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Executive Summary

Deliverable 2.4" Public Report" is the culminating document that aims to provide a concise overview of all the work and results obtained from the iCAREPLAST project.

This document has been structured into four sections. The first section provides an overview of the project, its results and the impacts achieved as a result of the work carried out during the time of the project. Section 2 presents the context of the project and a detailed overview of the objectives achieved. Section 3 describes the work packages, the main results obtained and the deliverables presented in each of them. Section 4 shows the main innovations obtained with the project and finally section 5 describes the different impacts achieved by the iCAREPLAST project.

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1. Executive Summary

1.1 Context and objectives

Approximately 70% of European plastic waste (18.5 mt/year) is not being recycled due to technical or economic reasons and are thus sent to landfill (27%) or incinerated (42%). This situation affects negatively the environment in terms of pollution and greenhouse gases emissions, as well as social perception regarding waste management, consumer's products industry and policy makers.

The aim of the project iCAREPLAST is to provide a cost and energy-efficient alternative to recycle and valorise non-recycled plastic waste that, due to their characteristics or their contamination, are currently disposed into landfills or underexploited through energy recovery. The process combines pyrolysis, catalytic treatment and membrane separation technologies to obtain high added-value chemicals, as they are aromatics (BTXs and medium to long-chain alkylaromatics), that can be used for the production of virgin-quality polymers or as raw materials for other processes in petrochemicals, fine chemicals and surfactants industries. To ensure the efficiency and sustainability of the process, advanced control techniques are applied that aim at harmonising economic and environmental targets, making use of meaningful indicators defined taking into account LCA and LCC analyses. Hydrocarbon-rich side-streams are recovered for energy valorisation through oxy-fuel combustion integrated with CO2 capture, improving energy sustainability and avoiding greenhouse gas (GHG) emissions. The valorisation of by-products (char and CO2) contributes to economic sustainability of the process and avoids waste generation.

1.2 Work Performed

During the 54 months of work and the completion of all activities of the project work packages, all proposed objectives were achieved. The waste plastic raw materials were fully characterised both macroscopically and microscopically, and the pre-treatment operations were designed accordingly. The waste plastic mixture was optimised and selected to achieve high liquid yields with an adapted molecular composition. Numerous thermal and catalytic pyrolysis tests were carried out in different reactors resulting in high liquid yields and thus maximising the content of the products. Several commercial membranes were examined to assess their hydrocarbon separation capability and promising separation factors were obtained in the laboratory-scale model mixture. Also, the suitability of the membranes was demonstrated using a model mixture representing the mixture that could be obtained after pyrolysis or alkylation. Alkylation, aromatization, and oxycombustion tests have been carried out at lab-scale, and the materials, catalysts and optimised conditions were selected. The integration of the pre-treatment line and the solid extraction unit with the existing pyrolysis pilot plant at Urbaser CiAM facilities, the modifications of the catalytic area to adapt the catalytic reactor to the aromatization process and the installation of a bench-scale distillation unit were carried out. In addition, pyrolysis tests under different conditions were also performed to investigate their effect on the reaction rate, product distribution, and product quality. A durability test was also carried out, demonstrating that the pyrolysis technology is highly stable in operation and consistently produces high-quality pyrolysis liquids with a constant composition. A pilot-scale aromatization test was also carried out using the light fraction distilled from the pyrolysis liquids produced under optimal conditions and using an industrial zeolitic catalyst based on ZSM-5 (BioBTX - EXT1). Furthermore, the steady state model of the entire iCAREPLAST process was completed thus, plant status and operation can be well represented by this model. The LCA models were completed to assess environmental sustainability and LCC to include cost parameters. Market and techno-economic analyses of the production of linear alkylbenzenes (LAB) and potential commercialization of BTXs, CO2 and char as by-products process were carried out.

In terms of non-technical achievements, the strong interaction between the WPs and the partners is worth mentioning. This allowed different communication and dissemination activities of the

project and its results to be carried out. All partners participated in several events, conferences and fairs where they presented the iCAREPLAST work in posters and oral presentations. At the same time, training activities were developed involving different students who carried out their Master, PhD and Postdoctoral activities within the framework of iCAREPLAST. Other relevant activities were the different scenarios that were defined to determine the economic conditions, design the business plan and the business cases in order to make the business viable. A feasibility assessment, barrier and stakeholder analysis were also carried out. A patent analysis was carried out for freedom of enterprise compliance. Nine innovations have been developed. Last but not least, iCAREPLAST is integrated into the future scenario of plastic circularity, being the Project Coordinator a leading member supporting the Plastics Circularity Multiplier group.

1.3 Progress beyond the state of the art

The project aims to demonstrate the whole technology for plastic waste valorisation in a pilot plant able to process >100 kg/h of plastic. Advanced upstream waste sorting, pre-treatment and pyrolysis is strongly backed by the demonstration activities, know-how and commitment of the consortium. iCAREPLAST solution will enforce circular economy by substantially increasing the amount of recycled plastics to produce commodity products that can be used for virgin-quality polymers production or as raw materials for other industries.

iCAREPLAST offers a more efficient and sustainable chemical process utilising nowadays nonrecycled plastic waste as starting material. Compared with existing chemical recycling technologies for plastic wastes, iCAREPLAST will contribute to: (i) improve pyrolysis liquid yield up to 12% (increment), which with the selected plastics mixture, at the pilot scale, the liquid yields obtained ranged from 76-84%; (ii) reduce energy requirements up to 45%. It has been shown that when energy criteria are considered, the pyrolysis process can be run saving approximately 0.36 MJ/kg of treated plastic and obtaining similar liquid pyrolysis yields. Similar conclusions have been obtained for the joint operation of pyrolysis plus alkylation, where a trade-off solution gives an energy required of 1.8 MJ/kg of plastic and a yield of alkylaromatics of 0.4 kg/kg, whilst if the alkylaromatic yield is maximised 0.5 kg/kg with 2.7 MJ/kg of energy is required. Preliminary models including oxycombustion give a very optimistic view in which the energy balance of the entire iCAREPLAST process is negative (meaning that energy is recovered); (iii) reduce residues production up to 95%. Residue production from pyrolysis step is estimated less than 0.05 %. In addition, the membrane separation tests carried out are key aiming to maximize the products yield and to minimize the residues production of the integrated iCAREPLAST solution; and (iv) increase economic yield up to 200%.

Moreover, the iCAREPLAST process will contribute to reduce CO2 emissions for treatment of plastic waste compared to conventional incineration. When considering also the substitution of virgin by secondary materials, the impact of the recycling process can even reach below net zero level. Additionally, the application of the iCAREPLAST recycling process will decrease the use of fossil resources. The developed life cycle engineering models also show that the production of the delivered materials (benzene, toluene and styrene) can compare with the production of the same materials from primary resources in the impact category of climate change. The environmental assessment of the recycling process can be carried on continuously in the future through the developed Live LCA framework and can be used as a decision support tool for further process optimization.

As a result of its initial exploitation 250,000 t of plastic waste has been treated and valorised, which otherwise would have disposed into landfill, converting it into 1,500 t of alkylaromatics and 1,000 t of aromatics. Additionally, liquid and gaseous streams comprising hydrocarbons and CO2, and solid by-products (char) have been recovered and valorised to maximise material and energy balance of the overall process, thus minimising environmental footprint and ensuring economic sustainability.





iCAREPLAST will expand the spectrum of plastic waste types that can be effectively recycled to almost every plastic. Thus, when no restrictions are put to the plastic materials that can be recycled, citizens will receive a confidence message that will reinforce their involvement in the recycling chain. This citizen engagement will contribute to the European targets for recycling 65% of municipal waste and 75% of packaging waste by 2030.

2. Project context and overall objectives

2.1. Project context

iCAREPLAST is a four-year, six-month project that aims to provide a cost and energy-efficient alternative to recycle and valorise non-recycled plastic waste (ca. 70% of European plastic waste) that, due to their characteristics or their contamination, are currently disposed into landfills (27%) or underexploited through energy recovery (42%). iCAREPLAST project, summarized in Fig.1, combines pyrolysis, catalytic treatment and membrane separation technologies to obtain high added-value chemicals, as they are (alkyl-)aromatics (BTXs and medium to long-chain alkyl-aromatics), that can be used to produce virgin-quality polymers or as raw materials for other processes in petrochemicals, fine chemicals and surfactants industries.



Figure 1. The overall concept behind iCAREPLAST process

To achieve the project's objective, the iCAREPLAST consortium has been crucial for the smooth running of all activities. Thus, the iCAREPLAST consortium brings together a group of ten partners (see Fig. 2) from five European countries (**Spain**, the **Netherlands**, the **United Kingdom**, **Germany** and **Portugal**), which brought together a complementary and outstanding set of experiences, skills, competencies and resources that have enabled the achievement of the innovation objectives, the design of an efficient and sustainable integrated recycling process, as well as the successful communication, dissemination and exploitation of the project results.



