In-built Triggered Enzymes to Recycle Multi-layers: an Innovation for Uses in plastic-packaging

D8.3: Scientific workshop organisation

WP8: Communication, dissemination and exploitation activities; assessment of legislative and economic aspects

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Abbreviations

BOPP	Bi-oriented PolyPropylene
CEFLEX	Circular Economy for Flexible Packaging
CIW	Common Industrials Waste
EFSA	European Food Safety Authority
EPR	Extended Producer Responsibility
EVOH	Ethylene Vinyl Alcohol
LDPE	Low Density PolyEthylene
MDO	Machine Direction Oriented
NGO's	Non-Governmental Organization
NIR	Near Infra-Red
OPA	Oriented PolyAmide
PBS	Polybutylene succinate
PBSA	Polybutylene succinate Adipate
PCL	Polycaprolactone
PET	PolyEthylene terephtalate
PRE	Plastic recycled Europe
PUR	PolyURéthane
PVDC	PolyVinylidene Chloride
SMART	Specific, Measurable, Achievable, Relevant, Time-bound

Executive summary

During Terminus project, the partners must organize an open scientific workshop held faced to the international scientific community to deliver the main outputs of the academic results of the Terminus project.

Deliverable report

Introduction

For the organization of the scientific workshop, an organization committee has been built-up. It is constituted of:

- The Terminus management team: A. Beakou (general coordinator), T. Langon (administrative coordinator), V. Verney (Scientific and Technical coordinator)
- Laura Sisti (Bologna University)
- Nina Knezic (Terminus dissemination manager)

The main goal of the organization committee has been to collect propositions from the partners to be able to propose a fully built-up Terminus session to be included in the program of an international scientific congress. For that, the organization committee has launched a call to oral communications inside the Terminus consortium. Seven oral propositions have been received covering all the working activities of the Terminus project.

With this content, we have been able to start discussion together with the European Bioremediation Federation which holds each three years its international meeting in Chania in Crete. Because Covid pandemic, the 8th EBC meeting has been postponed by one year and was scheduled to be organized during June 2022. After discussions with the two chairmen, Prof. **Nicolas Kalogerakis** (TU - Crete) and Prof. **Fabio Fava** (University of Bologna), they agreed to welcome a Terminus project session within the topic SS-1. Plastics and Microplastics: fragmentation, monitoring, biodegradation, fate, recycling. They also asked for us to give a plenary lecture devoted to plastic recycling, with a focus on plastic multilayers recycling.

Program

Finally, after discussions a one-day Terminus specific session (June 14th, 2022) was proposed with the following time schedule and program:

TUESDAY, JUNE 14TH, 2022	
8:30 - 9:15	PLENARY LECTURE #3 – ROOM A Chairpersons: Nicolas Kalogerakis & Fabio Fava
ID 188	TERMINUS PROJECT: PAVING THE WAY FOR CIRCULAR AND RECYCLABLE PLASTIC MULTILAYERS Dr Vincent Verney *Recycle-Consulting / CNRS / Sigma Clermont / Clermont Auvergne University*

