properties of PP recyclate blends** Jessica Hinczica, Polymer Competence Center Leoben, Austria LECTURE **Development of reusable adhesives systems for polymer bonding applications** Daniel Bautista, Polymer Competence Center Leoben, Austria LECTURE **Effect of recycling on the properties of glass fiber reinforced injection molded polymer products** Maja Csapo, BME, Hungary LECTURE **Development of polymerization of T-RTM technology using 3D printing** Zsófia Luca Csepel, BME, Hungary LECTURE **Fourier-Transform Infrared Microscopy Analysis of recycled copolymer materials** Alen Erjavec, University of Maribor, Faculty of Mechanical Engineering, Slovenia LECTURE **Characterisation of the chemical composition and thermal properties of SARS-CoV-2 rapid antigen tests cassettes present in Slovenia** Janez Slapnik, Faculty of Polymer Technology, Slovenia LECTURE **Mechanical Recycling of Carbon Fibre Reinforced PA66** Rebeka Lorber, Faculty of Polymer Technology, Slovenia
14:50 – 15:00	Closing remarks of the 1st IPPT_TWINN conference

RECORDED LECTURES (link sent to all participants of the conference)

- **Tunning the surface properties of recycled thermoplastic composites by the injection moulding process parameters: Atomic Force Microscopy study**
  Klementina Pušnik Črešnar, Faculty of Polymer Technology, Slovenia

- **On-Demand Activation of Dynamic Bond Exchange in Covalently Crosslinked Polymers**
  David Reisinger, Polymer Competence Center Leoben, Austria

- **Influence of Compatibilizer on Viscoelastic Properties of the Thermoplastic Polymer-Based Biocomposites**
  Silvester Bolka, Faculty of Polymer Technology, Slovenia

- **Stiffness and Strength Steering of Recycled Polypropylene (rPP) with Addition of Waste Paper**
  Silvester Bolka, Faculty of Polymer Technology, Slovenia

- **Use of Recycled Materials Based on PE in Rotational Moulding Technology**
  Blaž Nardin, Faculty of Polymer Technology, Slovenia

- **Use of sustainable polyolefins for injection moulded closure packaging**
  Blaž Nardin, Faculty of Polymer Technology, Slovenia

- **Neoteric Solvolysis Methods and how are they being developed**
  Carlos Hornero Montalvo, Moses Productos S.L., Spain

- **Recycling of PET via aminolysis**
  Irena Pulko, Faculty of Polymer Technology, Slovenia

- **Novel Eugenol Based Acrylate Oligomers: Their Photochemical Curing Behavior and Mechanical Properties with Reproducity**
  Zhejiang University, College of Chemical and Biological Engineering, China

Chemical Technologies for Improved Circularity of Thermoset Polymer Composites

Stefano Turri*, Oussama Boumezgane, Gianmarco E. Griffini, Eleonora Manarin

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Polymer composites offer many technical advantages over other competing manufacturing materials, but the management of composite products at their end of life is very challenging, especially when thermoset fiber reinforced materials are concerned [1]. Most of composites are still landfilled – where not banned – or incinerated, practices clearly not compliant with the principles of circular economy. Glass fiber reinforced polymers (GFRP) represent by far the largest portion of the market, due to their lower cost. On the other side, carbon fiber reinforced polymers (CFRP) are still a niche in the market, but their recovery at the end of life is more economically appealing. The reference (TRL 8-9) recycling technologies for these materials are mechanical shredding for GFRP and pyrolysis for CFRP [2]. The former method however doesn't allow a true liberation of fiber from the matrix residue and it is essentially a downcycling, while in the latter the organic phase is generally lost or burnt.

Recently, smart dismantling coupled with chemical recycling (chemcycling) by solvolysis have gained a renewed interest as an integrated circular technology suitable for the current generation of composite materials, since it can be able to recover and separate well cleaned and undamaged fibers with a reusable, depolymerized resin fraction [1,3]. A careful selection of solvent media and catalyst may help to optimize the process conditions with minimization of capital cost investments, typically quite high for solvolysis plants. For the next generation of sustainable composite materials, the definition of a roadmap towards the development of new thermoset resins *recyclable by design* is needed to improve the overall circularity of the sector. Key steps are a smart chemical design of resin structures, the use of biobased building blocks, the replacement of reactive diluents, the incorporation of latent depolymerization catalysts. Specific examples are represented by the so-called Covalent Adaptable Networks (CANs), either dissociative or associative, that in principle may allow for both an extended product lifetime through repair strategies, as well as an easier chemcycling at their end of life [4,5].

Keywords: thermosets; polymer composites; recycling; covalent adaptable networks; circular economy

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A Journey from Processing to Recycling of Multilayer Waste Films: Main Challenges and Prospects with innovative approaches

Khalid Lamnawar,
INSA Lyon, France

In a circular economy context with the dual problems of depletion of natural resources and the environmental impact of a growing volume of wastes, it is of great importance to focus on the recycling process of multilayered plastic films. This talk is dedicated first to the general concepts and summary of plastic waste management in general, making emphasis on the multilayer films recycling process. Then, in the second part, the focus is dealing with multilayer films manufacturing process, including the most common materials used for agricultural applications, their processing, and the challenges of their recycling, recyclability, and reuse. Hitherto, some prospects are discussed from eco-design to mechanical or chemical recycling approaches.

In the context of a circular economy, the first illustrations in in this plenary lecture were devoted to the study of the processing ability of recycled flexible films from agricultural bale wrapping film's waste. These multilayer films initially contain an additive that gives the final product a sticky character. During recycling, this additive is still present and migrates to the surface. A particular emphasis has been placed on understanding and studying the migration mechanisms of this additive according to the molecular architecture of the polyethylenes used with different branching content. Model blends, with or without mineral fillers, were prepared for this purpose. In addition to morphological and microstructural studies, original experimental methodologies have been set up to study the migration/diffusion kinetics with tribo-rheometry and film surface's "tack" monitoring devices. As for the rheological behavior in shear and elongation, it is influenced by the presence of this additive. The second part of this study was devoted to understanding the realities of the recycling activity of other polypropylene- and polyethylene-based multilayer films. Given the complexity of these materials, we chose to study equivalent model blends by combining them with physical compatibilizers. The influence of the latter on the rheological, morphological and mechanical properties was then evaluated. This study was then transposed to post-consumption multiphase systems.

The last studies were devoted to a future-oriented approach. This involves going from eco-design to the development of easily recyclable multi-micro/nanolayers. The originality of this study consists in limiting the number of constituents, reducing/controlling the thickness of the layers and avoiding the use of tie-layers. Finally, their rheological, morphological and mechanical properties have been evaluated according to number and type of mechanical recycling cycles.

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Sustainability and Chemical Recycling of Polymers

Christian Paulik[a,b]

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Without doubt, we are living in the "plastic age" producing around 380 million tons/yr on a global scale. Plastics are a fantastic material, finding applications in all areas of our modern live from healthcare to packaging. However, the flexibility in tailoring macromolecules for each specific application also makes a reuse difficult. Thus, only 5% of the municipal plastic waste in the US are recycled [1], meaning the vast majority is incinerated or ending in landfills. The recovery of resources (recycling) on the other hand is gaining increasing attention. Guidelines, strategies and framework conditions need to be created to accelerate the development towards a closed material cycle.

The paper discusses different aspects in chemical recycling of plastics from the pre-treatment (washing) of the plastic waste flakes [2] to contamination effects of derived feed streams in downstream polymerization processes [3].

Keywords: chemical recycling, polyolefins, polymerization, catalyst poisoning.

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Industry-focused developments to introduce recycling and sustainability in plastic and composite materials

Sonia García-Arrieta[1], Amaia de la Calle[1], Isabel Harysmendi[1], Oihane Echeverria-Altuna[1], Luis Palenzuela[1], Raquel Rodriguez[1], Olatz Ollo[1], Damien Salle[1], Cristina Elizetxea[1]

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Nowadays, polymeric materials (plastics, resins, and composites) are widely used in millions of industrial applications. During the last 50 years, research in polymeric materials has been directed by the requirements demanded by the industry, which mainly are to increase properties and/or to reduce cost. However, the current society, governments and our planet require us to redirect our research towards sustainability, recyclability and circular economy. Millions of tons of polymeric waste are in our landfills, floating in our seas or burning without any treatment.

