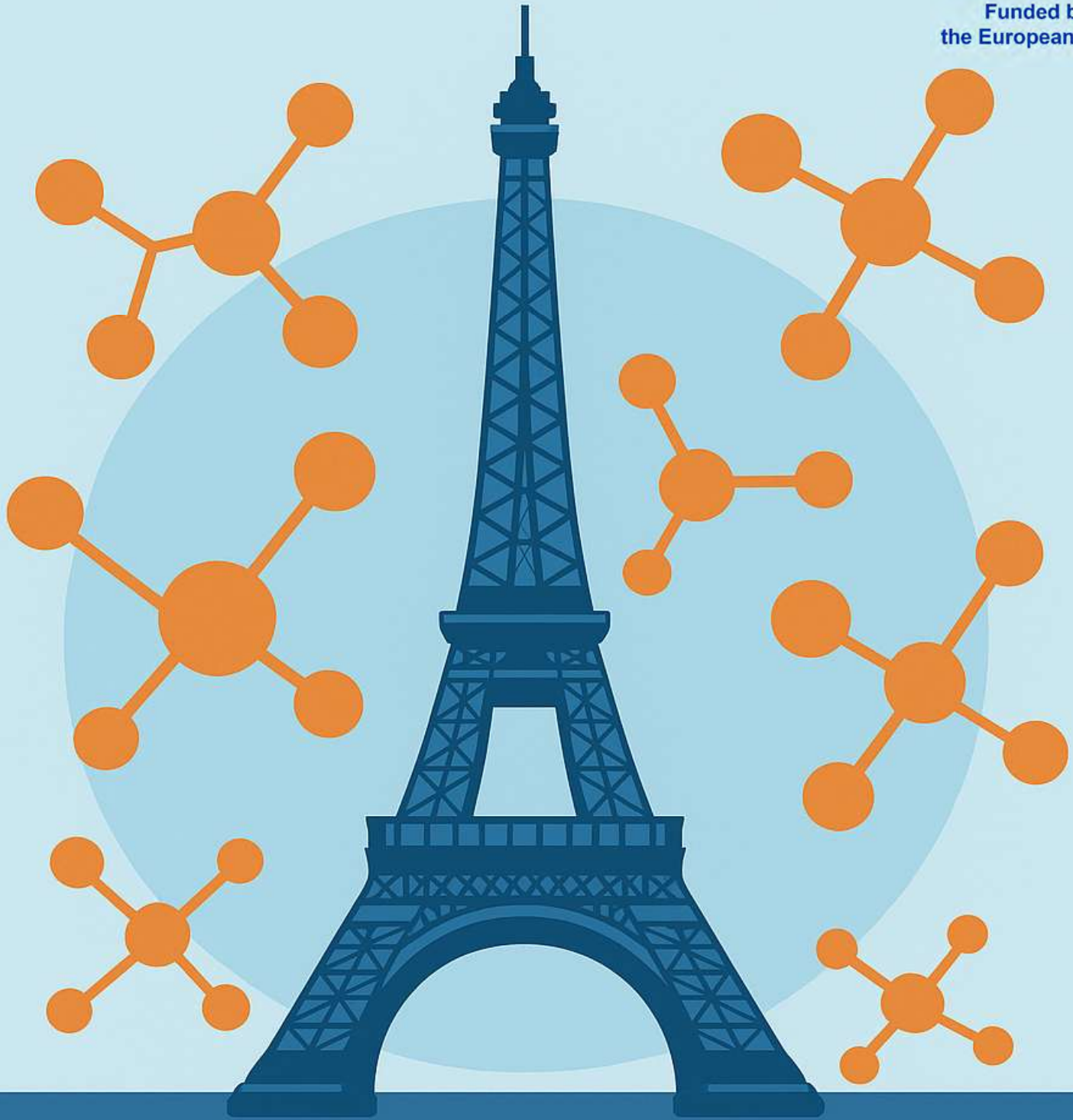
THE REBOND NETWORK

NEWSLETTER

Paris 2025 edition

Funded by
the European Union



ReBond

MSCA DOCTORAL NETWORK

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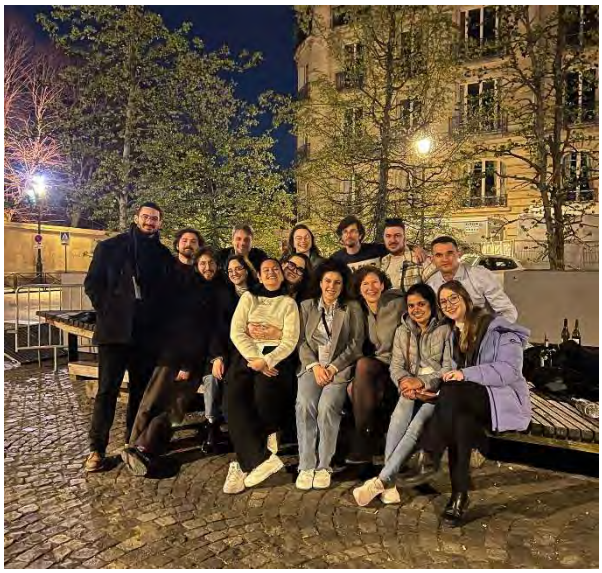
The Meeting: In short

From synthesis to mechanical dynamics of vitrimers:

*During the meeting the first series of advanced training modules for our Doctoral Candidates was performed. PhDs from the C3M group joined by professors **Renaud Nicolaÿ** and **Costantino Creton** to deliver extremely interesting lectures. The lectures focused on giving insight on both synthesis paths for vitrimers and their mechanical behavior, particularly their role in fracture mechanics.*