Figure 2. iCAREPLAST consortium





2.2. Overall objectives

To reach the main objective of the iCAREPLAST project, thirteen specific objectives (O) were established. During the fifty-four (54) months of the project, the work carried out by the iCAREPLAST consortium allowed each of these specific objectives to be achieved. The work carried out to achieve each of them is described below.

O1. Characterisation of plastic waste streams in terms of flow, macroscopic composition (polymer distribution), microscopic composition (additives, fillers, etc.), seasonal fluctuations, and physical characteristics (format, shape, strength, etc.). These characteristics will be used in O2. to design pre-treatment and feeding processes and in O3. to optimise composition of feed mixtures.

In the context of WP3, plastic waste feedstocks from URB treatment plant were fully characterized. Parameters such as CHNS, chlorine presence, humidity, volatile matter among others were analysed on white and coloured LDPE streams, PP and PS. Thermo-gravimetric analysis (TGA) of LDPE, PP and PS fractions was also performed. Macroscopic characterization of plastic waste feedstocks was carried out. These data were essential in the design of the pre-treatment line in O2.

Completion degree of O1 is 100%.

O2. Identification of pre-treatment operations and design of feeding systems to feed the waste raw materials in optimum conditions of quality into pyrolyser at a rate of 100 kg/h. Physical characteristics analysed in O1. for each different stream will be taken into account.

Physical, chemical and compositional data of plastic waste feedstocks determined in O1 were used to define the most suitable ranges of composition and morphology for an optimal feeding of waste streams into the pyrolyser. The fulfilment of O2 includes the identification of plastic waste treatments for cleaning from organics, metals and abrasive materials, and mechanical actions such as shredding and densification to facilitate feeding. The pre-treatment operations have been successfully identified, and the pilot plant was fully designed, while the units' assembly, building and integration stage was successfully carried out. Assembly and Installation Certificate and commissioning were performed.

Completion degree of O2 was 100% at M36.

O3. Optimisation of feeding mixtures. Taking into account the characterisation obtained from O1, different mixtures of plastic wastes, including mainly PE, PP, PET and PS, and lower amounts of other plastics, such as PVC and polycarbonates, as well as fibre-reinforced or other composites, will be tested. The objective is to identify composition of mixtures that maximise the amount of aromatics and olefins obtained at the pyrolysis step and are as much coherent as possible with the polymer distributions identified in O1.

The most relevant point achieved in O3 was the choice of the best plastic waste mixture to produce high liquid yields with the most suitable composition to be used in WP4 processes. Finally, a mixture of 80 % of LDPE, 10 % PP and 10 % PS (wt. %) was selected taking into account the major contribution of plastic film in a future scenario in which recovery of PP and PS reaches a significant but minority contribution. The values for PP and PS contribution have been chosen considering the current recovery ratio PP/PS 1:2, the PP content in the recovered waste plastic film and keeping the contribution of PS in a 10 % according to the original selected mixture (30 % LDPE, 60 % PP and 10 % PS). In order to assess if the pyrolysis technology is adequate to process other kinds of plastics and what is its influence on the product yields and composition, tests with small incorporation of PET and PVC were tested. In addition, a procedure based on simulation and multiobjective

optimization was explored in WP5, where optimal mixtures of plastics wastes have been selected. Specifically, when the amount of olefins and aromatics are maximized at the pyrolysis step. *Completion degree of O3 is 100%*.

O4. Optimisation of operational parameters of pyrolysis reactor. Adjustment of temperature, pressure, and flow rate, decision between thermal or catalytic pyrolysis, and selection of catalyst (if any), to achieve a minimum liquid yield of 85 wt.% and maximise aromatics and olefins content in the products after the pyrolysis step.

Several pyrolysis tests were performed in three different batch reactors to achieve the goals of WP3. After assessing the most favourable experimental conditions and taking into consideration the operation window of URB plant, it was decided to use 440 °C during 30 minutes to test different plastic mixtures. These operational conditions were chosen considering the products' yields and composition as well as the characteristics of the pyrolysis plant in URB. Futhermore, many actitvities were developed to compare the thermal and catalytic pyrolysis. Both catalytic and non-catalytic pyrolysis can influence the product distribution to a certain extend by altering the feedstock composition, temperature, residence time, pressure and operation mode. A scale and type of pyrolysis reactor effect was observed. In addition, a procedure based on simulation and multiobjective optimization was explored in WP5, where optimal pyrolysis step when the liquid yield is also maximized.

Completion degree of O4 is 100%.

O5. Design of separation processes to selectively remove oxygenates and bulky hydrocarbons from pyrolysis products. Due to the nature of the substances to be removed, a two-step membrane separation process is required. Membrane-based extraction using selective solvents will be used to remove oxygenates from the primary pyrolysis products whereas the heavier hydrocarbons will be separated from light aromatics and olefins by solvent-resistant nanofiltration membranes.

The presence of oxygenates turned out to be minor due to good pre-sorting of the plastic waste. Therefore, O5 focused on the removal of heavier hydrocarbons from aromatics and olefins. Various commercial membranes were screened in order to assess their separation capabilities; promising separation factors were obtained on lab scale model mixture. Validation on real pyrolysis liquids indicated severe fouling resulting in the lack of membrane performance. A systematic study into membrane performance suggested this could be attributed to sorption of various impurities.

Completion degree of O5 is 80%.

O6. Optimisation of operational parameters of alkylation reactor. Adjustment of temperature, pressure, flow rate, and selection of catalyst, to maximise conversion of aromatics and olefins into alkylaromatics, achieving a minimum yield of 95%. The products obtained from the alkylation reaction will also depend on the mixtures entering the reactor, which will be conditioned by feeding mixtures identified in O3., operational parameters of the pyrolyser identified in O4. and the separation process designed in O5.

Different studies were carried out for optimizing both catalyst and main parameters of alkylation reaction: (1) optimization of alkylation reaction parameters (contact time, aromatic/olefin ratio, T, P, etc.) by using selected commercial zeolites as catalysts (H-MOR, H-USY and H_MCM22); (2) effect of transition metals incorporation into the zeolites on the alkylation performance, also including industrially-practiced H2 feeding; (3) effect of zeolite modification (mesopores creation) on the catalyst performance and stability; and (4) catalytic deactivation studies (long time tests), including catalyst regeneration and reuse. Lab-scale experiments performed at optimal reaction





conditions with modified H-MOR zeolite provided high mono-alkyl-aromatics selectivity (>90%) and high catalytic stability (TOS >30 h). In addition, a procedure based on simulation and multiobjective optimization has been explored in WP5, where optimal plastic mixture and operating temperatures for pyrolysis and alkylation reactors was analysed. Specifically when the amount of alkylaromatics are maximized.

Completion degree of O6 was 100% in M36.

07. Design of membranes to separate alkylaromatics from mixtures obtained after the alkylation reaction. Commercially available tight ceramic nanofiltration membranes will be required to separate long-chain alkylaromatics from the mixture (olefins, paraffins and unconverted light aromatics). In case the cut-off of the commercial membranes is insufficient, organics groups will be grafted into the pores to tune these membranes to the targeted alkylaromatic molecules. The feed composition from the alkylation reactor will depend on parameters identified in O3 to O6. A minimum purification yield of 99% must be achieved.

Activities related to O7 were fully aligned with those related to O6 and the new aromatization step (see O8). The selection carried out regarding separation selectivity associated to molecular weight in pyrolysis liquids can be directly used in the selective separation of (bulky) alkyl-aromatics, from mixtures exiting the alkylation reactor. Aromatic reactor mixture composition was determined in cooperation with BBTX and membrane characterization experiments using lab-scale setup initiated. As a result of the potential switch of alkylation reactor to aromatization reactor, various (pre-)commercial membranes were screened to assess their performance, but 99% purification could not be obtained. A start was made to develop new organic solvent reverse osmoses membranes, which were promising in terms of selectivity but exhibit low permeances. Permeances were so low that they were not practical for purification purposes. Nonetheless, successfully nearly defect free torlon based hollow fiber membranes were developed.

Completion degree of O7 is 70%.

O8. Optimisation of operational parameters in hydrotreatment for final hydrogenation of unsaturated alkyl-chains to improve the quality of the final products (stability, viscosity, colour, etc.). Adjustment of temperature, pressure, flow rate, and selection of catalyst in the process, to process the alkylation reaction mixture with the aim of getting complete saturation of aromatic substituent chains in >90% yields and meet the final product specifications.

The activities proposed in O8 were not carried out as originally scheduled. The experiments performed in WP3 and WP4 revealed that a hydrotreatment step is not necessary to get complete saturation of aromatic substituent chains (alkyl chains) in >90% yield, for the plastics mix selected. The first part of deliverable D4.3 detailed, in any case, the state-of-the-art commercial catalysts for hydrotreatment processes. In addition, and due to the results of pyrolysis of plastics obtained in WP3, increasing for aromatics production is now envisaged for the overall benefit of the process.