It is the duty of researchers to work hard to change the world and propose sustainable polymeric processes and materials to the current industry. Tecnalia has been (and continues being) very active in achieving these sustainability objectives that are so highly demanded by society.

Tecnalia participates in regional and European projects in this line, whose objective is to involve the industry as main actor in the need to implement sustainable processes and materials and integrate circular economy. A review of objectives and main result will be presented for next projects:

Regional projects:

- Avansite: New generation of sustainable composites for advanced manufacturing.
- Neoplast: New raw materials from technologies and processes of chemical recycling of plastics for the industrial sectors of the Basque Country in an applied approach to the circular economy
- Ze-Konp: Sustainable and intelligent manufacturing processes of thermoplastic composites of new generation
- Biopolyurethanes: Development of biobased polyurethanes and polyurethanes with dynamic bonds

European Project:

- FiberEuse: Large-scale demonstration of new circular economy value chains based on the reuse of end-of-life fiber-reinforced composites
- Digiprime: Digital platform for circular economy in cross-sectoral sustainable value networks
- Maelstrom: Smart technology for Marine Litter Sustainable Removal and Management
- DeremCo De & Remanufacturing for Circular Economy Investments in the Composite Industry
- EverPV: Highly efficient delamination technologies to recover and reuse metals, glass, polymers from end-of-life photovoltaic panels

Keywords: circular economy, sustainability, plastic recycling, composite recycling, remanufacturing.

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Chemical Recycling of Polymers

Ema Žagar

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Chemical recycling of polymers containing chemically labile bonds in the backbone, and especially those that are difficult to reprocess or have a cross-linked structure, offers an alternative for obtaining high quality secondary raw materials suitable for the production of the same or other polymer materials. Here we discuss the possibilities of microwave-assisted chemical recycling of high-performance polyamides and their composites [1,2] and of polyurethane foams with a cross-linked structure [3–5].

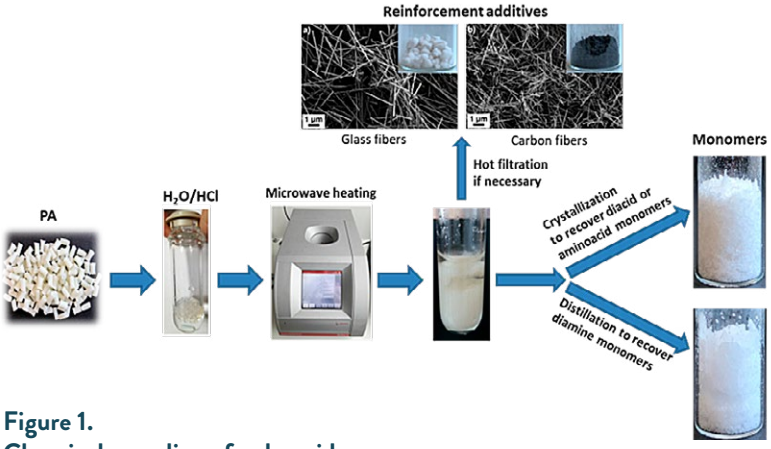

Figure 1.
Chemical recycling of polyamides a nd PA-based composites.

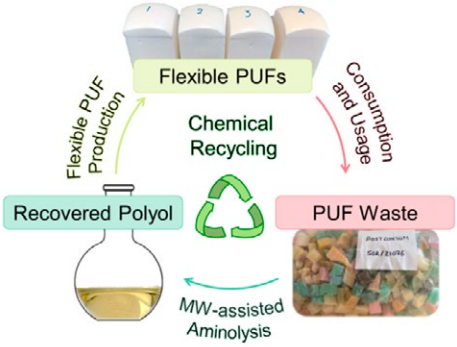

Figure 2.
Chemical recycling of PU foams to recover high quality polyether polyols.

Keywords: chemical recycling, microwave heating, secondary raw materials.

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Recycling of polyurethane thermosets – a recurring challenge for over 50 years

Hynek Beneš

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Worldwide production of polyurethanes (PUR)s is constantly growing. From 2020 to 2022, sales of PURs grew from 23.9 to 25.2 million tons, and the global market is expected to reach almost 27 million tons in 2025 [1]. PURs are a versatile class of materials, which are mainly produced as thermosets and found applications as foams, coatings, adhesives, etc. in many industrial sectors. However, due to their variable chemical composition and crosslinked structure, their recycling has been a challenging issue for over 50 years.

Physical recycling is limited because PURs cannot be reprocessed. Waste PUs (mainly foams) can only be ground into a fine powder and added (max. 10%) to the polyol stream during the production of new PU products. Chemical recycling (hydrolysis, glycolysis, acidolysis, etc.) of PUs are generally highly energy-intensive processes that usually require high temperature, pressure, and the addition of expensive catalysts. Therefore, new end-of-life options for PURs are being investigated, which would enable their integration into the circular economy. In this contribution, various methods of PUR recycling are critically reviewed and the most challenging issues are discussed.

Keywords: polyurethane, recycling, solvolysis

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Recycling of Polyamide 11 and 12 for 3D printing applications

Soeren Griessbach

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Nowdays still a huge amount of used laserintering material is being dumped. Only 25% of the fresh material what is used were taken out the process as parts. All other material is dumped. Especially with large frame machines this is an environmental desaster. We present a technology how handle the powder and bring back recycled material into the production process to reduce waste up to 100%.

This technology is available for any all mid-size and large frame machines by all common machine suppliers with all standard PA12, PA11 and PA12-12. Saving 1kg of PA12 means saving 7kg of CO_2.

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Bio-based polyurethane foams with enhanced recyclability

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In 2021 57.2 Mt of plastic was produced just in Europe, making the plastic industry the 8th biggest one on the continent. Yet only 16.4% of the produced plastics were recycled [1]. These days more and more attention is being put on the problem of polymer recycling. However, developing new recycling methods on an industrial scale is time-consuming and not always cost-effective. Another approach to the problem is developing more sustainable materials which would be better degradable.

In recent years, the polyurethane (PUR) market has experienced significant growth as the product demand for the construction, automotive, and packaging sectors is constantly rising. Currently, PUR foams are the major product segment in the PUR market, accounting for over 50% share of the global revenue [2]. However, a big part of those materials are still non-degradable. The degradability of PUR is generally obtained by the incorporation of hydrolytically labile groups into the polymer chain. This is typically achieved by using a polyol with hydrolyzable (e.g. ester) groups. Moreover, incorporating hydrophilic entities such as glycol chains into the polymer's structure can increase the rate of degradation [3].

In the study, three series of bio-based aliphatic poly(ester-ether) polyols of different functionalities (2, 3, and 4) based on succinic acid (SA) and tetraethylene glycol have been synthesized. These SA-polyols have been further successfully employed for the preparation of ultra-low density polyurethane foams as a full (100%) or partial (80%) replacement of high-functional polyols, petrochemical (commercial) or bio-based (tall oil-based) polyols. All prepared PUR foams were semi-rigid with partly-open cellular structures having a beneficially low apparent density of 15-16 kg·m-3. The influence of SA-polyols bearing hydrolysable ester linkages on the chemical recyclability of the PUR foams was evaluated. It was found that the introduction of SA-polyols into the PUR foam structure led to facile foam recycling via solvolysis. Up to 2.7 times faster progress of glycolysis was observed when the SA-diol was incorporated into the structure of PUR foam. Moreover, it was found that the recycling time is not only influenced by a degree of crosslinking but also by the supramolecular structure of the PUR foam segments.

Keywords: Polyurethane, foam, biobased, recycling, solvolysis.