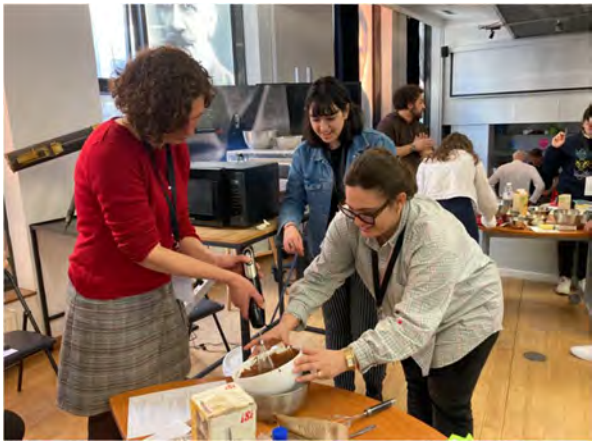


What have we been up to?

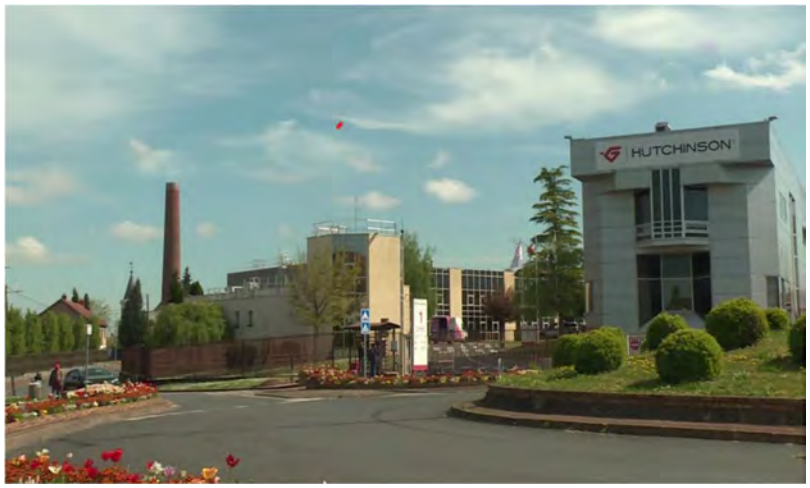
During this meeting the Doctoral Candidates presented their first results achieved in their PhD journey. Follow us in a series of presentations diving in the exciting world of polymers, starting from their laborious synthesis and processing, diving into the ever-evolving field of their dynamics which will be blown wide open through the efforts of both molecular and thermodynamic simulations!
Whatever the field of interest is, we're sure that at least one of our PhD will be able to satisfy your hunger for scientific advancements in the field!