17:00 - 18:30	SESSION - 6A: H2020-TERMINUS Conference (ROOM A) Chairpersons: Vincent Verney and Patrick Shahgaldian
ID 43	ENZYMATIC DEGRADATION OF THE MOST COMMON ALIPHATIC BIO-POLYESTERS A. Rosato, A. Romano, G. Totaro, A. Celli, F. Fava, L. Sisti, Giulio Zanaroli Dept. of Civil, Chemical, Environmental and Materials Engineering – DICAM, University of Bologna, Italy
ID 105	RECYCLING OF NON-FIBRE FRACTION OF CARTON PACKAGES Åsa Olsson Tetra Pak Packaging Solutions AB, Lund, Sweden
ID 111	THE POWER OF NADES - BOOSTING ENZYME STABILITY TOWARDS THERMAL DEGRADATION Simona Varriale[1], Astrid E. Delorme[2], Jean-Michel Andanson[2], Julien Devemy[2], Patrice Malfreyt[2], Vincent Verney[2], and Cinzia Pezzella[1,3] [1] Biopox srl, Viale Maria Bakunin 12, Naples, 80125, Italy [2] Université Clermont Auvergne, CNRS, SIGMA Clermont, ICCF, F-63000 Clermont-Ferrand, France [3] Department of Agricultural Sciences, University of Naples "Federico II", Naples, Italy
ID 119	ADDITIVE EFFECTS ON CURING ESTER-BASED DEGRADABLE POLYURETHANE ADHESIVES S. Mačiulytė, P. Nemaniutė, D. Bražinskienė, O. Eicher-Lorka, S. Joseph Asadauskas Dept. of Chemical Eng. and Technologies, Center for Physical Sciences and Technology (FTMC), Lithuania
ID 135	COMPLEXITY OF MULTILAYERS, MAIN CHALLENGES AND SOLUTIONS TO REACH THE REALITY OF A SUSTAINABLE INDUSTRIAL PROCESSES, AN SMES POINT OF VIEW Alexandre Fontaine
	STTP Emballage, Sainte-Sigolène, France
ID 137	RHEOLOGICAL IMPLICATIONS OF FUNCTIONAL NANOCHARGES IN POLYURETHANE-BASED ADHESIVES AND THEIR LAMINATION PROCESS M. Ngom, C. Chevalier, S. Mani, Rigoberto Ibarra-Gómez Centre Industriel de la Plasturgie et des Composites (IPC), France
ID 144	NANOBIOCATALYTIC DEGRADATION OF DESIGNER POLYURETHANES Patrick Shahgaldian[1], S. Amirabbas Nazemi[1], C. Wu[1], R. Burn[2], C. Pezzella[3,4], S. Varriale[3,4] and P. F.-X. Corvini[2] [1]Institute of Chemistry and Bioanalytics, School of Life Sciences, FHNW, Switzerland [2]Institute of Ecopreneurship, School of Life Sciences, FHNW, Switzerland [3] Biopox, Naples, Italy [4] Dept of Agricultural Sciences, University of Naples "Federico II", Portici, Italy

The session was successfully organized on June 14th 2022 with full congress attendance for the plenary lecture and interested scientists for the Afternoon session

The program and the abstracts of all the presentation can be found on the congress website:

https://www.ebc-viii.tuc.gr/en/home

https://www.ebc-viii.tuc.gr/fileadmin/users_data/ebc-viii/_uploads/EBC_VIII_DETAILED_PROGRAM_v5.pdf

PLENARY LECTURE #3 : TERMINUS PROJECT: PAVING THE WAY FOR CIRCULAR AND RECYCLABLE PLASTIC MULTILAYERS

Paving the way for circular and recyclable plastic multilayers

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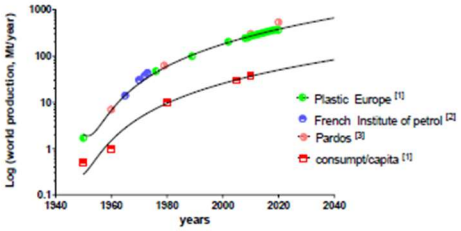
ABSTRACT

Since the second half of the twentieth century, plastic materials have seen an exponential growth. Now, the world production is estimated at more than 380 Mt each year. Plastics are widely used for a great number of different applications. They can be classified according to the time scale of their use: short-term (less than 1 year like for packaging), middle-term (electronics) or long-term applications (automotive or construction sector). Consequently, huge quantities of plastics wastes with different time history are generated and accumulated along the years. If they are non-recycled, they can stay in the natural environment with adverse potential effects like microplastics pollution (atmospheric contamination, fresh and sea water contamination, and soils contamination).



The high level of plastic world contamination makes it mandatory to improve and develop the recycling processes of plastic waste. Today, different plastic recycling processes like (bio)chemical, mechanical recycling or a combination of both are developing. Each are specific, aiming either to come back to the starting base units (monomer) or to reprocess a new secondary stable material for a second cycle of use.

Terminus project aims at unlocking the recyclability of plastic multilayers packaging. These materials with a high barrier level of properties towards oxygen and water are extensively used in packaging for food preservation. They are made of a certain number of different plastic layers with various incompatibilities (polyolefins, PET, paper board, aluminium,..). Thus, they are difficult to recycle, achieving mainly their service life in landfilling or in incineration. Terminus project focuses on adding a recyclable capability to these materials throughout innovative conceptual design. New designed polyurethane adhesives and/or tie layers embarking specific enzymes with a special thermal protection are synthesized to allow an on-demand triggered biodegradation of these layers. Separated delaminated layers can be then collected and recycled individually, unlocking the route towards recycling. Thus, Terminus brings together biotechnological and chemical processes with mechanical recycling to face this challenge.