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Improving the quality of polypropylene recyclates

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In recent years, the recycling of polypropylene (PP) has gained increasing attention. This is due to the European Union's targets to increase the recycling rate of plastic packaging and the use of recycled plastics in new products [1, 2]. However, the quality of recycled PP is still inferior to virgin PP due to non-polymeric and polymeric contaminants and degradation processes [3, 4]. In addition, the blending of different PP grades during recycling is an obstacle to the use of recyclates in certain applications and processing methods. In particular, the suitability for processing methods requiring low melt flow rates (MFR) is limited. This work deals with the improvement of the quality of PP recyclates based on improved sorting techniques in mechanical recycling, with particular focus on reducing the MFR.

To get an overview of the source material for PP recycling, the composition of different PP waste bales was analysed. The proportions of different contaminants and the different processing methods were determined. In order to demonstrate the possible improvement through improved sorting, a recyclate from a manually sorted fraction containing only low MFR PP waste products was compared with a commercially sorted PP recyclate. The two recyclates were compared using different testing methods, with special focus on the MFR. Using the waste samples obtained from the bales, data was collected on industrial Near Infrared (NIR) sorting lines. This data was used to develop new data processing methods in order to enable automatic sorting by processing method.

Quite high levels of contaminants were found in some waste bales, highlighting the need for additional sorting and treatment of the waste prior to reprocessing into recyclates. The proportions of the different processing grades varied seasonally, which could lead to variations in the properties of the recyclates. Injection moulded products were found to have the highest percentage in the waste bales, making sorting by processing method important to produce recyclates with a low MFR. Manual sorting by processing method produced a recyclates with a lower MFR suitable for processing methods such as thermoforming or extrusion. The developed data processing methods showed promising results in enabling automatic sorting of low MFR fractions from high MFR fractions.

Keywords: polypropylene recycling, sorting, recyclate quality, melt flow rate

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Long-term properties of PP recyclate blends

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Recently, the recycling of polymers in particular gained increasing attention in the scientific, industrial and commercial community as well as the wider public. One the one hand, due to the materials economic and technical relevance, and on the other hand to the environmental and social challenges connected with the waste perspective and circular economy of plastic products. Currently, only 13 % of all plastic waste in is processed into recyclates and used in new products, the highest amount of 46 % of the recyclates are used in the building and construction sector [1]. By adding virgin material, the mechanical properties of the recyclate can be improved and higher amount of recyclates can be processed into new products [2]. In this work, several virgin polypropylene (PP) materials, which are commonly used in the building and construction sector and recyclates, especially post-consumer recyclates, were compounded with different weight fractions of 10 % and 40 % of recyclates. Different virgin grades, which are used in the common processing methods like injections moulding, blow moulding or extrusions process were analysed and compounded. Afterwards, the rheological, mechanical and long-term properties of the different blends were evaluated. The melt flow rate (MFR), which is often used in quality control, was used to determine the rheological properties. The flow behaviour of the pure materials (virgin and recyclate) has an influence on the MFR of the blends. For the identification of the mechanical properties, tensile tests and Charpy impact test were performed to determine the strength and the deformation behaviour as well as the impact strength. All mechanical measurements demonstrate the similar trend, as high recyclate content yields to a lower mechanical performance. To analyse the crack resistance of the blends, the for polyethylene standardized, crack round bar test (CRB-Test) was applied. With increasing amount of virgin material, the crack resistance can be improved. There is a non-linear correlation between the crack resistance and the recyclate content. These applied measurements are able to identify the influence of the recyclates on virgin materials. Especially for long-term relevant material properties a highly non-linear dependency of the recyclate content must be considered.

Keywords: crack resistance, polypropylene, recycling, post-consumer

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Development of reusable adhesives systems for polymer bonding applications

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The use of carbon and glass fiber reinforced polymers in the transportation industry has considerable increased during the last years as an effective method to reduce the weight of their components. Moreover, a strong focus has been set on adhesive bonding strategies for composite materials, especially for structural applications where guaranteeing the safety of the passengers is of crucial importance.

Adhesives based on polyurethane (PU) and epoxy formulations are mainly used as standard systems in the composite industry. Nevertheless, there is not a common and convenient method to lose and reprocess adhesive networks in a controlled manner. A solution to overcome this challenge is the development of reworkable systems, which permit a debonding on demand avoiding any damage in the substrates. This work aims at the preparation and further re-processability study of a PU adhesive with associative dynamic covalent bonds.

Inspired by previous works of Zheng [1] and Yan [2], model polymer networks consisting of polyethylene glycol (PEG), 4,4'-methylene-bis-(phenyl isocyanate) (MDI), and ethylenediamine-N,N,N',N'-tetra-2-propanol (EDTP) and glycerol were prepared and further characterized. Thermogravimetric analysis (TGA) were done to study the decomposition and stability behavior of the dynamic networks, obtaining first degradation onsets in the range of 180°C and 238°C. Plate-plate rheometer tests were performed at different temperatures to investigate and probe the bond exchange reactions [Figure 1a]. In a second step, a polymeric MDI was used to prepare a formulation more suitable for a real case application. TGA tests showed a thermal stability up to 180°C. Plate-plate rheometer tests were also conducted, demonstrating an increased stress relaxation and viscoelastic flow behaviour at high temperatures, giving rise to the re-workability of the final formulation [Figure 1b]. Finally, lap-shear tests using CFRP and GFRP as substrates were carried out prior to and after reprocessing to investigate whether the bonding performance could be retained.

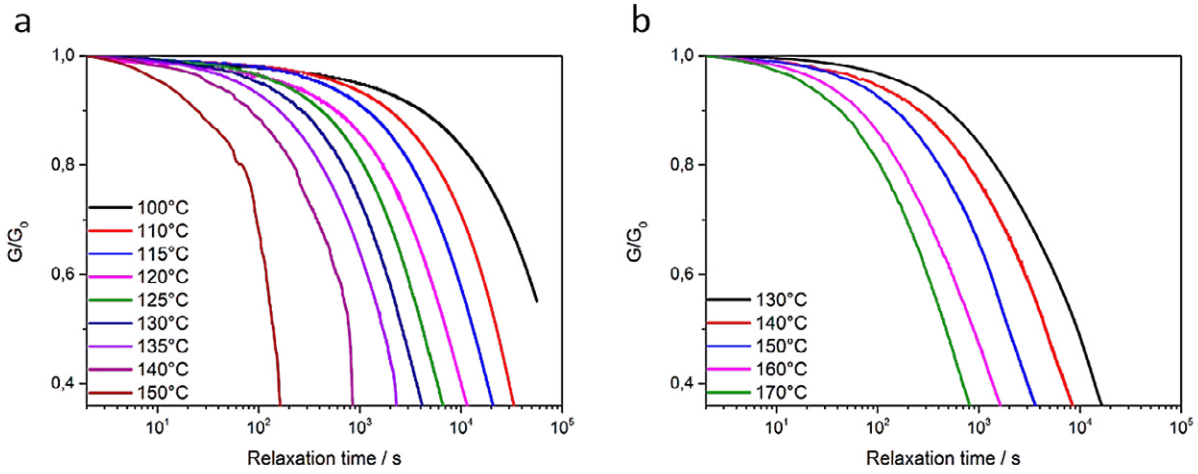

Figure 1 - Stress-relaxation curves at different temperatures of the most stable model network (a) and the final adhesive formulation (b)

Keywords: adhesive, reprocessing, vitrimer, polyurethane, bonding.

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Effect of recycling on the properties of glass fiber reinforced injection molded polymer products

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The use of glass fiber-reinforced polymer composites in the automotive industry has been widespread for a long time due to their excellent technical properties. However, due to environmental regulations and consumer demands, using recycled materials is challenging for the industry [1]. Recycling the raw material can weaken the mechanical properties, thus reducing the maximum performance of the final products [2]. Several studies have investigated the recycling of glass fiber-reinforced polyamide. As a result, we need to minimize the effect of fiber fragmentation during processing [3]. Our study investigated the mechanical properties of glass fiber-reinforced PA materials with 15 m%, 30 m%, 45 m%, and 60 m% glass fiber as a function of fiber content and the effect of fiber fragmentation due to recycling. We performed tensile and bending tests on the specimens to examine specific mechanical properties. Knowledge of changes in mechanical properties increases the reliability of recycled materials and, thus, their market potential.