From art to cooking, passing through rubbers:

The meeting wasn't only a moment to share scientific knowledge, but also to experience many new things for the first time. The days in Paris were an incredible amalgamation of sensations and experiences: from the beautiful art and monuments of the city to the delicious food we scientifically prepared and shared together, to the visit of a cornerstone company in rubber processing.
The schedules might have been tight, but we savored every moment of this beautiful experience and now it's finally time to share that same excitement with all of you!



31/03/2025: Visit to Hutchinson

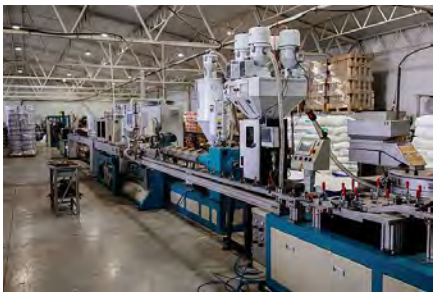


*The first day of the meeting started with a visit to a facility of one of industrial partners, Hutchinson, located in "**Châlette-sur-Loing**". During the day, we were accompanied by "**Laina Guo**" and "**Gaétan Grimaldi d'Esdra**" on a tour of the facility where we were able to see and feel first hand what the rubber industry is all about, while also receiving insight on the scientific side by the many intersting presentations which took place during our stay. The tour started in the production facility, where we observed the process of tyre production, starting from the stocks of raw materials, passing through the assembly line which inevitably led to the vulcanization ovens, one of the most fundamental process in the rubber industry. During the evening the visit moved on to the elastomers and material characterization laboratories, which, as fellow material scientist ourselves, gave us the same feeling as a kid entering in a candy shop! The visit concluded with a detailed tour of the company's showroom, where we were able to observe many of the materials, prototypes and products through which Utchinson built its legacy and sector dominance since it's foundation in 1853, in that same exact facility.*

*The first scientific presentation of the day was delivered by "**Clément Robin**", currently employed in Hutchinson as "Rubber Material Specialist". It focused on the basic principles of rubbers as materials and their rheological behavior, such as the theory of rubber elasticity and the effects of fillers, such as carbon black and hard particles, on the material's mechanical and conductive properties, giving a strong theoretical kickoff into our journey into the world of rubbers and elastomers!*

*The second presentation was delivered by **Florence Lim**, employed in Hutchinson as a "R&I Engineer". Her presentation focused on giving us insight into a very strong and difficult to master analytical technique: Solid State NMR. During her talk we learned of the outstanding range of use of such a technique and how invaluable it can prove in chemical analysis, particularly for insoluble polymers such as vulcanized elastomers, giving a more analytical twist to our growing understanding of rubbery materials!*

*The last presentation of the day was delivered by **Gaétan Grimaldi d'Esdra,** Elastomers and Rubbers R&D Project Manager. His work was focused on how the concepts of non-linearity in elastomers' rheological properties impact industrial applications, displaying for us a series of past and present projects and issues encountered during his career as case studies. His presentation bridged the gap between industry and academia exemplifying how lab scale measurements and observation can be scaled up and applied to truly mastodontic dimensions!*



*The second day of the meeting started our deep dive into the field of polymer synthesis, with a strong focus on vitrimeric materials with Professor **Renaud Nicolaÿ**, who pushed the discussion further on the topic of synthesis of polymers and vitrimers, picking up from where we left off at the previous meeting in Crete. The lecture described different synthesis pathways that can take place for polymeric materials and introduced fundamental parameters to quantify polymerization reactions. Network formation in polymeric materials was thoroughly discussed and explained to us. The lecture culminated with an analysis of step growth polymerization processes, giving us a comprehensive outlook on the field.*

Following the first round of lectures, where everyone warmed up to the more basic concepts regarding the synthesis of polymeric materials, the PhD students from the C3M group, including our own DC1, Matteo Conti, were the hosts of the first round of lectures! Matteo gave a 30-minute lecture on the synthesis of amorphous vitrimers, explaining the theory behind common techniques he uses. The C3M students gave a practical lecture on the theory behind basics of polymerization and analytical techniques, culminating in an interactive moment in which the less chemically savvy DCs were presented with some exercises to solve, testing themselves and enhancing their portfolio of knowledge!