Acknowledgements: The financial support by H2020 project Terminus contract No. GA-814400 is greatly appreciated.

References:

1 Plastic Europe the facts : https://plasticseurope.org/

2- French Institute of Petrol, Rev. Inst. Fr. Pet., 30, 1, 1975, https://doi.org/10.2516/ogst:1975005

3- Pardos marketing : https://www.pardos-marketing.com/

SESSION - 6A: H2020-TERMINUS Conference

1. ENZYMATIC DEGRADATION OF THE MOST COMMON ALIPHATIC BIO POLYESTERS
A. Rosato, A. Romano, G. Totaro, A. Celli, F. Fava, L. Sisti, Giulio Zanaroli

ENZYMATIC DEGRADATION OF THE MOST COMMON ALIPHATIC BIOPOLYESTERS

Antonella Rosato, Angela Romano, Grazia Totaro, Annamaria Celli, Fabio Fava, Laura Sisti, Giulio Zanaroli

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ABSTRACT

Background information

Massive amounts of non-degradable petroleum-based polymers are being produced worldwide, causing significant plastics accumulation in the environment. Biodegradable plastics have thus received a lot of interest, as several polyester-type plastics have high susceptibility to enzymatic hydrolysis and microbial degradation and show properties often comparable to conventional plastics (Larrañaga and Lizundia, 2019; Urbanek *et al.*, 2020). In this study we investigated the degradation ability of several hydrolytic enzymes belonging to different subclasses (i.e. lipase, esterase, proteinase, etc.) against some common aliphatic commercial polyesters, namely poly(butylene succinate) (PBS), poly(butylene succinate-co-adipate) (PBSA), poly(caprolactone) (PCL), poly(lactic acid) (PLA) and poly(propylene carbonate) (PPC). A comparative study was performed in terms of degradation rate and mechanism of hydrolytic degradation, to provide additional elements for the design of end-of-life management strategies for biodegradable plastic wastes or the design of innovative self-degrading plastic materials, in a circular bioeconomy perspective.

Acknowledgements: The financial support by H2020 project TERMINUS contract No. 814400 is greatfully acknowledged.

2. RECYCLING OF NON-FIBRE FRACTION OF CARTON PACKAGES Åsa Olsson

RECYCLING OF NON-FIBRE FRACTION OF CARTON PACKAGES

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ABSTRACT

Tetra Pak packages consist of a paperboard structure for mechanical robustness and a multilayer laminate consisting of different polymers and Aluminium that serves as the essential barrier for food protection. The high-quality virgin fibre part is very attractive for paper mills recycling carton packages. After fibre recovery, these mills also generate the residual non-fibre fraction called PolyAl. This material consists of different polymers and Aluminium foil which can be mechanically recycled as a composite material (Aluminium and polymer mix) to produce pellets. This is done on industrial scale (close to 100.000 tons/year) today. Pellets like these can be used in different applications for injection moulding. Some products made of PolyAl are pallets, crates, flowerpots, office equipment, dispensers and tissue holders.

Enhanced mechanical recycling by separation of the different layers is expected to give benefits such as increased purity and recycling into new non-food contact applications. Several companies are offering solutions for separation using different methods for triggering delamination.

The TERMINUS-principle using a pre-made adhesive layer containing enzymes that can trigger delamination on demand is an interesting methodology where the components for separation are already built into the packaging material structure.

Acknowledgements: The financial support by H2020 project Terminus contract No. GA-814400 is greatly appreciated.

