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A comprehensive techno-economic and environmental comparison of plasma-treated adhesive-bonded thermoplastic composites

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A B S T R A C T

Thermoplastic composites are gaining attention for their ability to support a circular economy, shifting away from traditional cost-driven design. Unlike thermoset composites, commonly used in automotive and aerospace sectors, thermoplastic composites offer recyclability, promoting environmental and economic sustainability. Adhesive bonding in thermoplastic composites provides even stress distribution, maintaining structural integrity and reducing delamination risks, which is crucial for lightweight design in transport and renewable energy applications. Optimising adhesive bonded joints is essential to improve structural integrity, efficiency and sustainability. This involves balancing technical, economic and environmental factors, with surface preparation, plasma treatment in particular, playing a key role.

This paper provides a comprehensive techno-economic-environmental comparison of adhesive bonded joints. The study integrates cost-effectiveness, production efficiency and environmental impact assessments to guide informed decision-making. The methodology includes preparing substrates with low-pressure plasma surface treatments, forming adhesive-bonded joints with different adhesives, and optimising process parameters using Response Surface Methodology (RSM). A Life Cycle Assessment (LCA) on optimised scenarios enhances Tensile Shear Strength (TSS). The study concludes with a comprehensive comparison, offering insights for resilient, efficient and sustainable engineering solutions.

Introduction

In today's engineering landscape, the growing interest in thermoplastic composites lies in the need, now more than ever, to abandon the traditional design logic, aimed at ensuring the functionality of the product at the lowest cost, to acquire a circular perspective of the goods manufactured (Campbell, 2010).

For several decades, the most popular fibre-reinforced polymers, especially in the automotive and aerospace industries, are those with thermoset matrix, by virtue of their ability to combine high mechanical properties with remarkably light weight. However, they generate a significant amount of waste, due to the difficulty with which they are recycled. (Lunetto et al., 2023; Hao et al., 2021; Mativenga et al., 2017; Lefeuvre et al., 2017)

In contrast, thermoplastic composites, due to their ability to be remelted and remoulded, allow total component recovery at the end of life, even for structural applications, and with it a step forward in the environmental and economic sustainability challenge experienced by the industries operating in the field. In addition to shorter production cycles, they also provide a variety of options available for making joints, including adhesive bonding. This is a particularly suitable process for joining composite materials due to several inherent advantages it offers. Unlike traditional mechanical fastening methods such as riveting or welding, adhesive bonding distributes stress more evenly across the joint area, thereby minimising stress concentrations that can weaken composite structures. This characteristic is crucial for composites, which are prone to damage from localised stresses. Moreover, adhesive bonding does not require drilling or punching of holes in the composite, preserving its structural integrity and reducing the risk of delamination or fibre damage. This non-invasive nature also contributes to weight savings, as it eliminates the need for additional hardware components. Overall, adhesive bonding not only enhances the whole performance and durability of composite structures but also supports lightweight design principles critical for applications in transport and renewable energy sectors.

However, optimisation of adhesive bonded joints plays a pivotal role in enhancing structural integrity, efficiency and sustainability across various applications. Selection and optimisation of adhesive systems involve intricate trade-offs among technical, economic and

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Nomenclature

ANOVA	ANalysis Of VAriance
CJ	Control joints
DoE	Design of Experiments
EP	Epoxy
GFRTPU	Glass Fibre Reinforced Thermoplastic Polyurethane
HY	Hybrid
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LPP	Low-Pressure Plasma
PU	Polyurethane
RSM	Response Surface Methodology
SLJ	Single Lap Joint
TSS	Tensile Shear Strength

environmental considerations. The adhesive bonding process is complex and requires significant interaction between substrate characteristics, interface properties and chemical-physical nature of the adhesive. A preliminary and fundamental step in achieving good quality bonded joints is the surface preparation of the substrates, the purpose of which is to clean them of any contaminants, which are the result of previous processing, increase their wettability and, in some cases, create an appropriate roughness, which favours adhesion forces.

Numerous studies highlight plasma treatment, whether conducted under vacuum or atmospheric conditions, as an effective method to prepare both reinforced and non-reinforced polymer substrates. This process can achieve the desired level of interaction with the material and ensure proper surface activation once the process parameters are appropriately regulated (Mandolfino et al., 2021; Palasakar et al., 2020; Gao et al., 2009; Hegemann et al., 2003). Additionally, plasma treatment is viewed not only as an environmentally friendly option but also as one that can easily be integrated into various manufacturing settings.