D4.3 introduced the concept of the **aromatization unit** and described the state-of-the-art catalysts. For that purpose, studies on catalytic aromatization over metal-zeolites based catalysts were made by comparing cataltysts developed at CSIC, BBTX and commercial catalysts. Promising results in terms of BTXs and other mono-aromatics production by converting alkanes/alkenes mixtures mimicking different pyrolysis liquid compositions were achieved at CSIC (lab-scale). In addition, catalytic tests of distilled pyrolysis liquids from 80LDPE10PP10PS pyrolyzed a the pilot plant are being carried out. The pilot plant scale catalytic aromatization experiment is still pending. *Completion degree of 90%.*

O9. Optimisation of operation parameters of the distillation column. Adjustment of number of plates and temperatures to fraction the different alkylaromatics obtained in the process. Targeted products will depend on parameters identified in O3 to O8. A separation yield per fraction > 95% must be achieved.

The bench-scale distillation unit with a capacity of 20 L and maximum number of theoretical plates ~50 was installed at URB facilities. Optimization of the reflux ratio and the cut temperatures to obtain the chosen fraction was performed, once the targeted inlet product to the aromatization stage was determined (light fraction) and samples from pilot testing campaigns was available to make operational adjustments. Finally, a separation yield of about 55 % (wt.%) was achieved (hydrocarbons with boiling points under 290 °C).

Completion degree of 100 %.

O10. Design of oxyfuel combustion units with CO2 capture for energy valorisation of hydrocarbonrich side-streams obtained at different points of the process: gases obtained at the pyrolysis step (O4.), oxygenates and other hydrocarbons discarded at the separation before alkylation step (O5.), and part of the unreacted substrates and diluents obtained after alkylation step (O7.). Taking into account the mass balance between main streams and side-streams, it is seen that the higher the energy obtained from side-streams the lower the final products yield. The objective will be to find the trade-off value that maximises percentage of energetic self-sufficiency (thus minimising environmental footprint) while ensuring economic sustainability.

It has been achieved the design and manufacturing of, first, a single cell stack in order to validate the materials, operation modes and testing setup, and, second, a 3-cell stack assembly working in fuel cell oxycombustion mode that generates oxygen and reforms an incoming current of CH₄ in order to obtain pure CO₂ and generate electricity. The final intended 20-cell stack at a 700 W power level was not achieved due to the multiple challenges found during development and testing of more than 20 prototypes. In any case, the developments achieved paves the way to identify cost effective industrial solutions that can be implemented in the future.

Completion degree of O10 is 75%.

O11. Identification of efficiency and sustainability indicators, and real-time optimisation and control of integrated operation. The optimum operating conditions for the demonstration plant will be determined as part of the process design (O1.-O10.). However, during plant operation, the optimum process conditions may change frequently e.g. due to changes in equipment availability, economic conditions, and process disturbances. Hence, the optimum operating conditions need to be re-calculated on a regular basis. This control activity is defined as real-time optimisation (RTO). RTO utilises the plant operating conditions as variables to predict properties such as product characteristics. A suitable optimisation problem statement needs to be formulated and solved, usually by developing two models: (a) the operating model and (b) the economic (or sustainability) model. The operating model (a) is a steady-state process model and contains all process variable constraints. The objective is determining the optimal process control parameters for real-time operation. The economic model (b) depends on an objective function including costs and expected product values that either needs to be maximised or minimised for overall optimisation. Therefore, the optimal operating parameters determined by the operating model (a) will be fed into the economic model (b) to determine if the suggested set of parameters leads to an economically feasible recycling process or if the set of parameters needs further optimisation. Only after the second optimisation through the economic model (b) the set of parameters represents the overall/global process optimum, allowing for maximum productivity, minimal operational costs, energy and resource efficiency and low environmental footprint.





The overall steady-state model of iCAREPLAST process is finished thus, plant status and operation can be well represented by this model (D5.1). The LCA models to assess the environmental sustainability and the LCC to include the cost parameters are completed. These models and their methodological approaches were developed in Task 6.2 and listed in MS8. Scenarios based on process parameters have been identified and analysed within the environmental models and are presented in D6.3. Further, jointly with URB, six sustainability indicators have been identified as very important for the process and a procedure for communication between optimizer application and LCA-LCE application defined and tested. Therefore, the architecture developed is ready to feed the plant status to the LCA application and get the sustainability KPIs and take them into consideration for integrated operation.

Completion degree of O11 is 100%.

O12. Pilot plant demonstration and integration of individual processes. Individual feeding, pyrolysis, alkylation, hydrotreatment and separation processes designed and optimised according to previous objectives (O1-11) will be integrated into a pilot plant able to process > 100 kg/h of plastic waste with a minimum 70% yield of transformation into alkylaromatics. This pilot plant is located at the Centre of Technological Innovation "Alfonso Maillo" (CiAM), in the city of Zaragoza (Spain), and it is operated by the waste treatment and management company URB.

The activities related to O12 developed until M56 included the integration of the pre-treatment line and the solid extraction unit with the existing pyrolysis pilot plant at URB CiAM facilities and the modifications of the catalytic area to adapt the catalytic reactor to the aromatization process. In terms of the pre-treatment line, the main activities carried out include: i) completion of the line with the acquisition, reconditioning, and assembly of a second-hand agglomerator and an industrial water cooling equipment; ii) pre-commissioning and commissioning activities obtaining the Assembly and Installation Certificate, the Low Voltage Installation Certificate and the Occupational Risk Assessment; and iii) performance tests obtaining the Ready for Operation Certificate and fulfilling KPIs. The performance tests have also been carried out for the solid extraction system, validating KPIs and getting the Ready for Operation Certificate. The results of the trials carried out in the pilot plant are collected in the deliverable D7.6 and have allowed the study of the influence of main factors in pyrolysis process: feedstock, pressure, and reactor level. A durability test was also conducted using the selected plastic waste mixture (80LDPE10PP10PS) under optimal cracking conditions (5 barg, 425 °C, and 55 % level). The thermal degradation behaviour presented a high operational stability with a constant production of high-quality pyrolysis liquids (results are reported in D7.7). A pilot scale aromatization test was also carried out with an industrial zeolitic catalyst based on ZSM-5 (BioBTX - EXT1) and using the light fraction distilled from the pyrolysis liquids produced under optimal conditions as liquid feed. These results are collected in the deliverable D7.7.

Completion degree is 100 %.

O13. Characterisation of products and valorisation of by-products. Alkylaromatics obtained from the operation of the pilot plant (O12.) will be characterised to ensure compliance with market specifications. Solid char obtained from the operation of the pilot plant at the pyrolysis step will be characterised and assessed for its use as filler in composite manufacturing. Other applications, like its use as gas absorbent, will be explored.

Market and techno-economic analyses of the production of linear alkylbenzenes (LAB) using a combination of polyethylene, polypropylene and polystyrene waste and potential commercialization of BTX as by-products from the LAB production process were carried out in Task 8.1. Market prices obtained in the market analysis were used to assess the economic viability of the

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process, which is presented in D8.1 and D8.2. The potential utilisation of solid by-products from the pyrolysis process was carried out in Task 8.2, and results presented in D8.3. *Completion degree of O13 is 100%.*





3. Progress and achievements at WP level

For the development of the project, the iCAREPLAST consortium structured the work plan in ten different work packages (WP), whose activities have been developed and reported in the forty-three deliverables that have been submitted to the European Commission, thus fulfilling the ten milestones (MS) of the project (see Fig. 3).



Figure 3. Structure of iCAREPLAST Work Plan and Milestones

- WP1. Ethics requirements.
- WP2. Project management and coordination.
- WP3. Validation of key enabling technologies in upstream processing.
- WP4. Validation of key enabling technologies in downstream processing.
- WP5. Plant modelling, control and optimisation.
- WP6. Life cycle engineering
- WP7. Pilot plant engineering commissioning and testing.
- WP8. Characterisation of products and by-products and their integration into existing markets.
- WP9. Communication, dissemination and training.
- WP10. Exploitation and business plant.

Each work package was structured in different tasks that were developed by the project partners throughout the 54 months of project implementation. The following section will explain the objective of each of these work packages (WP), as well as the main key results achieved with the development of all activities and tasks and the deliverables submitted for each WP.

3.1 WP1. Ethics requirements

WP Leader	CSIC		
Duration	M1-M54	WP Team	UPV, TUBS, IPT, LNEG, BBTX, IC, UT, KER, URB
WP Objective			

This work package sets out the 'ethics requirements' that the project must comply with.