CHIMIE
MOLECULAIRE
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*The concluding act of the day was a two-part lecture from Professor **Constantino Creton**, treating us to a complete overview on the mechanical behavior of polymers, from regular melts to strongly crosslinked systems. The first part of the lecture focused strongly on the concept of stickiness, what are the physical and chemical causes of its insurgence, how it could be quantified through mechanical and rheological means and how it could be exploited to obtain tunable materials such as pressure sensitive adhesives. The second part of the lecture instead focused on the concepts underlying the mechanics of fracture in soft materials, ranging from crack formation regular polymer melts to the effect of dynamic crosslinks in the crack formation and ultimate failure of vitrimeric materials.Prof. Creton's lecture shed light on a whole new field for many of the DCs, further expanding our scientific horizons.*

Project Meeting Presentations

ECPCI, Paris France, 2nd – 3rd April 2025

Guisi Merola, ESPCI / Hutchinson / UCLouvain

Explores the development of vitrimer elastomers by combining static and dynamic boronic ester crosslinkers in alkyl acrylate copolymers. Initial experiments with imine-protected crosslinkers faced challenges: poor crosslinking efficiency, discoloration, solubility issues, and evidence of supramolecular aggregation instead of stable networks. Characterization (DSC, TGA, DMA) showed Tg and thermal stability close, but only static crosslinkers gave stable rubbery plateaus, while dynamic ones failed to form insoluble networks. Next steps involve redesigning protected amines, model polymer studies, and reactive extrusion to improve homogeneity and validate dynamic crosslinking chemistry.

Arya Vijayan, UCLouvain / UOI

Focuses on synthesizing PS-PMMA copolymers with different architectures (random, block, gradient) using RAFT polymerization for compatibilizing blends. Random copolymers were obtained successfully at various molecular weights, but block copolymers showed reproducibility issues and incomplete conversions, while free radical polymerization (FRP) gave more reliable results. The next step is to prepare PS and PMMA vitrimer precursors with dioxaborolane units and use them, alone or combined with vitrimer copolymers, to compatibilize PS/PMMA blends through dynamic covalent crosslinking.

Matteo Conti, ESPCI / UCLouvain

Showcased the synthesis of well-controlled PnBMA and PEHMA vitrimers using RAFT polymerization to achieve homogeneous systems with low dispersity. It details the production of homopolymers and copolymers, followed by RAFT end-group removal and functionalization into vitrimer precursors through condensation with phenylboronic acid. The resulting vitrimers were characterized by DMA, showing tunable properties based on crosslinking density and molecular weight.

Arcangela Russo, UOC / UCLouvain

Investigates the viscoelastic behavior of polystyrene vitrimers, focusing on how dynamic dioxaborolane crosslinks affect their melt and solution properties. It details issues like material instability, side reactions, and the presence of heterogeneities that influence rheological measurements like annealing time and phase locking. The study also explores using these vitrimers as reinforcing agents in blends with higher molecular weight matrices.

Alexandros Andreas Rispo Constantinou, UN / UCLouvain

Develops a mathematical model based on Flory-Huggins theory to predict phase separation in blends of a homopolymer and a dioxaborolane vitrimer. The model explores how factors like the proportion of crosslinked sites, mixture composition, and the interaction parameter influence the stability of the mixture. It concludes that the crosslinker is necessary for phase separation and that too much of it, or a lower overall vitrimer content, makes separation more likely.

Project Meeting Presentations

ECPCI, Paris France, 2nd – 3rd April 2025

Balint Magyari, UN / UOC

Showcased the use of coarse-grained molecular dynamics (MD) simulations to model the rheology of vitrimers. It employs a hybrid MD-Monte Carlo approach to simulate dynamic bond exchange events, which are key to vitrimer behavior. The results show how bond exchange rate influences stress relaxation, chain mobility, and the transition from solid-like to liquid-like flow.