Vacuum or atmospheric-pressure plasma are also applied to enhance the adhesion properties of reinforcements to the matrix in the production of natural fibre composite materials (Mohsen et al., 2024).

Most studies on bonding reinforced polymers focus on thermosetting matrix composites, given their more frequent use in structural applications (Akman et al., 2020; Schweizer et al., 2017; Dighton et al., 2019; Ruzafa-Silvestre et al., 2024). On the contrary, studies on the adhesive bonding of thermoplastic composites are still quite limited. For example, two recent studies addressed various surface treatment strategies to enhance the properties of bonded joints of different types of thermoplastic composites. Avci et al. investigated the effects of two different surface treatments (wet blasting and atmospheric plasma treatment) on CF/LM-PAEK thermoplastic composites. They examined changes in surface characteristics and adhesion strengths. The surfaces treated were bonded using an epoxy-based film adhesive and subjected to single lap shear testing to assess adhesion strength under tensile loading. The results indicated that both surface treatments enhanced the adhesion strength of the bonded pre-consolidated CF/LM-PAEK thermoplastic composites compared to untreated specimens (Avcı et al., 2024).

Similarly, S.J. Lim et al. applied laser surface treatment to CFRP and CFRTP surfaces to improve adhesive bonding strength with a thermoset adhesive. They investigated the effect and mechanism of laser surface treatment. Single lap shear adhesive bonding tests and tensile adhesive tests were conducted to determine the adhesive bonding strength (Lim et al., 2020).

Both studies focused on the use of epoxy adhesives, highlighting a specific interest in evaluating the performance of these adhesives with different surface treatments.

Conversely, this paper addresses the critical need for a comprehensive techno-economic-environmental comparison of adhesive bonded joints of composite materials. Such an analysis goes beyond mere mechanical performance assessment to integrate cost-effectiveness, production efficiency and environmental impact throughout the lifecycle of structural joints. By evaluating these interconnected aspects, engineers and decision-makers can make informed choices that balance performance requirements with economic feasibility and environmental sustainability.

Through this integrated approach, this paper aims to provide insights into optimising bonded joint technologies that not only meet rigorous performance standards but also align with sustainable development goals. By advancing our understanding of the techno-economic-environmental aspects of bonded joints, we can pave the way for more resilient, efficient, and environmentally friendly engineering solutions in the future.

Experimental activity

The general approach involved five phases: (i) preparing thermoplastic composite substrates various plasma surface treatment setups; (ii) adhesive-bonded-joint preparation using three different adhesives; (iii) statistical analysis using the Response Surface Methodology (RSM) technique in order to optimise process parameters for each joint configuration; (iv) life cycle assessment of the different optimised scenarios, using the plasma parameters that maximise Tensile Shear Strength (TSS); (v) a comprehensive techno-economic-environmental comparison of the bonded joints. The goal of this method is to explore and compare different adhesive systems through a multidimensional lens, contributing valuable insights to adhesive bonding methodologies, guiding material selection and joint design strategies for enhanced structural integrity and reliability.

Materials, planning of experimental campaign using DoE and joint manufacturing

A glass-fibre reinforced polyurethane-based thermoplastic (GFRTPU) composite, Tepex® Dynalite 108-FG290(4)/45% (Lanxess, Germany), was used as the substrate to produce adhesive bonded joints (the main characteristics are shown in Table 1).

Three different adhesives were used to thoroughly assess adhesive bonding performance across various scenarios. This systematic exploration considers the interaction between substrate, adhesive and surface treatment, crucial to achieve optimal mechanical response. The adhesives selected from three different producers were: (i) a two-component polyurethane adhesive (PU); (ii) a two-component structural epoxy adhesive (EP); (iii) a two-component hybrid cyanoacrylate and acrylic

Table 1
Material properties of the GFRTPU composite substrate.

Matrix	Polymer	Thermoplastic polyurethane (TPU)
Reinforcement	Fibre	E-Glass filament
	Fabric	Twill 2/2
	Fabric thickness (mm)	0.25
	Area weight (g/m^2)	290
	Yarn (tex)	204
Laminate	Density (kg/m^3)	1820
	Fibre content (vol%)	45
	Thickness (mm)	1
	Tensile modulus (parallel and normal) (MPa)	23,000
	Stress at break (parallel and normal) (MPa)	440
	Strain at break (parallel and normal) (%)	2.3
	Glass transition temperature 10 °C/min (°C)	94

adhesive (HY).