WP Key Results

Information was collected in D1.1 on ethical issues that may arise in the project and provides guidance on how they are managed within the iCAREPLAST project. The report indicates the consortium's ethical procedures in relation to management, research activities (research integrity, environment, health and safety aspects), data management and dissemination. It indicates the legal framework and ethical requirements in the URB pilot plant, including Regulations and Standards (with all certifications attached), Corporate Social Responsibility and URB's Quality, Prevention, Environment and Energy Policy.

M6

Deliverables

D1.1. EPQ-Requirements No.1





3.2 WP2. Project management and coordination

WP Leader	CSIC		
Duration	M1-M54	WP Team	UPV, TUBS, IPT, LNEG, BBTX, IC, UT, KER, URB
WP Objective			

The aim of this work package is to ensure an efficient project execution leading to the achievement of iCAREPLAST project objectives. The project management has been implemented to assure: Intensive, flexible and open dialog among the partners concerning key scientific and technical issues; Rapid and effective decision-making on technical and organisational issues; Compliance with EC administrative and reporting requirements; Implementation and management of the technological infrastructure; Effective risk management and project quality.

WP Key Results

The results achieved with the development of WP2 are as follows:

- Day-to-day consortium coordination and communication.
- Organisation of online progress technical meetings.
- Organisation of face-to-face plenary meetings and the two iCAREPLAST workshops.
- Progress activity reports and cost statements requested and revised half-yearly.
- An amendment to extend the project 6 months (budget neutral) was requested and approved by the EC, mainly due to changes needed due to COVID-19 outbreak impact on the project activities and readjustment of budget and efforts.
- Continuous revision of the management and the quality plan (D2.1) established measures.
- Control the timeline and status of all the deliverables using the iCAREPLAST shared folder. Revision and submission.
- Validation of all the milestones achievement.
- Risks analysis updated in D2.3 (M18, M24, M36 and M48). Due to the fast implementation of mitigation actions (if needed), the project tasks and objectives were completed giving high-quality results

Deliverables	
D2.1. EPQ-Requirements No.1	M6
D2.2. Progress activity and management reports with cost statements according with EU rules	M12; M24; M36
D2.3. Risk Report	M12; M24; M36 M48
D2.4. Public Report	M54

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WP Leader	LNEG		
Duration	M1-M36	WP Team	CSIC, UPV, TUBS, IPT, BBTX, IC, KER, URB
WP Objective			

The main purpose of WP3, is to validate the different upstream processes needed for the adequate treatment and transformation of plastic mix (essentially derived from MSW and end-of-life composites) into mainly profitable liquids together with gases and solids by using thermal (and catalytic) pyrolysis. The main objective of the WP is to provide the required experimental data by treating real materials and process characteristics to enable WP7 activities (design, commissioning and operation of individual units at TRL>6) and process modelling and control activities (WP5-WP6). The overall process for obtaining the desired pyrolytic liquids involves the following major steps: (i) the selection/sorting and characterisation of the plastic raw materials; (ii) the pre-treatment and adequate feeding of the plastic into the pilot plant of plastic waste valorisation; (iii) thermal and catalytic pyrolysis processes; and (iv) the analysis and characterisation of the liquid products, as well as side-products (gases and carbon/char). All these steps are contemplated in the WP validation activities and summarised in the following tasks: Definition and characterisation of waste streams; Design of pre-treatment operations of raw materials; Thermal and catalytic pyrolysis.

WP Key Results

The key results achieved with the development of the five tasks of WP3 are the following:

- Selection/sorting and characterization of the plastic raw materials.
- Assessment of the pre-treatment and adequate feeding of the plastic into the pilot plant of plastic waste valorisation.
- Study the thermal pyrolysis processes to optimise the operating parameters and test the optimised thermal pyrolysis parameters on individual plastic waste in two reactors with different operating modes.
- Tests with different mixtures of plastic waste with the aim to identify the ratio olefins/aromatics suitable for the alkylation reactions in WP4 in order to choose the most adequate plastic waste mixture.
- Thermal pyrolysis tests with small incorporation of PVC and PET.
- Studies of the effect of experimental conditions used in the thermal pyrolysis of the plastic waste mixture selected (PE, PP and PS).
- Experiments with catalytic pyrolysis of individual plastics and with the plastic waste mixture selected. Thermal and catalytic pyrolysis tests with small incorporation of cardboard, metals and organic matter.
- Production of aromatics out of mixed plastic waste via thermal catalytic pyrolysis proven.
- Aromatization catalyst optimization studies performed.
- Analyse and characterize the liquid products, as well as side-products (gases and carbon/char).
- Round-robin of composition analysis of URB pyrolysis liquids obtained with the selected plastic waste mixture.

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		I U		C 3

D3.1. Report on selected feed streams characterization	M24
D3.2. Report on selected pre-treatment procedure that enables a proper and continuous feeding of the selected plastic mix into pyrolyser	M24
D3.3. Report on optimal feeding composition and operation parameters for thermal pyrolysis step	M24
D3.4. Report on optimal feeding composition, best catalyst options and reactor operation parameters for catalytic pyrolysis step	M24
D3.5. Report on comparison between thermal and catalytic pyrolysis	M30 M48
D3.6. Report on main indicators for up-stream process control	M30





3.4 WP4. Validation of key enabling technologies in downstream processing

WP Leader	CSIC		
Duration	M6-M46	WP Team	UPV, TUBS, IPT, LNEG, BBTX, IC, UT, KER, URB
WP Objective			

The main objective is to technically validate different unitary processes at lab/bench scale to provide data and models to (a) accurately design the corresponding units/sub-processes and (b) establish the plant operation protocols for the commissioning and efficiency optimisation. The processes under study in this WP are: Catalytic upgrading of pyrolytic liquids via alkylation of aromatics with olefins over solid catalysts; Fractionation and separation of (i) light hydrocarbons (\leq C5) and (ii) oxygenated molecules (acids, phenols, etc.) from pyrolytic liquids; Fractionation and separation of aromatics and alkylaromatics final products; Separation and catalytic hydrotreatment (if needed) of hydrocarbons side-streams; Gases treatment, oxycombustion in oxygen transport membrane (OTM) reactors and CO₂ capture.

WP Key Results

The main results achieved through the completion of the four WP4 tasks are as follows:

- Catalyst screening and selection of the best catalyst using model and complex mixtures and subsequent testing and optimisation/modification of catalysts by incorporation of metals, creation of mesopores, etc. Assessment of the activity and stability of the catalyst at long reaction times (deactivation control) and evaluation of the catalytic aromatisation process and selection of the best catalyst.
- Bench-scale testing rig built and operative for organic membrane separation. In membrane separation screening, good separation of heavier hydrocarbons from model liquid mixtures at ambient temperature has been achieved. Membrane characterisation on synthetic and real pyrolysis products and performance testing was also carried out using a laboratory-scale configuration to determine the operating window and fouling potential. Finally, alternative membrane systems (polymeric/ceramic) were prepared and tested.
- Selection of materials (catalysts and oxygen selective membrane) and operation mode for the
 oxy-combustion unit. Design and manufacture of components for the small-scale OTM module
 and subsequent validation of the experimental set-up for the oxy-combustion tests of the
 medium-scale OTM module. This was followed by the assembly of full-scale modules for
 medium-scale experimentation. Then a system analysis and detection of critical points of design
 and operation was performed.
- Final operational conditions optimized to improve BTXs yield in aromatization and assessment of catalyst activity and stability (with pyrolysis liquids).

Deliverables	
D4.1. Report on catalyst selection for catalytic alkylation	M18
D4.2. Start-up of research apparatus for testing membrane separation technologies	M18
D4.3. Report on catalyst selection for catalytic hydrotreatment	M28
D4.4. Report on optimal feeding composition and operation parameters for catalytic alkylation/aromatization step	M46
D4.5. Report on membrane separation units for pyrolysis and alkylation reactor products including choice of membrane and operating conditions envelop.	M36
D4.6. Report on oxyfuel combustion of fuel gas integrated with CO2 capture	M36
D4.7. Report on main indicators for downstream process control	M36

3.5 WP5. Plant modelling, control and optimisation

WP Leader	UPV		
Duration	M1-M54	WP Team	TUBS, CSIC, IPT, IC, URB
WP Objective			

Move the operating point of the process towards its optimum taking into account simultaneously different objectives or specifications related to safety, environmental, plant economics, energy use, etc. This goal will be reached using different methodologies related to control engineering and optimisation.