Teresiana Guarino, UOC / UM

Investigates the extensional rheology of polystyrene vitrimers and vulcanized natural rubber elastomers. It compares their behavior under uniaxial extension, showing the precursor deforms ductily while vitrimers fail in a brittle manner, with strain hardening that increases with the applied strain rate. The study also applies the Mooney-Rivlin model to natural rubber and outlines future work on biaxial extension to better understand material behavior under processing conditions.

Anthi Sapouna, UCLouvain / UOI

Presented the degradation of high-density polyethylene (HDPE) through multiple processing cycles and the effect of adding HDPE vitrimers as additives. It characterizes the degraded samples using rheology, differential scanning calorimetry (DSC), and molar mass distribution analysis, finding that degradation reduces relaxation times and slightly affects crystallinity. The future work will focus on understanding how vitrimer addition influences these properties to potentially enhance the recyclability of polyolefins.

Frances Abygail F. Genio, ESPCI / UG

Focuses on developing semi-crystalline vitrimers to improve the recycling of mixed polyolefin waste, specifically polyethylene (PE) and polypropylene (PP). It explores three main strategies: modifying HDPE with dioxaborolane maleimides via reactive extrusion, synthesizing functional polyolefins from hydrogenated polybutadiene, and using azidotriazine grafting agents to compatibilize polymer blends. The work aims to create dynamic networks that enhance material compatibility and enable thermal reprocessing, ultimately upcycling mixed plastic waste into higher-value products.

Claudia Spina, ESPCI / UOC

Investigates the adhesion properties of polystyrene (PS) and poly(methyl methacrylate) (PMMA) based vitrimers. It explores the use of dioxaborolane metathesis reactions to create dynamic covalent bonds at the interface between otherwise immiscible polymers. Preliminary Double Cantilever Beam (DCB) tests compare the fracture behavior of standard PS-PMMA interfaces against vitrimer-modified ones (vPS-vPMMA), suggesting potential changes in adhesion mechanics and crack propagation. The work aims to leverage vitrimer chemistry to enhance interfacial adhesion in polymer blends.

Project Meeting Presentations

ECPCI, Paris France, 2nd – 3rd April 2025

Stefano Chiani, UOI / ESPCI

Studies how incorporating azidotriazine-based vitrimers affects the structure and dynamics of high-density polyethylene (HDPE). It finds that increasing vitrimer content reduces crystallinity and lowers the glass transition temperature, enhancing molecular mobility within the amorphous regions. These changes are attributed to vitrimers disrupting crystal packing and promoting crystal thickening at lower temperatures.

Michail Danikas, UOI / UOLeeds / UM

Investigates the molecular dynamics of vitrimers, focusing on poly(n-butyl methacrylate) (PnBMA) and poly(2-ethylhexyl methacrylate) (PEHMA). It analyzes how molecular weight and crosslink density influence thermal behavior (glass transition temperature increases with both factors), viscoelastic properties (stronger networks and topology freezing with more crosslinks), and dielectric dynamics (merging of α- and β-relaxations into an αβ-process). Overall, crosslinking restricts chain mobility, raises Tg, and alters relaxation dynamics, with effects more pronounced at higher molar mass.

Konstantina Lyroni, UM / UOC

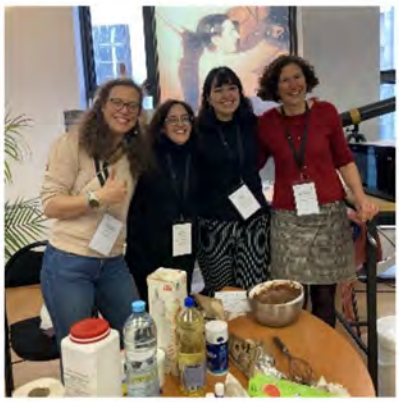
Showcased the development and testing of a multispeckle light scattering (MSLS) setup to study the dynamics of polymeric materials and vitrimers. The setup was successfully validated using Brownian particles and initial tests on a PnBMA homopolymer, revealing a long relaxation time of 4 hours at room temperature that decreased at 40°C. The research aims to apply this technique to characterize the spontaneous and driven dynamics of vitrimer samples provided by the ReBond network.