3. THE POWER OF NADES - BOOSTING ENZYME STABILITY TOWARDS THERMAL DEGRADATION Simona Varriale , Astrid E. Delorme , Jean-Michel Andanson , Julien Devemy , Patrice Malfreyt , Vincent Verney , and Cinzia Pezzella

THE POWER OF NADES - BOOSTING ENZYME STABILITY TOWARDS THERMAL DEGRADATION

Simona Varriale[1], Astrid E. Delorme[2], Jean-Michel Andanson[2], Julien Devemy[2], Patrice Malfreyt[2], Vincent Verney[2], and Cinzia Pezzella[1,3]
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ABSTRACT

In recent years, natural deep eutectic solvents (NADESs) have gained increasing attention as promising nontoxic solvents for biotechnological applications, due to their attractive properties including low flammability, low volatility, facile preparation, high solvability, compatibility with enzymes and the ability to enhance their activity. In this regard, one of the challenges in the implementation of enzymes in industrial processes is their ability to remain active for a longer time or survive harsh operative conditions, resulting in the need of reengineer enzymes to fine-tune their properties toward the end application.

Betaine-based NADESs at a concentration of 25 wt % in a buffered aqueous solution were used as media to inhibit thermal inactivation of POXA1b laccase and its five variants when incubated at 70 and 90 °C. All the tested laccases showed higher residual activity when incubated in NADES solutions, with a further enhancement achieved also for the most thermostable variant. Furthermore, the residual activity of laccases in the presence of NADESs showed a clear advantage over the use of NADESs' individual components. A computer-aided approach based on molecular docking simulation was performed to understand the role of NADESs in the stabilization of laccases toward thermal inactivation, evaluating the interaction between each enzyme and NADESs' individual components gaining more insights into this structure– function relationship. A correlation within the binding energies between laccases and NADES components and the stabilization of the enzymes was also demonstrated.

These findings establish the possibility of preincubating enzymes in NADESs as a facile and cost-effective solution to inhibit thermal inactivation of enzymes when exposed to high temperatures. The precise combination of interactions and molecule orientation determined a different stabilizing effect for each enzyme, suggesting the possibility to tailor the NADES composition for every enzyme of interest, taking advantage of computer-aided approaches for a preliminary screening of different combinations of NADES' components.

Acknowledgements: The financial support by H2020 project TERMINUS contract No. 814400 is greatly appreciated.

4. ADDITIVE EFFECTS ON CURING ESTER-BASED DEGRADABLE POLYURETHANE ADHESIVES S. Mačiulytė, P. Nemaniutė, D. Bražinskienė, O. Eicher-Lorka, S. Joseph Asadauskas

ADDITIVE EFFECTS ON CURING ESTER-BASED DEGRADABLE POLYURETHANE ADHESIVES

Sandra Mačiulytė, Paulina Nemaniutė, Dalia Bražinskienė, Olegas Eicher-Lorka, S. Joseph Asadauskas [1]
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ABSTRACT

Faster degradation rates of adhesives under environmental conditions could significantly accelerate bioremediation processes of glued items. Ester-based polyurethanes (PUR) are more submissive to hydrolysis and enzymatic degradation than traditionally used polyether PUR. However, adhesives from ester-based PUR are not widely used, partly due to kinetic uncertainties during their polymerization. In this investigation, caprolactone and adipate macrodiols were reacted with difunctional and trifunctional isocyanates, monitoring their polymerization rates by titration, FTIR and viscometry. Temperatures and molar ratios were also varied, leading to unexpected changes in the course of the polymerization. Inclusion of caprolactam, acetate and several other additives affected the curing rates significantly. Better understanding of polymerization kinetics could accelerate the replacement of polyether PUR adhesives with the ester-based ones, which would improve bioremediation efforts of glued waste items.

Acknowledgements: The financial support by H2020 project TERMINUS contract No. 814400 is greatfully acknowledged.

5. COMPLEXITY OF MULTILAYERS, MAIN CHALLENGES AND SOLUTIONS TO REACH THE REALITY OF A SUSTAINABLE INDUSTRIAL PROCESSES, AN SMES POINT OF VIEW Alexandre Fontaine

Complexity of Multilayers, main challenges and solutions to reach the reality of a sustainable industrial processes, an SMEs[1] point of view.

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ABSTRACT

Multi-layers packaging, even if they provide various and functional services, represent a serious preoccupation for a whole part of people all over the world. The complexity of the main generated issues is as significant as the complexity of their own manufacturing.

The complexity of multi-layers packaging is characterized as much by the diversity of multi-layers represented on the market, whether due to: - the nature or diversity of the packaged products; - the different functions that can be provided by the packaging; - their manufacture using different industrial processes and technologies; - or their difficulty in being fully recycled at the end of the chain.