WP Key Results

The main results obtained from the work carried out in WP5 are as follows:

- Development of the steady state model for the overall iCAREPLAST process based on blocks or sub-processes that can be managed independently (pyrolysis, alkylation, aromatisation and oxycombustion). It will be useful for decision-making in process operation: from experimental data to simple steady-state models, using Aspen plus and fitting techniques (polynomial regression and genetic algorithms) ready for use in optimisation algorithms.
- Application of multi-objective optimisation techniques to obtain optimal operating points of a
 process taking into account several criteria simultaneously related to yields, energy and
 sustainability. At the same time, state several multi-objective optimisation problems based on
 the metamodels developed and to solve these problems to obtain optimal operating points of
 the different process units, such as the optimal operation of the alkylation, aromatisation and
 oxy-combustion units.
- Development of dynamic models of different iCAREPLAST sub-processes for use in advanced control techniques. This has made it possible to show how advanced control can improve pyrolysis performance through dynamic simulations using dynamic models and multivariable predictive control.
- Design of a proposal that combines production data with sustainability indicators to select the optimal operating point of the iCAREPLAST process taking into account both criteria: plant economics and environmental issues.
- Settle and dispose all data generated during simulations, creating and publishing a dataset of iCAREPLAST process, under License: Creative Commons Attribution-NonCommercial (CC-BY-NC).
- Design and develop a communication architecture between applications to share data related to the plant status and the LCA indicators related to this status. Test the communication architecture using simulated plant data with the full LCA model developed in WP6 for real-time LCA purposes. Finally, design a proposal for mixing real-time pilot plant data with simulation data for real-time LCA.

Deliverables	
D5.1 . Report on the complete plant simulation and digital-twin model building	M24
D5.2. Report on advanced control system specifications. Tool for multivariable predictive control	M48
D5.3. Report on real time	M54
D5.4. Report on Real Time LCA specifications. Interface for Real-time LCA optimisation	M54





3.6 WP6. Life cycle engineering

WP Leader	TUBS		
Duration	M1-M48	WP Team	UPV, IPT, URB
WP Objective			

This WP aims to develop an integrated methodology for the live assessment of life cycle environmental and cost assessment of secondary chemical commodities from the recycling of plastic containing products. This integrated approach includes models depicting both the background system (e.g. considering regional differences in infrastructure, weather, collection systems and electricity mixes, etc.) and the foreground system (technological development inside iCAREPLAST from a process, machine and factory perspective). The methodology to be developed within this work package includes the following objectives: (i) Definition of product system, software requirements and architecture; (ii) Development of background and foreground system models, and life cycle engineering models; (iii) System evaluation: sensitivity (elasticity) analysis regarding process parameter threshold values; (iv) Development of interpretation and visualisation techniques and interfaces.

WP Key Results

- Identification of necessary data to perform an LCA static and in real time.
- Definition of the architecture for the computational models and to capture and transfer the process data in real time.
- Development of a framework for live LCA (incl. technical requirements).
- Parameterized inventory model building for all processes within the iCAREPLAST process chain and for required materials (e.g. membranes).
- Integration of cost parameters into LCA/LCC model and basic evaluation.
- Scenario analysis of LCA models by variation of input parameters (e.g. temperature, pressure, plastic waste mix composition) and background systems (energy provision).
- Design and development of a Guided User Interface for iCAREPLAST pilot plant managers providing appropriate results visualisation, analysis tools and an overview of the environmental parameters.

Deliverables

D6.1. Description of the product systems, software requirements and architecture	M12
D6.2. Computational models describing material and energy flows. Integrated computational Life Cycle Engineering models and live LCA software system/tool (to be implemented in the pilot plant)	M36
D6.3. System evaluation: report of system behaviour in extreme scenarios	M50
D6.4. LCA- and LCC-data interpretation and adequate visual analytics interfaces	M50

3.7 WP7. Pilot plant engineering, commissioning and testing

WP Leader	URB		
Duration	M6-M54	WP Team	TUBS, IPT, UT, KER

WP Objective

The main objective is the demonstration of the technology at pilot scale under industrial-relevant environment (at least TRL-7). This objective entails secondary objectives related to the design, construction, assembly and commissioning of different units and subprocesses. The core of the technology will be the demonstration in the pilot plant at URB facilities of the transformation of the plastic mix into refined and stabilised alkylaromatics.

Apart from this central piloting objective, other lateral but crucial technologies will be demonstrated ex-situ to reduce the demonstration activity, allow for higher pilot plant flexibility complexity and warrantee the objective accomplishment in a shorter timeline. These two ex-situ piloting activities comprise (a) technology on advanced separations for organic separation and fractioning (UT); (b) oxycombustion technology to produce heat to partly satisfy the plant demands and a CO2 stream product, limiting GHG emissions.

WP Key Results

As a result of the work carried out in WP7, the following main results were obtained:

- Plastic waste pre-treatment line: Detailed engineering, building, mechanical and electrical
 assembly of the agglomerator; definition of the safety protocols and the operating procedure;
 PLC/SCADA programming; equipment calibration and operation adjustments; consumption data
 integration in the OPC project; commissioning of the whole line in progress; and characterization
 of the LDPE agglomerated material; acquisition and integration of a new industrial water cooling
 equipment; pre-commissioning and commissioning tests of the agglomerator completed; and
 performance tests of the whole pre-treatment line completed.
- Solid extraction system: Detailed engineering, main equipment purchasing; equipment manufacturing; skid assembly and definition of working principles (normal working condition, start-up and safe stop); electrical and mechanical integration in the pilot plant; definition of the plant operation control logic, control loops and key sequences, safety interlocks and alarms; PLC and SCADA programming; commissioning tests without material and completion of tests by using a simulated reactor content (thermal oil).
- Pilot plant operation: Pyrolysis tests under different conditions to investigate their effect on the reaction rate, product distribution, and product quality completed. Analysis of experimental results and characterization of samples ongoing. A durability test was also carried out demonstrating that the pyrolysis process has a high operational stability.
- Catalytic tests: a) pilot tests: commercial catalyst procurement completed; b) lab-scale tests using industrial catalysts completed; pilot-scale test using a commercial catalyst completed.
- Design on advanced membrane module for the selective separation of alkylaromatics; detailed engineering, HS3 studies and 3D modelling of the membrane module of selective separation of alkylaromatics; Construction of the membrane module pilot setup for the selective separation of alkylaromatics
- Oxycombustion module design, with components, materials and dimensions; balance of Plant (BoP) for the pilot testing of modules under oxycombustion conditions assembled, including the required turbomachinery and heat-exchange systems; module validated for 1 single cell; all materials and components validated; 3-cell stack tested for oxycombustion conditions.
- The separation of a complex pyrolysis oil mixture derived from the pyrolysis of plastics was
 tested with commercially available OSN membranes. However, for the complex pyrolysis oil
 mixture studied here, no commercially available membranes are suitable to perform the desired
 separation. It is believed that taking a holistic approach and tuning the pyrolysis process more
 to the suitability of the OSN membranes, e.g. adding an integrated catalytic step, could
 potentially resolve this. Similarly, an alternative route could be to combine OSN with other





separation methods like fractionated condensation and/or distillation. By combining these with membrane technology, the energy requirements for separation are likely to be reduced.

Deliverables	
D7.1. Drawings of new and modified units	M15
D7.2. Protocols and procedures for plant operation	M30
D7.3. Report of the pilot plant commissioning results at URB facilities	M48
D7.4. A pilot scale membrane module suitable for testing preferred membrane technology	M51
D7.5. Ceramic membrane oxycombustion reactor, composed of a set of membrane units, assembled and sealed, and provided with covers, tubbing and thermal insulator, with an electric power of 700 W	M54
D7.6. Report of plant operation tests	M54
D7.7. Report of operation optimisation and durability test	M54

3.8 WP8. Characterisation of products and by-products and their integration into existing markets

WP Leader	IC		
Duration	M18-M48	WP Team	CSIC, TUBS, LNEG, BBTX, UT, KER, URB

WP Objective

The main objectives of this WP are: (i) Determination of the key markets for the main alkylbenzene products and specification of detailed product composition required to reach those key markets; (ii) Characterisation of by products from the pyrolysis (char, pyrolysis gas) and synthesis (oxygenates, and other hydrocarbons) processes; (iii) Determination of potential uses for these by-products through reprocessing in the plant or as product streams.

WP Key Results

The main results obtained from the work carried out in WP8 are as follows:

- Identification of several pyrolysis-based value chains.
- Assessment of the potential of the plastics pyrolysis route.
- Techno-economic analysis of the production of alkylbenzene and other industrially applicable chemicals that can be derived from the pyrolysis of plastics; Further development of the techno-economic analysis of the main products and by-products.
- Assessment of the potential of carbons from the pyrolysis process for various uses.
- Study of the integration of CO2 into existing markets.
- A high carbonaceous content was achieved in the solid products from the pilot plant, the main use of the solid products would be in providing energy for the pyrolysis process.
- The current percentage of the CO2 employed in its different uses compared to that emitted is so low that there is still a very high potential to increase its use. A complete study was performed on the identification of optimal pathways for CO2 utilisation.