Keywords: recycling, fiber fragmentation, glass fiber reinforcement, injection molding, mechanical properties

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Development of polymerization of T-RTM technology using 3D printing

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Nowadays, industry – especially the automotive industry – is placing great emphasis on using and developing composite materials, as their specific strength exceeds that of metals, which enables significant weight reduction. In addition to the necessary mechanical properties of the materials used, the possibility of recycling the finished products has become an important aspect. Conventional thermoset composite materials meet the necessary strength and stiffness specifications. Still, recycling is not yet possible [1].

A solution for recycling is using thermoplastic matrix composites. Still, the high viscosity of the polymer matrix does not allow the proper impregnation of long fiber reinforcement structures. In-situ polymerization can overcome this difficulty, which can be achieved by T-RTM (thermoplastic injection moulding) technology [2]. In T-RTM reactive process, thermoplastic polymers are present as monomers and polymerization occurs during the manufacturing process. Caprolactam is the monomer of polyamide, which can be polymerized in-situ by anionic ring-opening polymerization to produce PA6 at the end of the manufacturing process. The presence of two substances, initiator and activator is necessary for the appropriate progression of the chemical reaction during polymerization [2]. The quality of the polymerization in a closed, temperature-controlled mould depends on the proper mixing of activator and initiator, which is characterised by the degree of conversion [3].

My aim is to analyse the properties of PA6 products manufactured by T-RTM reactive technology. I have investigated the quality of polymerization in the prepared products at given distances along the flow path. A 3D-printed part was placed in the inlet of the injection mould in order to modify the flow. I have analysed the effect of this component on the polymerisation process at different stages of the flow path. The testing methods used included thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC).

Keywords: T-RTM, polymerisation, PA6, 3D printing.

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Fourier-Transform Infrared Microscopy Analysis of recycled copolymer materials

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The recycling of plastics through the production of new copolymers or polymer composites is increasingly being researched and is a promising method in the field of waste management. The European Commission published a European Strategy for Plastics in a Circular Economy in early 2018, which emphasises improved design and production of plastics and plastic products to facilitate reuse, repair, and recycling [1]. This is the document that, in addition to the Paris Agreement on Climate Change stimulates further developments on this topic. Mechanical recycling is common in the polymer industry to reprocess plastics, usually using extruders or injectors [2, 3]. Recycled materials can be mixed with virgin resin or natural fillers to obtain better properties at reduced costs [3, 4]. At the same time, using plastic waste as a raw material for new products reduces waste accumulation, which is leading to a cleaner environment [5, 6].

Vibrational spectroscopy is an ideal tool that is widely used for complete chemical and physical structure determination of polymers, qualitative and quantitative analysis of polymer blends, determination of their compatibility and study of their degradation processes. Fourier transform infrared spectroscopy (FTIR) is the traditionally preferred method [7-12]. However, conventional FTIR spectroscopes do not allow us to record the spectra of a sample at the microscopic level, so a system of FTIR microscopes has been developed. This type of microscope gives us a better insight into the microstructure of different copolymers or polymer composites.

In this study, FTIR microscopy was used to investigate the distribution of different polymer structures in the recycled material obtained from disposable surgical masks (DSM). DSMs mainly consist of three main parts - a three-layer filter part made of nonwoven and melt-blown polypropylene, a nose wire made of iron wire covered with polypropylene, and ear loops containing different thermoplastic and thermosets polymers such as polyester, polyamide 6, and polyurethane [13]. The recyclate was first prepared by grinding the sample and then extruding it using a twin-screw extruder with temperature zones ranging from 165 to 200 °C. The resulting filament was granulated and then used to produce bend test specimens by injection moulding. An FTIR microscope was used to map the surface of specimens and the cross-section area of the samples to monitor the dynamics of the distribution of different polymers in bend test specimens. Such analysis is important for understanding recycled materials, especially when they are composed of a variety of different polymer materials.

Keywords: FTIR, microscopy, spectroscopy, recycled materials, copolymers

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Characterisation of the chemical composition and thermal properties of SARS-CoV-2 rapid antigen tests cassettes present in Slovenia

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The covid-19 pandemic resulted in an enormous generation of medical waste due to the increased use of personal protection equipment, vaccination accessories, and single use diagnostic devices. The fast generation of infective waste caused a high burden on waste management systems and emphasised the need for the establishment of effective systems for recycling of medical waste. Rapid antigen tests for SARS-CoV-2 contain valuable materials (nanogold and polymeric materials), which can be recycled. The establishment of effective systems for recycling polymeric materials from rapid antigen test cassettes requires good knowledge of the composition of different tests in circulation, as many polymer types are not compatible and need to be separated before recycling. The study aimed to characterise the chemical composition and thermal properties of nine different rapid antigen test cassettes that are present in Slovenia. Chemical composition was characterised using infrared spectroscopy with Fourier transformation (FT-IR), while thermal properties were characterised using differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA). FT-IR analysis revealed that rapid antigen test cassettes are based on high-impact polystyrene (HIPS), except for one sample which was composed of acrylonitrile butadiene styrene (ABS) copolymer. DSC was used for the determination of glass transition temperatures (Tg), which ranged from 90 °C – 99 °C for HIPS, while ABS had a Tg of 107 °C. Seven samples exhibited exothermic peaks of various enthalpies during cooling runs of DSC measurement that were ascribed to the crystallisation of unknown components (most likely demoulding agents or impact modifiers). TGA was used to determine decomposition temperatures which ranged from 426 °C to 433 °C and ash contents, which ranged from 1.3 % - 3.7 %.

Keywords: medical waste, rapid antigen tests, recycling, chemical composition, thermal properties

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Mechanical Recycling of Carbon Fibre Reinforced PA66

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Fibre-reinforced polymer composites are known for their excellent mechanical properties and are increasingly used in structural and lightweight applications as alternatives to traditional materials. However, at the end of their life cycle, these materials are typically disposed of in landfills. Carbon fibre polymer composites (CFRPs) are very valuable from the functional side and proportionately expensive materials that should be considered for recycling and reuse due to environmental and economic aspects. This study focuses on the influence of multiple cycles of mechanical recycling, involving grinding and injection moulding, on the mechanical and thermal properties of PA66. The mechanical and thermal properties were evaluated for mechanical recycling of up to five cycles. The results showed that the mechanical properties, such as modulus and strength, generally deteriorated slightly with each recycling cycle, while the thermal properties remained more or less in the same range. The observed changes in the properties occurred due to the degradation of the polymer matrix and the shortening of the carbon fibres. The findings emphasize the importance of mechanical recycling for carbon fibre-reinforced PA66 composites since they retained superior properties even after five cycles of recycling. Therefore, it is recommended to recycle carbon fibre-reinforced composites to minimize waste and maximize resource utilization, at least once or even more if possible.

Keywords: mechanical recycling, carbon fibre, PA66, carbon fibre reinforced composites.

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From post-consumer polymer waste to technical parts

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The growing emphasis on sustainability, driven by new laws, regulations, and consumer demand, is pushing plastics processing companies to reduce the carbon footprint of their products without compromising performance or cost. Incorporating recycled materials presents a significant opportunity to achieve this goal, but availability and quantity constraints hinder its widespread adoption. Recycled polyethylene terephthalate (rPET), a material readily available in large quantities, shows promise in meeting these requirements. Switzerland's successful PET bottle recycling system demonstrates the potential for reducing CO_2 emissions through circular economy practices. However, the recycling process often results in materials that no longer meet the required specifications and therefore cannot be returned to the cycle. Today these materials are either spun into polyester fibres or used for energy recovery and remain practically unused for technical parts. Beside the residuals from the PET bottle stream there is another source for rPET, which is ocean-bound plastic material collected from the sea or beach.