Traditional and LPP surface treatments were performed and compared, as follows: (i) acetone cleaning followed by mechanical abrasion using 3M™ Hookit 255P P400-grit abrasive paper, with second acetone cleaning to eliminate all traces of dust (MA), (ii) LPP treatment was carried out with various setup configurations. Case (i) was designated as control joint (CJ), against which the differences observed with treatments (ii) are referenced, since it is the most commonly used surface treatment and is recommended by standards (ASTM International, 2003).

Specifically, experimental campaign on LPP treatment was designed according to a Central Composite Design (CCD) with two factors. This design was chosen because it enables efficient modelling of the experimental response, allowing accurate estimation of first- and second-order regression terms.

The control factors chosen for LPP treatment were power (P) and exposure time (t), while adhesive was set as a scenario variable, thus resulting in three adhesive-bonded-joint scenarios, namely respectively, with reference to the type of adhesive used, PU-scenario, EP-scenario and HY-scenario. Table 2 displays the factors investigated along with their three levels (low, medium and high), represented in both actual (natural) and coded forms, normalised within a range from -1 to 1. The levels were selected to operate under the stability conditions of the plasma reactor, allowing for a broad range that is centered on the machine's full scale.

Immediately after treatment, single lap joints (SLJ) were fabricated following the geometries specified in the ASTM D1002 standard (Fig. 1) (ASTM International, 2009), ensuring an adhesive thickness of 0.05 mm, using an appropriate jig.

Three replicates were realised for each treatment configuration. Upon the complete curing of the various adhesives following the manufacturer's instructions, the SLJ underwent tensile shear testing using an Instron 8802 servo-hydraulic testing machine (Instron, MA, USA) at a test speed of 1.3 mm/min.

Analysis of Variance (ANOVA) was conducted to assess the impact of process factors and their interactions on the TSS response variable, utilising Design-Expert Software Version 23.1.1 (Stat-Ease, Inc. MN, USA). After the screening phase of factors, in order to identify only those that have a statistically significant effect on response the TSS was modelled using RSM, allowing the development of first- or second-order regression models that accurately describe the response behaviour within the investigated domains. The resulting response surfaces facilitated the identification of setup parameters that optimise TSS performance, with the aim of determining statistically equivalent conditions in terms of mechanical performance but more cost-effective or easier to implement in the manufacturing process.

Life cycle assessment

A key part of the experimental campaign was the comparison of the three joint configurations in terms of environmental impact, using the Life Cycle Assessment (LCA) methodology. The analysis was conducted in accordance with the international standards ISO 14040 (ISO, 2020) and ISO 14044 (ISO, 2020), using SimaPro 9.4.0 software and its database, Ecoinvent3.

The aim of the environmental impact assessment was to highlight the differences between the three joint configurations, resulting as the optimal solution found from the response surface analysis conducted. For this reason, the LCA study considered only the differential aspects between the three joints, namely the type of adhesive (polyurethane-PU, epoxy-EP, hybrid-HY) and the respective optimal plasma-surface-treatment parameters (power and exposure time). The impact of the GFRTPU composite substrate was neglected, as it is the same for all three joints. Moreover, including the material in the LCA analysis would certainly have masked the effect of the other elements, since the impact of the substrate is much more significant compared to the other components of the joint, as demonstrated in a previous study conducted by the Authors (Mandolfino et al., 2024).

As already mentioned, the main goal of the LCA is not to calculate the absolute impact of each joint, but to provide a comparison between them. Therefore, the analysis focused exclusively on the bonding phase, neglecting the fibre and matrix production as well as the joint's service phase and end-of-life stages (recycling or reuse operations). Table 3 shows the inventory for the three scenarios analysed related to the adhesives' contribution.

It should be mentioned that some of the data implemented into the Life Cycle Inventory (LCI) were obtained through direct measurements, while others were collected from the literature, the products' technical and safety data sheets, or retrieved from the software database. The "*quantity*" of adhesive used in the specific configuration refers to the average mass of adhesive required to produce one joint. This value was experimentally determined by weighing five identical joints made with the same adhesive using a precision balance and calculating the average. In turn, the primary components of the three adhesives (i.e., their "*main constituents*" listed in Table 3) correspond to the specific ingredients reported in the relevant safety data sheets; their values are not provided here to prevent the identification of the corresponding commercial products.