Deliverables	
D8.1 . Report on technoeconomic analysis on the markets for alkyl-benzenes and their derivatives	M40
D8.2. Identification of optimal by-products utilisation	M48
D8.3. Identification of optimal char utilisation	M48
D8.4. Identification of optimal paths for CO2 utilisation	M48





3.9 WP9. Communication, dissemination and training

WP Leader	KER				
Duration	M1-M54	WP Team	CSIC, UPV, TUBS, IPT, LNEG	, BBTX, IC, UT, URB	
WP Objective					
The aim of this work package, led by the Dissemination Manager is to ensure the proper communication and dissemination of project outcomes. The main WP objectives are: i. Communicate the benefits of the iCAREPLAST process for Plastic Recycling and Plastic					
Waste Man and its pote economy; ii Disceminate	Waste Management Industries, as well as for Chemical and Petrochemical Industries, and its potential impact over plastic waste mitigation and contribution towards circular economy;				
iii. Develop lea disseminate generation	 Disseminate results of the project, technological innovations achieved and their potential impact over other processes or industries; Develop learning resources, and use them for training and academic activities, to disseminate major innovation outcomes of iCAREPLAST project to the current and next generation of employees in the sector of Process Industries, promoting their awareness 				
W/D Koy Results	Inplementation				
 The main results from the Create the project's communication active Video of the project Organisation and experse 2022 a perspectives-iCAREP Soriano Technology Four (4) conference are currently under Five (6) posters and disseminate the project's website Several press release the project's website Development of lear material was upload 	e development website and so vities and disse produced and ecution of two and (ii) Work LAST Project" a Park, Zaragoza proceedings w preparation. d thirteen (30) ject and the res ave been produces have been me (see annexe l arning resource to the project	t of WP9 are a ocial networks minate projec put online by (2) project wo shop "Plasti at the Alfonsc on January 2 vere published oral present sults of iCARE uced and pub ade on differe II). es: preparatio	as follows: (LinkedIn, Twitter and YouT ct news and events through t Arpil 2020 orkshops: (i) "live demonstrat c Recycling Strategies, Cu Maíllo Innovation Centre, Ic 023. d (see annexe I) and seven (7 rations were presented at c PLAst (see annexe II). lished in the iCAREPLAST we ent platforms, within the part on of presentations and sho Tube iCAREPLAST channel and	ube), and promote hem. tion LCA" online, in rrent status and ocated in the López 7) research articles lifferent events to bsite. thers' websites and ort videos. All the nd is being used as	
 educational material by the academic partners' in their university lessons. Four (4) Post-docs, seven (7) PhD students and fifteen (18) Master students have worked in the framework of iCAREPLAST and 4 Educational training activities were developed. iCAREPLAST has taken part of the future scenario of plastic circularity in Europe and led different activities in both, Plastics Circularity Multiplier Initiative and SusPlast Platforms. The project appeared in the list of Horizon 2020 projects contributing to Zero Pollution Action Plan. 					
D9.1. Project communica	ation and disse	mination plar	n (PCDP)	M2	
D9.2. Communication m and press clippings, proj	aterials, includ ect website and	ing newslette d intranet	rs, project related videos	M6; M12; M18; M24; M30; M36 M48	
D9.3. Report on monitorinactivities	ng and evaluatio	on of commun	ication and dissemination	M48	

D9.4. Learning resources to be uploaded to the project website

M48

3.10 WP10. Exploitation and business plan

WP Leader	URB		
Duration	M6-M54	WP Team	CSIC, UPV, TUBS, IPT, LNEG, BBTX, IC, UT, KER,
WP Objective			

The aim of this work package is to establish the exploitation and business plan for the exploitation and use of the project results, paving the way for wider exploitation of iCAREPLAST process and associated technologies and boosting innovation, as well as manage the IP rights among consortium members of the project.

WP Key Results

The main results from the development of WP10 are as follows:

- A techno-economic analysis of the alkylation/aromatization and the oxy-combustion processes.
- SWOT analysis of the iCAREPLAST technology.
- Definition and review of the Key Exploitable Results (KER).
- Definition and review of the Intellectual Property Rights (IPR) in connection with one of the patents associated with the KER.
- Definition of business scenarios.
- Stakeholder mapping.
- Characterization of the iCAREPLAST's innovations, including particular risks.
- Analysis of barriers for a hypothetical plastics chemical treatment plant.
- Elaboration of the iCAREPLAST Exploitation Plan where the business model analysis is included.

Deliverables

D10.1. Data Management Plan	M6; M18; M36
D10.2. iCAREPLAST Exploitation and Business Plan, including IPR strategy and	M30
agreement	M36





4. Project main innovations

The development and completion of the iCAREPLAST project resulted in nine innovations. These innovations are listed below

	Title of the Innovation
Innovation 1	Novel pre-treatment line for post-consumer plastic mixtures
Innovation 2	Novel char extraction system
Innovation 3	The Product Sustainability (iPPS) web application designed to provide plant managers with a near real-time analysis of the environmental performance of the plant.
Innovation 4	Novel separation unit for aromatic products
Innovation 5	Novel combustion unit for the oxycombustion of side stream gases
Innovation 6	Novel alkylation catalyst formulation
Innovation 7	Pyrolysis of plastic waste mixtures into aromatics: catalytic process proven
Innovation 8	Control architecture to maximize liquid pyrolysis yield, manipulating pyrolysis temperature and plastic mix composition of PE, PP and PS.
Innovation 9	Real Time monitoring LCA application

5. iCAREPLAST Impact

Expected Impacts	Expected Impacts KPIs Description		
	KPI-1.1. Pyrolysis liquid yield, measured as mass of liquid products obtained per mass of treated plastic.	At the pyrolysis step a minimum liquid yield of 85wt.% will be achieved at the final design.	At lab scale, many pyrolysis tests were performed in three different batch reactors. Thermal pyrolysis experiments with the selected plastic mixture achieved a liquid yield ranging from 70 to 95 wt.% for the different operational conditions studied. At pilot scale, liquid yields obtained in the range of 76 – 84 wt. % depending on feedstock, pressure, and reactor level. Note that with the optimal reactor level, the production rate could be tripled compared to other scenarios.
Expected Impact 1. More efficient and sustainable chemical process utilising plastic waste as starting material for the production of added value products such as chemicals but excluding fuels.	KPI-1.2. Net specific energy required, measured as energy consumed (in MJ) per mass of plastic treated.	Improvement of at least 40% in comparison with current processes (40MJ/kg), reaching a maximum value of 24MJ/Kg for treated plastic.	Development of sub-process models (aromatisation, independent alkylation, oxy- combustion, pyrolysis, and alkylation). Their development includes modelisation of energy consumed in kWh per kg of treated plastic. Relationships between plastic mix and operating conditions with energy consumed were adjusted for future use in WP5 optimization activities. Some preliminary results about optimal operating points selection related to minimisation of energy consumption have been obtained. It has been shown that when energy criteria is considered you can operate the pyrolysis process saving approximately 0.36MJ/kg of treated plastic and obtaining similar liquid pyrolysis yields. Similar conclusions have been obtained for the joint operation of pyrolysis plus alkylation, where a trade off solution gives an energy required of 1.8MJ/kg of plastic and a yield of alkylaromatics of 0.4kg/kg, whilst if you want to maximize the alkylaromatic yield you will obtain 0.5kg/kg with 2.7MJ/kg of energy required. Deeper analysis would be needed to adjust the oxycombustion model, preliminary results give a very optimistic viewpoint in which the energy balance of the complete iCAREPLAST process is negative (meaning that energy is recovered).
	production, measured as mass of residues	A maximum of 0.1% specific residue	formed by oligomers partially cracked and char can be used as