In the field of injection moulding, there is a significant opportunity for these materials, especially with the increasing trend towards using or switching to post consumer recycling (PCR) materials to reduce the CO_2 footprint. By adding suitable additives (nucleating agents, minerals, short fibers) to rPET, it can also be used to produce technical parts, currently made with polybutylene terephthalate (PBT), polyoxymethylene (POM) or polyamide (PA).

However, the demanding processing of the material makes it difficult to produce technically useful parts economically by injection moulding. The main problem is the crystallisation rate of PET, which leads to rather amorphous parts when cooling down quickly. The natural ageing process or use at higher temperatures then leads to post-shrinkage, which can lead to distortion and cracking of the part. Based on various tests and applications, it was possible to demonstrate that high quality technical parts can be manufactured economically from rPET with comparable cycle times in injection moulding.

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Tuning the surface properties of recycled thermoplastic composites by the injection moulding process parameters: Atomic Force Microscopy study

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Surface science touches every aspect of our lives. Research activities on the surfaces of polymer composites with wettability, adsorption and controllable adhesion, which ultimately determine the functionality of the polymer composite material, have been very active in the last year. New opportunities for the development of polymer nanocomposites have also been linked to the concept of nanoscale filler modification, which differs from conventional bulk fillers due to the higher number of surface energy atoms, providing increased surface area. Many different fillers have been investigated in polymer composites, altering the crystalline structure and morphology leading to changes in surface properties. In this study, addressing the vision of European Green Deal following the concept of sustainability and recycling, aspect of natural filler in polymer are more pronounced as well as the nanoscience challenges for sustainable tailoring methods.

Fibre-reinforced thermoplastic (nano) composites (nW-PPr) of recycled polypropylene (PPr) and nano functionalised wood fibre (nW) were developed by extrusion and injection moulding. In order to understand the effect of filler and processing parameters (mould temperature) on the composite crystalline structure, morphology and surface behaviour of polymer composites, an injection moulding technique was used to prepare a sample with a controllable skin-core surface structure. The differences in surface properties were characterised in detail. The atomic force microscopy (AFM) technique was used to analyse the surface topography and morphology of the samples. Measurements were performed in tapping mode at different scan sizes, acquiring height, amplitude, and phase images. The analysis showed clearly visible fibers incorporated into the surface of the samples. The mean surface roughness (Sa) was used to demonstrate the presence and the dispersion/incorporation of nW on the surface of the composite nW-PPr. The differences in topography between the samples prepared by varying the mould temperature (30 °C, 60 °C, 90 °C, 150 °C) were obtained. The highest Sa was measured in the composites with 30 °C mould temperature, indicating the filler effect on the surface. As the mould temperature increased, the value of Sa of the nW-PPr composites decreased.

Keywords: surface properties of fibre reinforced thermoplastic composites, atomic force microscopy (AFM).

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On-Demand Activation of Dynamic Bond Exchange in Covalently Crosslinked Polymers

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In covalent adaptable networks, a defined onset of bond exchange reactions, and the associated suppression of undesired creep at operating temperatures is difficult to realise. For numerous bond exchange mechanisms, e.g. dynamic transesterification, latent catalysts pose a straightforward approach to overcome this challenge [1]. In response to an external trigger, such as light or temperature, an active catalyst is released, and enables an on-demand reorganisation of the network topology exploitable for thermomechanical reshaping, welding and recycling on a macroscopic scale. We introduced a library of latent base catalysts to selectively release the catalysing species for thermoactivated transesterification in dynamic polymer networks. First, a photolatent base was incorporated in an optically transparent thiol epoxy polymer, and the spatiotemporal activation of dynamic transesterification was enabled by the light mediated release of a strong guanidine base from a quaternary ammonium salt [2]. Adverse effects of this class of photolatent catalysts (low solubility, narrow absorption window) were overcome by using photoactive N-substituted derivatives of amidine bases. Owing to their commercial availability, an important step toward the technical application of spatially resolved activatable dynamic polymers has been made [3]. In a subsequent study, the whole approach was transferred to a 3D printable thiol-ene photopolymer. The sequence depended wavelength-orthogonality between the curing reaction and the light-induced cleavage of the photolatent base catalyst enabled an efficient network formation without premature release of the catalysing species [4]. Advancing from photolatent bases, recently introduced thermolatent bases allowed the production of composites and objects in any spatial geometry without restrictions regarding their optical transparency. 3D-printed objects and composite structures exhibited an outstanding creep resistance prior to catalyst activation. However, once a defined temperature threshold was exceeded, they were efficiently reshapable and weldable [5].

Keywords: Covalent adaptable networks, Vitrimers, Dynamic Transesterification, Latent Base Catalysts.

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Influence of Compatibilizer on Viscoelastic Properties of the Thermoplastic Polymer-Based Biocomposites

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A composite material consists of two or more physically distinct phases, the combination of which produces mechanical properties that are different from those of the individual constituents. Thermoplastic polymer-based biocomposites are of great importance due to their remarkable strength and stiffness combined with very low weight. When using natural fiber reinforcement, the main advantage is that they have almost no effect on the wear of processing equipment. With glass fiber reinforced composites, wear of processing equipment is a major disadvantage. The strength to weight and stiffness to weight ratios are many times higher than steel or aluminum and property combinations that cannot be achieved with metals, ceramics or polymers alone [1]. Thermoplastic polymer based biocomposites have gained importance in all fields of research due to their environmentally friendly properties. The main disadvantage of natural fibers is their thermal stability. This can be avoided by using a suitable thermoplastic matrix. Polypropylene and polyethylene are the most suitable due to their high processing temperature, as the thermal decomposition temperature of natural fibers is higher [2, 3]. The hydrophilic nature of natural fibers and the hydrophobic nature of polypropylene (PP) and polyethylene (PE) pose a challenge to researchers. It can be solved in several ways: by thermal treatment of the natural fibers, by chemical modification of the natural fiber surface, or by the addition of compatibilizers. Modification by compatibilizers is often more commercially acceptable and can be carried out together with mixing and pelletizing of biocomposites based on thermoplastic polymers. Although twin screw extruders are suitable for processing, at least antioxidants should be added to prevent thermal degradation during processing. The addition of a compatibilizer improves the interfacial adhesion between natural fibers and thermoplastic matrix. The improved adhesion leads to higher stiffness and strength. The addition of maleic anhydride-grafted polypropylene (PP-*g*-MA) in red pine fibers PP biocomposites [4] increased strength and stiffness, but had no significant effect on toughness. Researchers reported [4] higher strength, stiffness and toughness by adding PP-*g*-MA in sawdust PP biocomposites. Biocomposites are characterized by traditional characterization methods and with the right combination, biocomposites can be characterized in detail. Dynamic mechanical analysis (DMA) is complementary to traditional mechanical (tensile and flexural tests) and thermal (DSC and TGA) methods. The dynamic nature of DMA results in measured quantities: storage modulus (E'), loss modulus (E") and loss factor (tan δ). The dynamic mechanical behavior of the measured samples allowed us to study the adhesion of the natural fiber-thermoplastic matrix at the interface, the morphology, the glass transition, the damping behavior and the elastic behavior. The viscoelastic properties of the thermoplastic polymer-based biocomposites can be evaluated with DMA and confirmed with additional tensile, flexural, DSC, TGA and impact tests. E' can be related to the stiffness of the sample, E" to the internal friction and tan δ as a mechanical damping factor to the molecular motions and viscoelasticity including the contribution of defects [5].