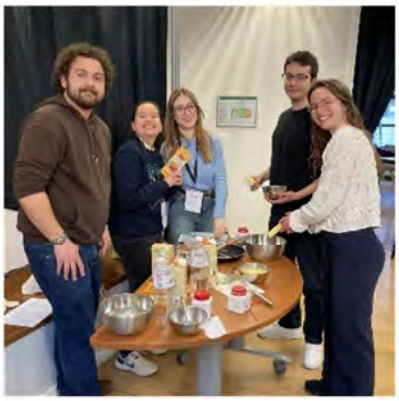
Anahita Karimi, UCLouvain / UG

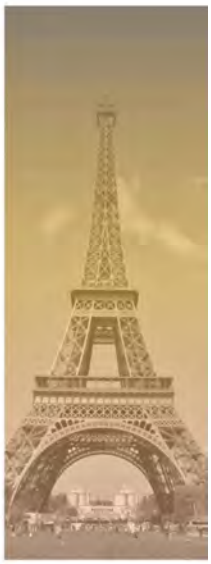
Presented how dynamic covalent bonds, introduced via two different cross-linkers, affect the rheological and thermal properties of polyethylene (PE) vitrimers. It finds that cross-linker density and nature significantly influence network formation, creep resistance, and crystallization behavior, with faster cross-linkers improving processability. The study also reveals that these vitrimers exhibit enhanced creep recovery and can act as nucleating agents, though crystallinity is reduced, providing a tunable approach to designing recyclable polyolefin materials.

Lorenzo Santori, UG / ESPCI

Investigates the rheology and morphology of immiscible polymer blends (PMMA/PS), analyzing how viscosity ratio affects blend behavior and the use of rheological functions to detect morphological changes. It also presents preliminary results on extruded homopolymer-vitrimer blends, demonstrating the processability of dynamic networks. Key findings include the identification of cocontinuity regions and the influence of blend composition on relaxation dynamics and viscosity.