	produced per mass of treated plastic.	production will be achieved.	bitumen, residue production from pyrolysis step is estimated less than 0.05 %. In addition, the membrane separation tests carried out are key aiming to maximize the products yield and to minimize the residues production of the
	KPI-1.4. Product purity (%), to ensure cost efficiency.	Purity achieved in final products will be at least 85%, according to market stock standards, that will be monitored and by tasks developed in WP8.	integrated iCAREPLAST solution. As membrane separation was unsuccessful to purify the streams after conversion, these purity were not achieved by membrane separation. Nonetheless, as one considers BTX as the product, a product purity of 85% is achievable with integrated catalytic pyrolysis.
Expected Impact 2. The technologies proposed should provide a decreased utilisation of primary fossil resources in the process industry of at least 30%.	KPI-2.1. Primary fossil resources usage measured in kilograms of oil equivalent (koe) required per kilogram of products obtained.	A maximum of 1 koe/kg plastic will be achieved, which means a decrease of around 60% compared with aromatics manufacturing from fossil feedstock.	Life cycle engineering models showed that the production of the delivered materials (Benzene, Toluene and Styrene) can have an impact of 0.69 kg CO2-Eq. per kg alkylaromatics (baseline scenario) in the climate change category. In this case, iCAREPLAST achieves an improvement from 58% up to 76% for each product type delivered (Benzene, Toluene and Styrene) when compared to the virgin raw materials. Depending on the mix of plastic waste input into the iCAREPLAST process, along with variations in the temperature and pressure in pyrolysis and alkylation, the output quantities of each delivered material can be further optimized.
Expected Impact 3. The concepts proposed should provide a decrease in CO2 emissions of at least 20%.	KPI-3.1. CO2 emissions in kg of CO2 per kg of treated plastic waste.	A maximum of 1.3 kg of CO2 per kg of treated plastic waste will be achieved, which, compared with incineration of the same waste, means a decrease of at least 50%.	The treatment of mixed plastic waste by iCAREPLAST can reach an impact of 0.6 kg CO2-Eq. and less per kg treated waste, depending on the chosen process parameters. The consideration of secondary product substitution (the production of the delivered materials through the iCAREPLAST process, as opposed to their production from virgin fossil fuels), can carries the recycling process below the net zero level. In addition to the climate change impact category, other environmental impact categories such as resource depletion (fossil) and particulate matter formation perform around zero or below zero, when including credits from the production of the delivered materials (Benzene, Toluene, Styrene). This means the recycling process offsets its environmental burdens with the potential credits from these added-value chemicals. The results of life cycle assessment

			studies list iCAREPLAST as a competitive plastic treatment option among others. This was further proven and optimized with scenario analyses of varying mixed plastic waste composition and varying temperatures and pressures during pyrolysis, alkylation and oxycombustion processes to determine the changes in the composition of the delivered materials.
Expected Impact 4. The concept should utilise at least 70% of waste material including at least 40% of plastic waste.	KPI-4.1. Percentage (weight based) of waste material used as raw material.	A minimum of 95% of waste material will be ensured.	Feedstock used for the trials was a mixture of waste LDPE, PP and PS in different proportions. That is, a 100 % of waste material.
Expected Impact 5. Effective dissemination of major innovation outcomes to the current next generation of employees of the SPIRE sectors, through the development, by education/training experts, of learning resources with flexible usability. These should be ready to be easily integrated in existing curricula and modules for undergraduate level and lifelong learning programmes.	KPI-5.1. Number of materials developed	At least 3 educational dossiers will be developed in WP9 (D9.4)	Learning resources timely started were developed (as a series of 6 short videos) and uploaded to the project website and social media. In addition, slides for all the presentations were prepared by the academic partners, shown and used in their students' courses.
	KPI-5.2. Number of current and future employees of the SPIRE sector reached, measured as number of students enrolled in courses utilising these materials.	The objective during the project will be to reach 2 students within the countries of all the partners involved in the consortium.	The total number of students who carried out their activities within the framework of iCAREPLAST was 29 (7 in Spain, 8 in the United Kingdom, 2 in Portugal and 12 in Germany). In addition, internships for international students were also promoted, bringing opportunities and valuable exchanges.





Annex I. Open Access Publications



Publications in Scientific Journal

No	Title	Authors	Title of Journal/Proc./Book	Number, Volume	Year	Open Access? Gold/Gre en	DOI	Repository Link
1	Comparative LCA of Municipal Solid Waste Collection and Sorting Schemes Considering Regional Variability	Erkisi, S.; Hagen, J. S.; Cerdas, F.; Herrmann, C.	Procedia CIRP	98	2021	Gold Open Access	10.1016/j.procir.2021.01.036	https://www.sciencedirect. com/science/article/pii/S22 12827121000597
2	Framework for the Life Cycle Assessment of non-permanent process units in volatile chemical recycling process chains	Hagen, J. S.; Erkisi, S.; Wit, P. de; Cerdas, F.; Herrmann, C.	Procedia CIRP	98	2021	Gold Open Access	10.1016/j.procir.2021.01.005	https://www.sciencedirect. com/science/article/pii/S22 12827121000287
3	Validation of the Application of the Pyrolysis Process for the Treatment and Transformation of Municipal Plastic Wastes	P. Costa, F. Pinto, R. Mata, P. Marques, F. Paradela, L. Costa	Chemical Engineering Transactions	86	2021	Gold Open Access	10.3303/CET2186144	https://www.cetjournal.it/i ndex.php/cet/article/view/ CET2186144
4	Catalytic Pyrolysis of Municipal Plastic Wastes (MPW) for the Production of Valuable Chemical Products	P. Costa, F. Pinto, R. Mata, P. Marques, F. Paradela	Chemical Engineering Transactions	94	2022	Gold Open Access	10.3303/CET2294143	https://www.aidic.it/cet/22 /94/143.pdf





Annex II. Communication and Dissemination Activities

Communication of iCAREPLAST project (project promotion):

Communication of iCAREPLAST project - PROJECT PROMOTION						
Name of the Event	Date	Place	Author(s)	Partner	Type of Presentation relevant for iCAREPLAST	
M6-M12						
Going Green - CARE INNOVATION 2018	28-29/11/18	Vienna, Austria	Andreas Schiffleitner	IPT	iCAREPLAST project Presentation.	
IFEMA - ChemPlast Expo	7-9/05/19	Madrid, Spain	Julio García Fayos	CSIC	iCAREPLAST project Presentation.	
Sustainability & Naturals in Cosmetics 2019	14-15/05/19	Berlin, Germany	Niels J. Schenk	BBTX	iCAREPLAST in one slide project introduction in the talk "Sustainable Building Blocks for Cosmetic and Packaging".	
12 th International Conference on Bio-Based Materials	15-16/05/19	Cologne, Germany	Pieter Imhof	BBTX	iCAREPLAST in one slide project introduction in the talk "Full Circularity Enabled: Sustainable, Cost Competitive Production of Platform Chemicals"	
International Plastics Processing & Recycling	19-20/09/19	Berlin, Germany	Marcelo Domine	CSIC	iCAREPLAST project Presentation.	
ISWA World Congress 2019	7-9/10/19	Bilbao, Spain	Ignacio Sanz Madroño	URB	iCAREPLAST in one slide project introduction in the talk "The future of plastics. Plastic to"	
4 th Chemelot InSciTe Annual Meeting	8-9/10/19	Horst, The Netherlands	Pieter Imhof	ввтх	iCAREPLAST in one slide project introduction in the talk "Full circularity in chemicals and plastics enabled"	
M12-M18						
Project CLIL & Science. Plastics a real challenge for today	22/10/19	Valencia, Spain	Laura Almar	CSIC	iCAREPLAST project Presentation as part of the Talk "Rethinking the future of plastics: moving towards a circular economy"	
23rd Ecomondo. The Green Technology Expo	5-8/11/19	Rimini, Italy	Laura Almar	CSIC	iCAREPLAST project Presentation.	
Biorizon Annual Event	28/11/19	Antwerp, Belgium	Pieter Imhof	ввтх	iCAREPLAST Project promotion as part of the Talk "Circularity with aromatics enabled".	
SusPlast Annual Conference	31/04/20	Online	Julio García Fayos	CSIC	iCAREPLAST project Presentation.	
Soluciones tecnológicas nuevas oportunidades de mercado	12/12/19	Valencia, Spain	Jose M. Serra	CSIC	iCAREPLAST project Stand.	
M18-M24						
Plastics Circularity Multiplier Online Conference	14/10/20	Online	Jose Manuel Serra	CSIC	Progress of iCAREPLAST project.	
CIRP LCE 2021 Conference CIRP LCE 2021 Conference	10/03/21 10/03/21	online online	Selin Erkisi Arici Johanna Hagen	TUBS	Oral Presentation "Comparative LCA of Municipal Solid Waste Collection and Sorting Schemes Considering Regional Variability". Oral Presentation "Framework for the Life Cycle Assessment of semi-permanent process units in volatile chemical recycling process chains".	
M30-M36						
EU Green Deal - Research and			Jose Manuel		iCAREDI AST project promotion at	
Innovation as a driver towards Climate Neutrality	3-4/10/21	Dubai, UAE	Serra and Laura Almar	CSIC	Networking Village.	