In this work, the preparation of thermoplastic composites with thermoplastic polymer-based biocomposites with the addition of different biomasses (waste paper, cellulose fibers, Fiber-Plast 35E, miscanthus fibers, beech sawdust and hemp husks) and various commercial compatibilizers (polyethylene grafted with maleic

anhydride (PE-g-MA), styrene maleic anhydride (SMA) and PP-g-MA) is described. Compatibilizers are widely used in composites and allow the use of standard processing equipment. In addition, an incompatible compatibilizer was chosen for the matrix PE-HD to test the viscoelastic properties and reinforcing effect for use in recycled thermoplastic materials where traces of PE are normally present. The viscoelastic behavior of these composites was investigated using dynamic mechanical analysis (DMA). The mechanical and thermal properties were investigated using Universal Testing Machine (UTM), Differential Scanning Calorimetry (DSC) and impact tests. The research work focused on different cases, forms and amounts of compatibilizers on the biocomposite materials. The greatest effect on the viscoelastic properties of the thermoplastic PE-HD-based biocomposites was measured with the PE-g-MA compatibilizer. The compatibilizer has no effect on the thermal behavior of the biocomposite, but the highest strength and stiffness of the biocomposites were obtained. All biocomposites were prepared with 30 wt.% of different natural fibers and the thermoplastic matrix PE-HD. Except the sample with hemp husks, all the biocomposites have higher strength compared to the untreated PE-HD matrix, all the biocomposites including the sample with hemp husks have higher stiffness. The storage modulus was improved for all biocomposite samples, although "incompatible" compatibilizers with PE-HD matrix were also tested. These particular compatibilizers showed that the traces of the PE in the recycled PP, PS would not affect the compatibilization with natural fibers. Based on the results, it can be concluded that there is an excellent improvement in storage modulus, strength and stiffness with compatibilized cellulosic fibers. Miscanthus fibers in the biocomposites also exhibited good mechanical properties but caused brittleness. A good combination of mechanical properties and toughness is waste paper incorporated in PE-HD matrix with compatibilizer. For blends as matrix, SEBS-g-MA compatibilizer is the best option to improve the toughness of biocomposites with natural fibers.

Keywords: thermoplastic biocomposites, compatibilizer, DMA, viscoelastic properties, natural fibers

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Stiffness and Strength Steering of Recycled Polypropylene (rPP) with Addition of Waste Paper

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The paper presents the preparation of a commercially useful engineering biocomposite based on recycled polypropylene (rPP) matrix and waste paper with the addition of compatibilizers. By combining the mixtures on a twin-screw extruder, we prepared several samples of biocomposites with different compositions (polymer matrix - waste paper fibers - modifier), injection-molded test samples, and then studied the interphase interactions between the polymer matrix and waste paper fibers. Interphase interactions were improved by the addition of various compatibilizers and evaluated by dynamic mechanical analyzes and with microscopic analysis of fracture sites after tensile and toughness tests. The characterization included mechanical, thermal, and rheological properties. As a matrix, we used polypropylene (PP), donated by a company engaged in the separation and recycling of polyolefins from post-industrial and post-consumer waste. A paper processing company donated the waste paper. As a reference material with which we compared the properties of the prepared biocomposite, we prepared an rPP composite that contained a waste thermoset composite with a moderate content of glass fibers. Waste ground thermoset composite with glass fiber was donated by a company engaged in the production of reinforced polyester products. During the research, we proved that by adding the right combination of modifiers, we can successfully increase the strength and stiffness of the prepared composites. Since fibers were added to rPP matrix, the material's toughness decreased, the fracture surfaces of these samples were used in the microscopic analysis, which proved a higher adhesion between the fibers and the rPP matrix in the samples with added modifiers. The mechanical properties of the prepared composites also improved at higher temperatures. The addition of modifiers and fibers did not significantly affect the thermal transitions of the material. With some additional research and modification, rPP composite with added waste paper fibers would definitely be interesting as a commercially useful engineering composite.

Keywords: Recycling, carbon footprint, recycled polypropylene, recycled biocomposite, stiffness and strength

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Use of Recycled Materials Based on PE in Rotational Moulding Technology

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The purpose of the paper is to present the development of a composite of plastic packaging waste and recycled material based on PE, for rotational moulding technology. The material for the rotational moulding of different products, which can be produced also in sports products needs to have high impact strength. So with use of recycled material, the mechanical properties of materials can be decreased, and as such the materials cannot be applicable for such applications/production. In the paper we will present how we used the process of injection and rotational moulding, to prepare different samples and compared them with each other. The materials were prepared by different contents of material from plastic packaging waste, from different suppliers, based on LLDPE with different concentration. The materials were then characterized and compared to the obtained values with recycled material and material from plastic packaging waste. Based on the results, we selected a composite that provided the appropriate mechanical and thermal properties. We also developed a three-layered composite from the chosen composite and made a sample based on the process of rotational moulding on one of the sporting goods cases to prove the concept. The structure of this case was made as a sandwich panel of two layers of virgin polyethylene, with a middle layer of polyethylene foam. The multi-layered materials allow us to achieve extreme wall thicknesses with low mass. In the composite, different body structures of composites were prepared. The intermediate layer was polyethylene foam. We also made a composite where the prepared recipe was used as the first layer and recycled polyethylene as the third layer. The results of the analysis showed differences between the properties of the samples prepared by injection molding and rotational moulding technology. Based on the obtained results, the best structure of composite and body structure was defined.

Keywords: Plastic packaging waste, PE – based recycled material, rotational moulding, single – layer composite, three – layer composite.

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Use of sustainable polyolefins for injection moulded closure packaging

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Changing global trends dictate changes in polymers for closure packaging. The paper presents an industrial-relevant need to invest in the development of sustainable materials solutions to keep up with the trends in the highly demanded market. The paper demonstrates that this is possible by monitoring and following the guidelines of sustainable polyolefins use, such as recycled or PCR polyolefins and bio-based polyolefins. The first step towards sustainability and circular economy is the so-called mono-packaging where PP is replaced by PE-HD. This contributes to the entire packaging being suitable for recycling in the same recycling stream. Every polymer change presents challenges in adapting the injection moulding mould, the injection moulding process, and ensuring adequate product quality. This paper summarizes all of these challenges and solutions. By using DoE, we will determine the suitable injection moulding parameters and check the sample's dimensional and functional quality. Additionally, we will perform the characterization of the used polyolefins and verify the suitability of polymer selection for the selected product.

Keywords: Sustainability, injection moulding, bio-based polyolefins, recycled polymers, biopolymers.

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Neoteric Solvolysis Methods and how are they being developed

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The search for new ways to reduce, reuse and recycle, leads to the search for sustainable developments in order to extend the life of materials through the application of recycling technologies, such as solvolysis.

Although a material has reached its end-life, it does not mean it has to be discarded. For this reason, several solvolysis techniques are being developed to provide a new life to the components of the composite materials; allowing to be separated from one another and recycling them so they can return to their raw state.

Solvolysis is a chemical reaction in which the solvent, the component in excess is able to break-up the chemical bonds of the substrate. It works up by displacing an atom or group of atoms in the substrate molecule. There are two main reaction mechanisms: SN1 and E1. They both work via carbocation formation.

Nowadays, using the previous knowledge about this methodology, the idea is to be able to substitute the more harmful solvents used by greener solvents with less health and safety issues. The solvolysis processes normally consumes a lot of energy and as the solvents are very harmful the posterior treatments are difficult to effectuate.

This way, newer strategies are being developed to recover the different parts of materials as composites. Fibres can be retrieved and separated from the resin in which they are imbued. Depending on the length of the fibres, the r-fibre can be used in new processes. Resin breaks-up and dissolves into the solvent so it can be extracted to be reused.

Keywords: Solvolysis, Green Solvents, Safe and Health, Composites

Recycling of PET via aminolysis

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Polyethylene terephthalate (PET) is a saturated polyester synthesized by the esterification of a dibasic acid (e.g. terephthalic acid) and a diol (e.g. ethylene glycol) and is used for fibres, bottles, films and other moulded products. PET is recycled by both physical and chemical routes. The physical route generally consists of remelting and forming new products with the help of suitable additives. PET can be chemically reprocessed by complete depolymerisation into oligomers, monomers and other by-products. In this way, a wide range of terephthalamide monomers can be produced, which can serve as building blocks for high-performance materials with desired mechanical and thermal properties.