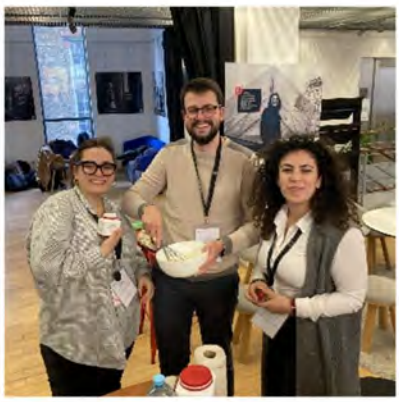


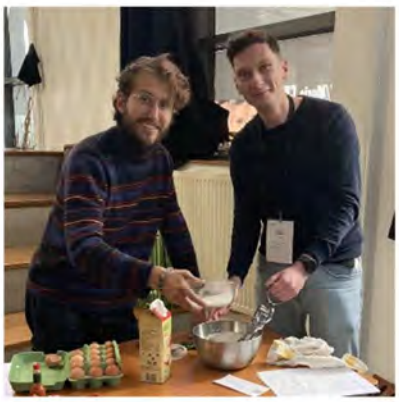




MOLECULAR COOKING

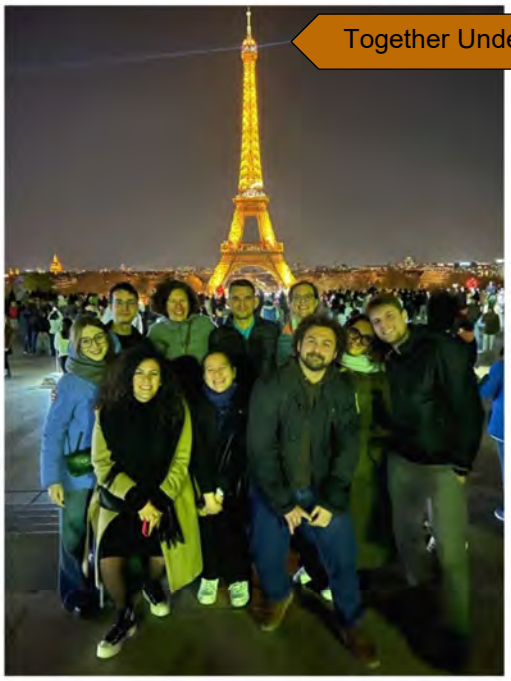
with P. Reutenauer and H. This

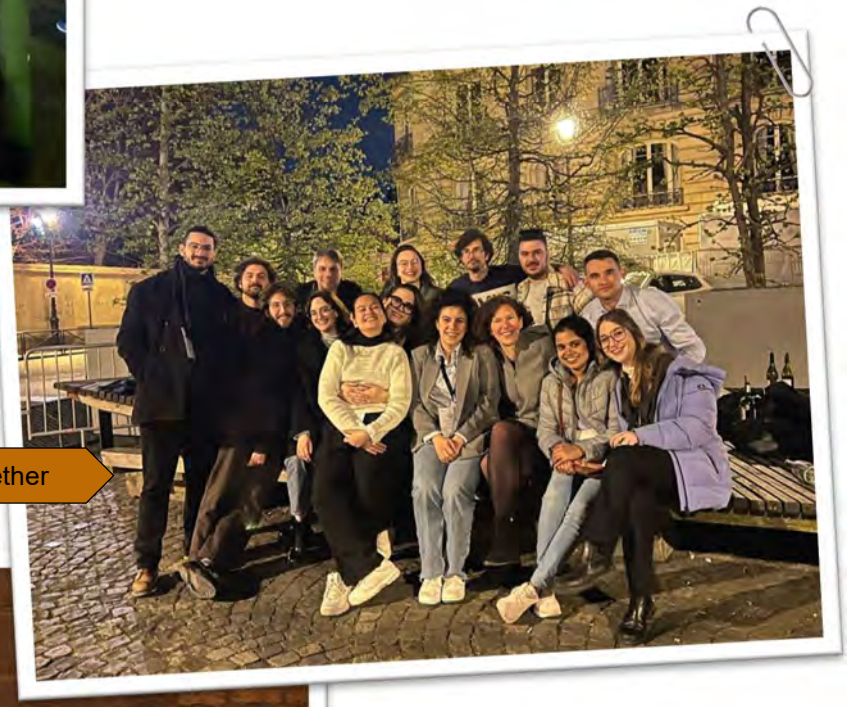




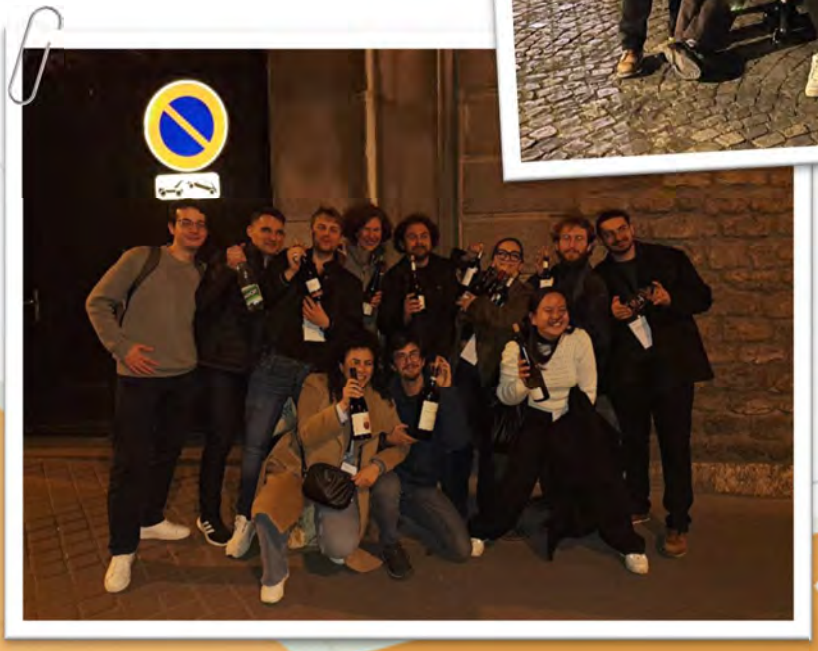
During our Paris meeting, we were introducerd to a unique molecular gastronomy workshop led by Philippe Reutenauer and Hervé This, the French chemist recognized as a founder of this field and pioneer of “note-by-note” cooking. With kitchenware provided by professors and students, we explored how principles of polymer chemistry - gels, foams, and reversible bonds - translate directly into culinary textures and tastes. Reutenauer guided us through the organization and scientific framework, while This demonstrated how chemical understanding can transform gastronomy into a creative laboratory. Together, they revealed how chemistry enriches cuisine, making flavour, texture, and innovation a truly shared experience.