M36-M42					
BlackCycle 1st Workshop	22/11/21	Cebazat, France / Online	Laura Almar	CSIC	iCAREPLAST project Presentation.
I Ciclo de Seminarios del IUNAN de Energía y Medioambiente	16/3/22	Córdoba, Spain	J. M. Serra	CSIC	iCAREPLAST project promotion
11 Conferencia del Programa Marco de Investigación e Innovación de la Unión Europea en España - Horizonte Europa - El nuevo Horizonte para Europa	06/04/22	Valencia, Spain	María Siurana, Laura Almar	CSIC	iCAREPLAST poster presentation.
M42-M48					
Mix-up Friends	-	Online	J. M. Serra and L. Almar	CSIC	Scientific community network https://www.mix-up.eu/friends
Pint of Science	23/5/22	Valencia, Spain	M. Laqdiem	CSIC	iCAREPLAST project promotion – Oxycombustion activities
SECV 2022: "Materiales para la descarbonización del sector energético"	3-6/5/23	Madrid, Spain	J. M. Serra	CSIC	Keynote presentation – iCAREPLAST project promotion
"Cambio Climático en las empresas" EL ESPAÑOL- ACCIONA	15/5/22	Madrid, Spain	J. M. Serra	CSIC	iCAREPLAST project promotion
UT & ITQ internal workshop	01/9/22	Valencia, Spain	J. M. Serra	CSIC, UT	iCAREPLAST project promotion
Asamblea General SusChem- España	27/09/22	Madrid, Spain	Cristian A. Severi	URB	iCAREPLAST project promotion
INNOTRANFER - symposium with a talk on the "Electrification and decarbonization of the processes industry".	5/10/22	Valencia, Spain	J. M. Serra	CSIC	iCAREPLAST project promotion
Environmental Solutions and Ecologic Transition, ecoFIRA	4-6/10/22	Valencia, Spain	J. M. Serra	CSIC	iCAREPLAST project promotion (flyers and ITQ-CSIC stand)
M49-M54					
IndrommedaTec	15/12/22	Valencia, Spain	CSIC and KER members	CSIC, KER	iCAREPLAST project promotion

Dissemination of iCAREPLAST project results:

Dissemination of iCAREPLAST Project Results					
Name of the Event	Date	Place	Author(s)	Partner	Type of Presentation (Poster/Oral) and Title
M6-M12					
XXXVII Reunión Bienal de la Real Sociedad Española de Química	6-30/05/19	San Sebastián, Spain	Julio García Fayos	CSIC	Poster Presentation "Ceramic membranes for O2 production and their applications in catalytic industrial processes".
22nd International Conference on Solid State Ionics	16-21/06/19	PyeongChang, Korea	Jose M. Serra	CSIC	Poster Presentation "Catalytic Membrane Reactors for Power Generation via Energetic Valorisation of Waste Plastic Recycling By- products Streams".
3rd International Congress of Chemical Engineering	19-21/06/19	Santander, Spain	Laura Navarrete	CSIC	Poster Presentation "Energetic Valorisation of Pyrolysis Gases from Plastic Recycling with Catalytic Membrane Reactors".
M12-M18					
7th International Conference on Organic Solvent Nanofiltration	28-30/10/19	Enschede, The Netherlands	Patrick de Wit	UT	Oral Presentation "Organic solvent nanofiltration for catalytic recycling of plastic residues (iCAREPLAST)"
2nd ITQ Winter Meeting	19/12/19	Valencia, Spain	Sara Escorihuela	CSIC	Oral Presentation "In-situ water removal from CO ₂ methanation process with a polymeric thin film composite membrane".
2nd ITQ Winter Meeting	19/12/19	Valencia, Spain	Marwan Laqdiem Marin	CSIC	Poster Presentation " <i>Best Poster Award</i> ": "Energetic Valorisation of Pyrolysis Gases from Plastic Recycling with Catalytic Membrane Reactors".
Sustainable Energy Futures Annual Conference - ICL	13/09/19	London, UK	Nana Owusu Nyantekyi	IC	Poster Presentation "An Analysis of the Valorisation of Waste Plastics to Produce Fuels and Petrochemicals"
M18-M24					
M24-M30					
CIRP LCE 2021 Conference	10/03/21	online	Selin Erkisi Arici	TUBS	Oral Presentation "Comparative LCA of Municipal Solid Waste Collection and Sorting Schemes Considering Regional Variability".
CIRP LCE 2021 Conference	10/03/21	online	Johanna Hagen	TUBS	Oral Presentation "Framework for the Life Cycle Assessment of semi-permanent process units in volatile chemical recycling process chains".
M30-M36					
ICHEAP15, 15th International Conference on Chemical and Process Engineering	23-26/05/21	online	Paula Costa	LNEG	Oral Presentation
16th Conference on Sustainable Development of Energy, Water and Environment Systems (SDEWES)	10-15/10/21	online	Paula Costa	LNEG	Oral Presentation
M36-M42					





Euromembrane	28/11- 02/12/2022	Copenhagen, DE	P. de Wit , R. van Lin ,T. Visser,	UT	Invited Talk "Organic Solvent Nanofiltration for Catalytic Recycling of Plastic Residues (iCAREPLAST)"
Sino-UK Workshop on Biomass Conversion and Utilisation	10/12/21	Liverpool, UK	Xiangyi Long	IC	Oral Presentation and iCAREPLAST project promotion as part of the Talk "Thermal pyrolysis of plastic to added-value chemicals".
M42-M48					
20th International Zeolite Conference (IZC-2022)	3-8/7/22	Valencia, Spain	P. Frigols- Arroyo, M. Parreño- Romero, L. Almar, J. M. Serra, M. E. Domine	CSIC	"Desilicated Mordenite-type zeolites as efficient catalysts for the alkylation of aromatics with olefins present in plastic's pyrolysis liquids"
20th International Zeolite Conference (IZC-2022)	3-8/7/22	Valencia, Spain	P. Frigols- Arroyo, M. Parreño- Romero, L. Almar, J. M. Serra, M. E. Domine	CSIC	"Ga- and Zn-ZSM-5 zeolites as efficient catalysts for the aromatization of paraffins and olefins present in plastic's pyrolysis liquids"
25th Conference on Process Integration, Modelling, and Optimisation for Energy Saving and Pollution Reduction	5-8/9/22	Bol, Croatia	P. Costa	LNEG	Oral Presentation "Catalytic pyrolysis of municipal plastic wastes (MPW) for the production of valuable chemical products"
INNOVATION FORUM 4PLASTICS	13/10/22	Mallorca, Spain	Cristian A. Severi	URB	Oral Presentation
M49-M54					
Horizon 2020- Innovation Fund Synergies Workshop	08/02/2023	Online	Tijmen Vries	BioBTX	Presented the iCAREPLAST project as showcase and possible opportunity for future Innovation fund application during workshop organised by European Framework Programme for R&I



Annex III. Press Releases





Press releases of iCAREPLAST project

Press release	Links
	https://www.icareplast.eu/fileadmin/media_iCAREPLAST/Documents/13- 03-2019_NP_ITQ_iCAREPLAST_CSIC.pdf
	https://www.csic.es/es/casos-de-exito/integrated-catalytic-recycling-
	plastic-residues-added-value-chemicals
	https://itq.upv-csic.es/wp-content/uploads/2019/03/iCAREPLAST-
Press release 1. The first	Pressrelease_last.pdf
promotion of the project. It	https://www.urbaser.com/en/2019/03/icareplast-a-key-to-closing-the-
was published in different	plastics-recycling-cycle/
partners' websites	https://www.upv.es/noticias-upv/noticia-10928-icareplast-es.html
	https://www.ipoint-systems.com/newsroom/news-detail/icareplast-the-
	answer-to-cost-and-energy-efficient-plastic-recycling/
	https://www.ipoint-systems.com/de/newsroom/news-detail-de/icareplast-
	die-antwort-auf-kosten-und-energieeffizientes-kunststoff-recycling/
	https://www.lneg.pt/project/icareplast/
Press release 2. Video promotion. Magazine Article in UmweltDialog (German, 24th July 2020)	https://www.umweltdialog.de/de/wirtschaft/circular- economy/2020/Kunststoffabfaelle-kosteneffizient-und-umweltvertraeglich- recyceln.php
Press release 3. Magazine Article in RETEMA. Revista Técnica de Medio Ambiente nº 227 (Spanish, November- December 2020), pages 94-99:	https://issuu.com/r.retema/docs/retema227/94
	https://www.lavanguardia.com/local/valencia/20230324/8850847/spin-
	off-kerionics-alcanza-hito-generar-hidrogeno-verde-tecnologia-propia.html
KERIONICS Press releases	https://www.lamoncloa.gob.es/serviciosdeprensa/notasprensa/ciencia-e- innovacion/Paginas/2023/270323-morant-transicion-energetica.aspx
	https://elpais.com/economia/negocios/2023-04-05/kerionics-investiga-
2023	como-conseguir-hidrogeno-verde-barato.html
	http://www.gentedigital.es/valencia/noticia/3592362/la-spin-off-kerionics-
	alcanza-el-hito-de-generar-hidrogeno-verde-con-tecnologia-propia/
	https://www.tendencias21.es/Dos-spin-off-de-la-UPV-premiadas-por-su-
	caracter-innovador_a41084.html

The iCAREPLAST Press Room section has been created at the website to collect all project press releases:

- O <u>https://www.icareplast.eu/documents/</u>
- J Press release 1: Spanish version UPV, Spanish version CSIC
- ∪ Press release 1: <u>German version IPT</u>