In this study, different amines (ethylenediamine, 1,4-diaminobutane, 1,6-diaminohexane) were used to depolymerize post-consumer PET beverage bottles. The bottles were cut into pieces with an edge length of less than 2 cm. Aminolysis of PET was carried out in the presence of organocatalyst (1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD)) and with an excess of amine (ethylenediamine, 1,4-diaminobutane, 1,6-diaminohexane; 1.5 and 3 eq.). After completion of the reaction the product was isolated, washed with organic solvents and dried. The products bearing amino functional groups were characterised by Fourier transform spectroscopy (FTIR), nuclear magnetic resonance (NMR), differential scanning calorimetry (DSC) and thermal gravimetry (TGA).

Keywords: PET, Aminolysis, depolimerization

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Novel Eugenol Based Acrylate Oligomers: Their Photochemical Curing Behavior and Mechanical Properties with Reproducity

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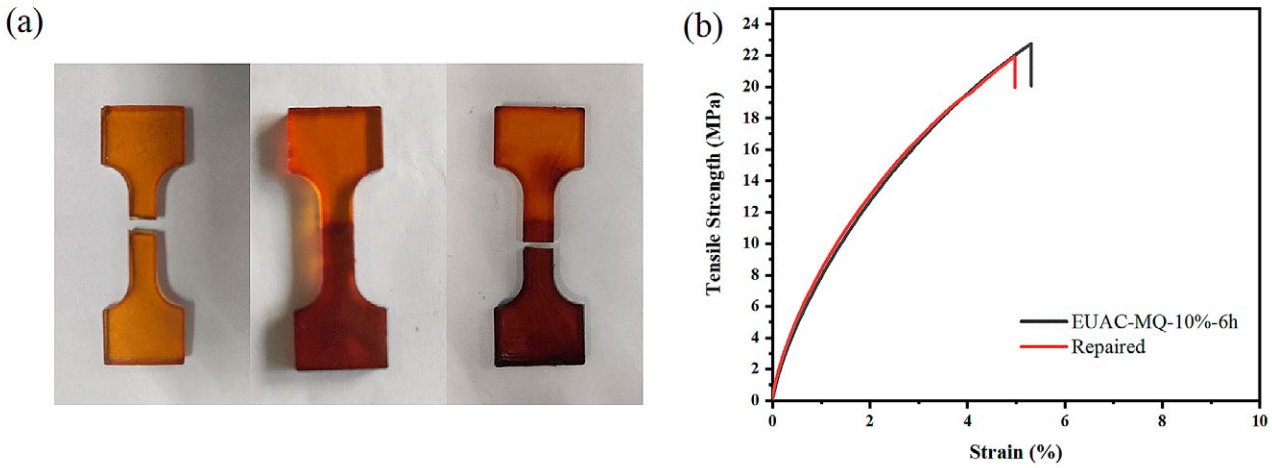
Acrylic oligomers are the most widespread light-curable 3D pringting materials in present, due to their many advantageous properties such as fast curing and low energy consumption.[1]. However, important issues in developing new light-curing based 3D printing materials are concerned including using more environmental raw materials and reproducity for printed materials. In this research works, we explored a new kind of eugenol-based epoxy acrylates. These photothermal curable oligomers are composed of monofunctional eugenol epoxy based acrylate (EUAC) and multifunctional eugenol epoxy acrylate based MQ silicone resins (EUAC-1.4MQ). The mixtures are first cured under UV-light to form the shape and then heated at high temperature to accelerate the rate of ester exchange reaction for enhancing the strength. The optimal formulation is determined by adjusting the ratios between EUAC and EUAC-1.4MQ. The addition of MQ silicone resins can significantly improve the mechanical properties of the cured thermosets, and the tensile strength increases with the extension of the heating time. The effect of heating temperature and time on the crosslinking network are investigated by DMA test and stress relaxation test. The activation energy of the ester exchange reaction (68.2 kJ/mol) is calculated by relaxation time, which is similar to the results reported in the literature. Thus, 3D printed assembled parts with excellent mechanical properties could be manufactured with higher efficiency. Also, based on the ester groups exchange interactions in the molecule chain networks, the self-healing behavior of the cured material was occurred. The repaired samples made by re-heating, exhibited the similar mechanical strength.



(a) Photos of the repaired samples;
(b) Tensile curves of the repaired samples

Keywords: Eugenol based acrylic oligomers, Photochemical curing, 3D printing materials, Reproducity

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Plastics in circular economy - requirements and challenges

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Pollution of the environment with plastic waste is one of the major problem areas of our time. The plastics industry continues to be a growing sector, with forecasts predicting a doubling of the volume by 2030 [1]. In 2021, we had a quantity of 390.7 Mt of plastic per year, of which only 32.5 Mt came from recycled materials and only 5.9 Mt of plastics were produced on the basis of renewable raw materials. The large remainder of 395 Mt is based on fossil raw materials [2].

Countries like Switzerland or Germany would need 3 Earths for their consumption, so Switzerland's earth overshot day is already on 13 May this year [2]. In order to achieve the goals, we have to decouple our value creation from the pure consumption of raw materials. Until now, our economy or the value chain has been linear and the processes and requirements have been optimised and defined in very narrow specifications. At the end of the product life cycle, the product is handed over for disposal and loses all its value. In many countries, a large part of the material flow still goes to landfills or the material is sent directly to thermal recycling. If plastic waste is collected and processed, possible areas of application are then sought. This procedure usually leads to downcycling of the material and only low-quality products can be produced. The aim must be to give value to plastic waste, so the linear economy must change into a circular one, but above all the development chain must also change. We must already have the possible end-of-life use in mind when creating the product and we must already know at the collection and sorting stage what qualities and in what products we want to end up, see Figure 1.

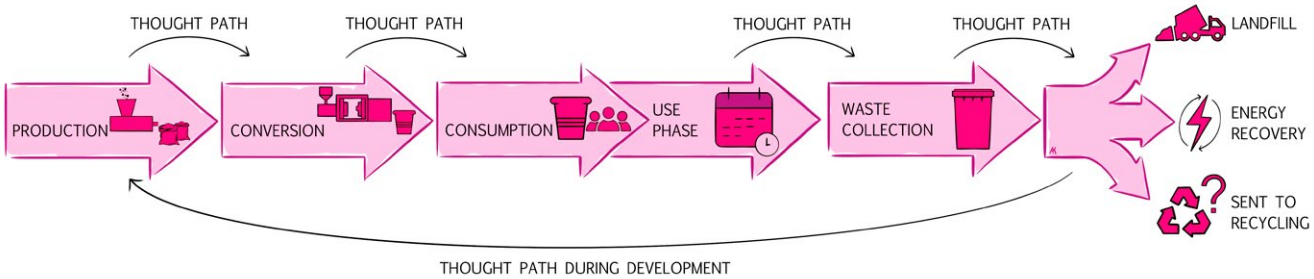

Figure 1: Schematic representation of the linear economy with the connection of the flow of thoughts end of life

When this cycle is closed, we enter a circular world, which, however, is divided into the material recycling area and a biogenic area, see Figure 2. In a circular world, we also work with specifications, but in order to ensure the use of recyclates, we often have to accept small compromises in the mechanical values and colour consistency, because the high values of the virgin material and the colour consistency are often not achievable. In principle, we should focus on the necessary required product properties; in some cases we have over-specified components on the material side in recent years. However, this is a very difficult undertaking, as development and testing have to be done from scratch again. Without opening up the specifications, the production of watch cases with recycled PET from Ocean Bound Plastics would not have been possible.