Together Under Paris Lights





Local Wines, Parisian Nights Together

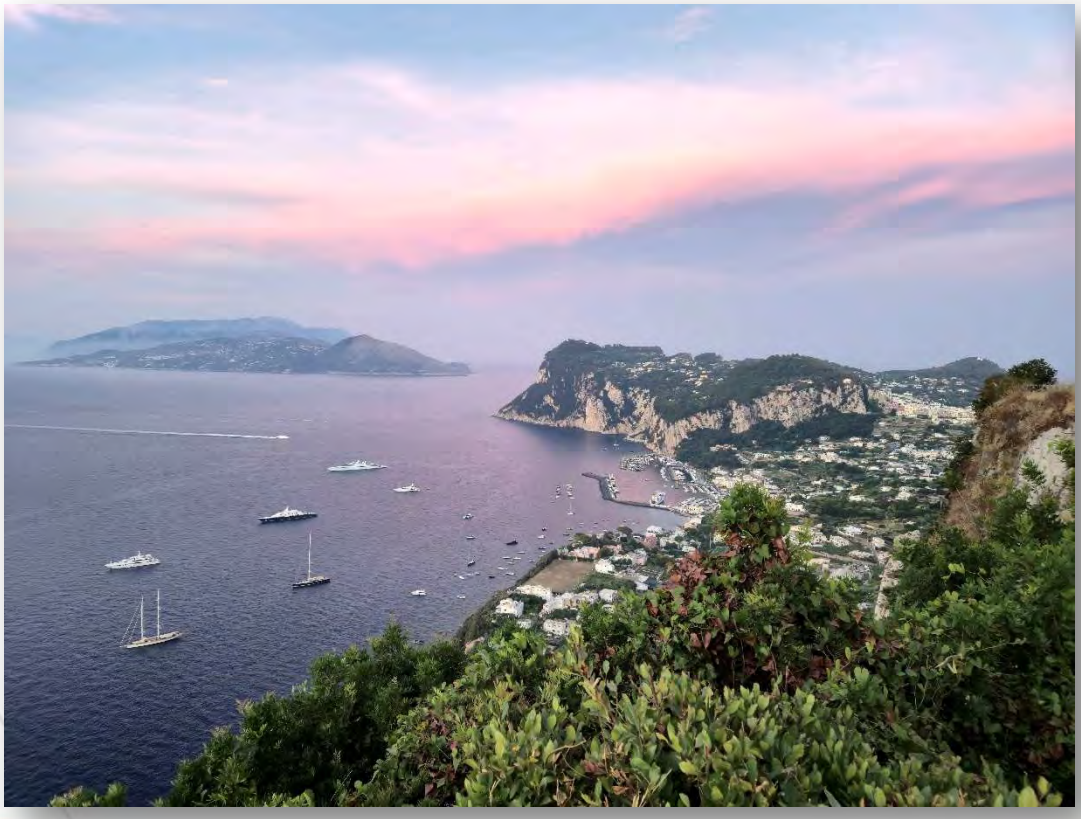


After the formal sessions, we had the chance to experience the city together, exploring Paris by night, sharing laughs, and building stronger connections on a more personal level. It was the perfect mix of work and connection, leaving us inspired and even more united as a team.

Summer School on Polymeric Networks for Sustainable Development

7-12 July, 2025 – Anacapri (Capri, Italy)

The next ReBond gathering will take us to the beautiful island of Capri for a vibrant summer school. This event will bring together not only members of our project but also students from outside Rebond, creating a wider community of exchange and learning. Participants will have the chance to hear from leading voices in polymer physics and related fields, gaining fresh insights into cutting-edge research. In addition, an industrial workshop will be held, where our partners will introduce interactive approaches to integrating vitrimers into production lines - bridging the gap between research and real - world applications.



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