So many steps and challenges which are impacting key actors of a complex chain with the main challenge: bringing recyclable functional packaging to the market. The development of an innovative solution such as TERMINUS, aimed at improving the recyclability of end-of-life multi-layers packaging, will help to meet this challenge in the reality of current industrial processes.

However, this raises many challenges that should be overcome. Firstly, from a technical point of view based on applied research work, and secondly from a more global point of view depending of key actors within the chain, from upstream packaging manufacturers to downstream recycling players.

At this intermediate stage of the TERMINUS Project, the vision of an SME manufacturer of multi-layers packaging and specialized on certain specific markets will highlight some of these challenges to meet the expectations of end-users of packaged products.

Acknowledgements: The financial support by H2020 project TERMINUS contract No. 814400 is greatly appreciated.

6. RHEOLOGICAL IMPLICATIONS OF FUNCTIONAL NANOCHARGES IN POLYURETHANE-BASED ADHESIVES AND THEIR LAMINATION PROCESS M. Ngom, C. Chevalier, S. Mani, Rigoberto Ibarra-Gómez

RHEOLOGICAL IMPLICATIONS OF FUNCTIONAL NANOCHARGES IN POLYURETHANE-BASED ADHESIVES AND THEIR LAMINATION PROCESS

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ABSTRACT

Background

Despite the positive impact of food packaging multilayer films on food wastes reduction and lowering of CO_2 footprint, the unfeasibility to recycle them makes these end in landfills, energy production facilities, and, to a much larger extent, as pollutants of soil, and particularly, sea[1]. This is why, in the project TERMINUS, a strategy to valorise multilayer wastes has been proposed: by incorporating functional materials into a target multilayer's adhesive it is possible to make it degradable, thereby allowing the separation and eventual recycling of the constituent plastic films.

In the context of the TERMINUS strategy, the objective of the present work is circumscribed to address the effect of the filler (encapsulated functional materials) in the rheological properties of polyurethane-type adhesives, its kinetics, and the overall laminating process in a continuous system. In literature, some papers have reported the employment of inorganic fillers in polyurethane adhesives with different but specific goals i.e. temperature stabilization[2], an increase of mechanical properties on use[3], and, pigmentation[4]. Unlike these cases, in TERMINUS, the main purpose of the filler does not obey primarily to the generation of particular structure-property outcomes but to protect the functional material from the processing conditions. However, as referred to in the literature, the sole presence of the filler, particularly nanostructured ones, affects the overall physical properties of the adhesive from rheological to final multilayer mechanical behaviour, making a need for the analysis of the properties' modification. Furthermore, an important particularity of the present work is that the lamination process was carried out in a continuous pilot laminating machine, unlike ones reported in literature where manual batch systems are normally employed. As such, the potential changes in rheological properties and kinetics of the adhesive as a result of the filler effect impacts also the coating operation.

Acknowledgements: The financial support by H2020 project TERMINUS contract No. 814400 is greatfully acknowledged.

7. NANOBIOCATALYTIC DEGRADATION OF DESIGNER POLYURETHANES Patrick Shahgaldian , S. Amirabbas Nazemi , C. Wu , R. Burn, C. Pezzella, S. Varriale and P. F.- X. Corvini

NANOBIOCATALYTIC DEGRADATION OF DESIGNER POLYURETHANES

Patrick Shahgaldian[1], Seyed Amirabbas Nazemi[1], Congyu Wu[1], Reto Burn[2], Cinzia Pezzella[3,4], Simona Varriale[3,4] and Philippe F.-X. Corvini[2]

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ABSTRACT

Background information

Polyurethanes (PUs) represent a major class of polymers used in a variety of commercial products. Among others, PUs are used as thin and flexible adhesives to produce multilayer packaging systems. The high stability of the urethane bond (*i.e.*, carbamate), bridging the building blocks of PUs, endows the resulting materials with enhanced chemical, physical and biochemical stability. This represents a major stumbling block for the biological/biochemical degradation of PUs for the recycling of multilayer materials.

Acknowledgements: The financial support by H2020 project TERMINUS contract No. 814400 is greatfully acknowledged.