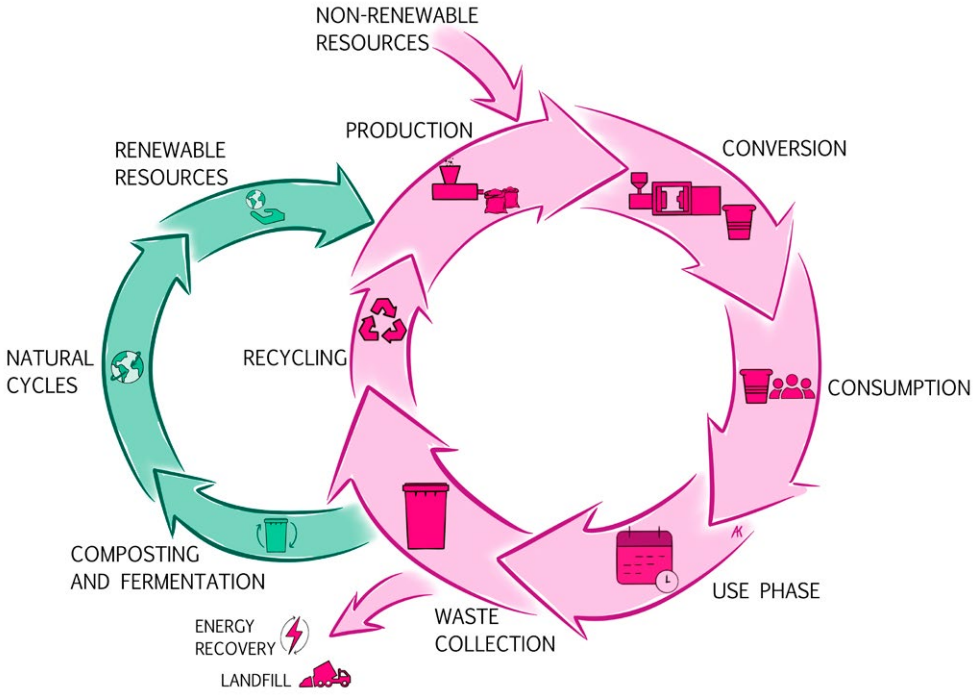

Figure 2: Schematic representation of the circular economy - biogenic and material based

It is important that we do not just target the completely circular approach to plastics. The polymer chains are damaged and apart from PET we cannot repair them so easily. The smell also plays a big role with polyolefins as recyclates. Therefore, we should definitely recycle all plastic materials before we dispose of them, see Figure 3. As a thought experiment, if we recycled only the 40% packaging plastics once, we would have a raw material saving of 156 Mt per year on earth, based on the 2021 figures.

Figure 3: Comparison Linear, recycling and circular economy.

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Thermomechanical devulcanization of rubbers

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Vulcanized rubber has exceptional properties such as flexibility, toughness, damping, and deformability, but its recycling poses challenges due to its crosslinked molecular structure. Unlike thermoplastics, common recycling methods like remelting cannot be utilized on them. Yet, there are promising recycling approaches, mainly focusing on material recovery. These methods begin by shredding rubber into crumb rubber, removing non-rubber components like steel or fibers. This crumb rubber finds use in applications like asphalt modification, artificial turf, or playground flooring, though these aren not true material recycling solutions. For raw material recovery, pyrolysis is an option, involving heating crumb rubber without oxygen, yielding gases, oils, and carbon black, unfortunately having much lower quality than fresh materials. One alternative is to dismantle the 3D crosslinked molecular structure of rubber and attempt to transform it back into the primarily linear structure of its uncured state. If the breakdown is selective, the process is called devulcanization; if not, it can be referred to as reclaiming. Reclaiming is a well-established method but is less selective, whereas various devulcanization methods can result in higher-quality materials that exhibit improved performance after revulcanization. This phenomenon is due to the resulting more advantageous molecular structure of devulcanized rubber than reclaimed rubber. Breakdown of the 3D molecular structure can be achieved by attacking the crosslinks from either a physical or chemical side, and the name of the process is derived from the method that is used for the cleavage of the bonds. Thermomechanical methods consist of presenting heat and shear load to the crumb rubber, while for example, thermochemical methods use heat and appropriate chemicals for the process. This presentation focuses on the thermochemical devulcanization of ground tyre rubbers. The basics of devulcanization will be introduced, then details of the process will be shown. Finally, the properties of the devulcanized rubber and vulcanizates made from it will be presented.

Keywords: rubber recycling, tyre recycling, crumb rubber, devulcanization, thermomechanical devulcanization

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Recycling opportunities for vulcanized rubber goods

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Vulcanized rubbers have outstanding performance, making them essential for our everyday life. Besides excellent flexibility, toughness, damping properties, and deformability, they have a significant drawback compared to plastics: their cumbersome recyclability. Due to their crosslinked molecular structure, the accessible and well-established recycling technologies of plastics based on remelting do not work. However, fortunately, there are many promising ways to recycle scrap rubber products. Recycling methods can be categorized into energy and material recovery techniques, from which material recovery is more beneficial in many terms. These technologies usually begin with shredding the rubber parts into crumb rubber to remove non-rubber components, e.g., steel or fibers. This crumb rubber can further be used for asphalt or concrete modification, filling for artificial turf football fields, or sports field and playground flooring. Nevertheless, these application fields do not mean complete material recycling. Two ways to regain raw materials for rubber products are present: pyrolysis and reclaiming or devulcanization. During pyrolysis, crumb rubber is heated without oxygen and degraded seriously. The resulting materials are gases, oils, and carbon black. However, their quality is far from their fresh counterparts. During reclaiming and devulcanization, the 3D crosslinked molecular structure of the rubber is broken down and transformed back into the mainly linear structure of the uncured state. If the breakdown is more or less selective, the process is called devulcanization; if there is remarkable degradation of the chains of the base polymer, then we can call it reclaiming. Devulcanization technologies are more promising because their selectivity results in a higher-quality material. They can be organized based on the concept used to break the bonds between the polymer chains. Thermal devulcanization means that heat is applied, mechanical devulcanization implies that the material is subjected to shear or other load, and chemical devulcanization means that a suitable chemical is used. The technologies that are used in practice are usually the combination of the abovementioned three approaches; the most widely used technologies are microwave (thermal), thermomechanical, (thermo)-mechanochemical, and biological (chemical) devulcanization. Based on throughput, microwave, thermomechanical, and thermo-mechanochemical-based techniques are the most encouraging. This presentation focuses on summarizing the material recovery opportunities for rubber goods with a particular emphasis on the devulcanization techniques.

Keywords: rubber recycling, tyre recycling, crumb rubber, devulcanization, material recovery

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Industry-focused developments to introduce recycling and sustainability in plastic and composite materials

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Nowadays, polymeric materials (plastics, resins, and composites) are widely used in millions of industrial applications. During the last 50 years, research in polymeric materials has been directed by the requirements demanded by the industry, which mainly are to increase properties and/or to reduce cost. However, the current society, governments and our planet require us to redirect our research towards sustainability, recyclability and circular economy. Millions of tons of polymeric waste are in our landfills, floating in our seas or burning without any treatment.

It is the duty of researchers to work hard to change the world and propose sustainable polymeric processes and materials to the current industry. Tecnalia has been (and continues being) very active in achieving these sustainability objectives that are so highly demanded by society.

Tecnalia participates in regional and European projects in this line, whose objective is to involve the industry as main actor in the need to implement sustainable processes and materials and integrate circular economy. A review of objectives and main result will be presented for next projects:

Regional projects:

- Avansite: New generation of sustainable composites for advanced manufacturing.
- Neoplast: New raw materials from technologies and processes of chemical recycling of plastics for the industrial sectors of the Basque Country in an applied approach to the circular economy
- Ze-Konp: Sustainable and intelligent manufacturing processes of thermoplastic composites of new generation
- Biopolyurethanes: Development of biobased polyurethanes and polyurethanes with dynamic bonds

European Project:

- FiberEuse: Large-scale demonstration of new circular economy value chains based on the reuse of end-of-life fiber-reinforced composites
- Digiprime: Digital platform for circular economy in cross-sectoral sustainable value networks
- Maelstrom: Smart technology for Marine Litter Sustainable Removal and Management

- DeremCo De & Remanufacturing for Circular Economy Investments in the Composite Industry
- EverPV: Highly efficient delamination technologies to recover and reuse metals, glass, polymers from end-of-life photovoltaic panels

Keywords: circular economy, sustainability, plastic recycling, composite recycling, remanufacturing.

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