In addition to the contribution of the adhesive(s), the environmental impact of surface treatment was also assessed. During execution of each plasma treatment for every LCA scenario, energy consumption was measured using a power meter (KTEM02, Ketotec, PRC). The energy values considered in the impact assessment account for the entire plasma process, including both the preliminary vacuum generation phase in the treatment chamber and the actual treatment phase (exposure time). For the analysis, it was assumed that the energy required for joint production is supplied by the European electricity grid mix, reflecting the location where the tests were conducted. The energy consumption values corresponding to the optimal treatment conditions for each scenario will be reported and discussed in section "Optimisation of TSS response in the different scenarios".

To determine the impact of each type of joint, the LCA analysis was set up considering a production batch of 5 bonded joints in order to maximise the utilisation of the load capacity of the LPP equipment.

The functional unit identified to compare the three different scenarios (PU, EP and HY) was therefore formulated as "the set of 5 adhesive bonded joints with optimised surface treatment, i.e., determining the highest average TSS increase compared to a mechanical abrasion".

There are two primary methods for evaluating environmental impact: midpoint and endpoint. Midpoint indicators address specific environmental impacts, such as climate change or acidification, across a wide range of ecological outcomes. The endpoint method aggregates environmental impacts into three broader categories, Human health, Ecosystem quality and Resource scarcity, simplifying the interpretation of LCA results.

In this study, the Life Cycle Impact Assessment (LCIA) was conducted using the ReCiPe2016 EndPoint method (Huijbregts et al., 2017). It offers three perspectives: (i) individualist for short-term effects, (ii) hierarchist, aligning with common policy principles, (iii) egalitarian for precautionary perspectives over longer timeframes. The hierarchist version with average weighting was chosen as the default, reflecting scientifically and politically accepted value choices (Huijbregts et al., 2017; Goedkoop et al., 2016).

Table 2
Factors and relative levels for all the three investigated scenarios.

Factors	Levels: Actual (Coded)		
	Low (-1)	Medium (0)	High (+1)
Power, P (W)	50	125	200
Exposure time, t (s)	10	120	230

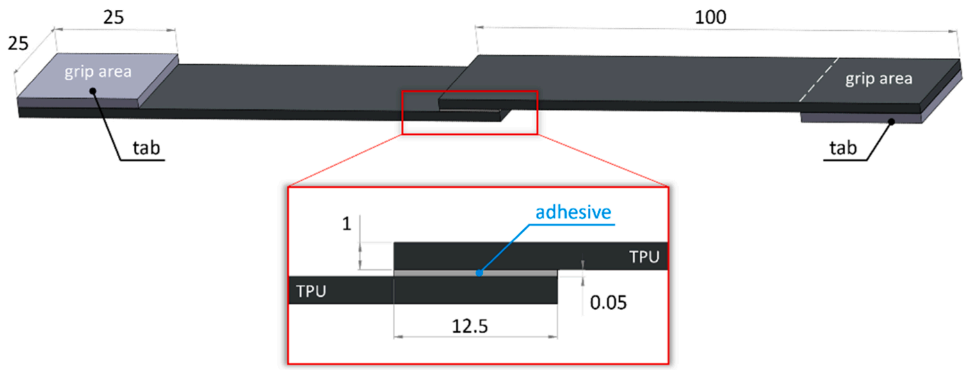

Fig. 1. Single lap joint sample geometry (dimensions in mm).

Table 3
Data used in the LCI for PU, EP and HY scenarios.

Scenario	Adhesive type	Quantity (g)	Main constituents
PU	Polyurethane	0.30	Methylene diphenyl diisocyanate Methylamine
EP	Epoxy	0.12	Epichlorohydrin Epoxy resin Acrylonitrile Butadiene Styrene
HY	Hybrid	0.15	2-Ethylhexyl Acrylate Methyl methacrylate

Techno-economic comparison

A product or process may possess desirable mechanical characteristics and exhibit a low environmental impact according to LCA. However, if it is economically or technically impractical to implement at scale, its adoption may be limited. Technical-economic considerations can aid in assessing the feasibility of scaling up production and commercialisation by providing critical information about time and costs.

On these premises, in evaluating various adhesive scenarios, to the analysis of the mechanical behaviour of bonded joints and the evaluation of environmental impact, a comparison was added to analyse performance based on three primary criteria: cost, working time and handling time. This comprehensive assessment aims to identify the most efficient and cost-effective adhesive solution for different applications.

Indeed, the cost of adhesives is a crucial factor influencing the overall feasibility of the bonding process. The analysis in this work includes the retail cost of 50 ml of adhesive (a common volume for a single cartridge), averaged across three online suppliers.

Regarding the comparison based on process timing, two time periods are fundamental in evaluating the feasibility of the process: (i) working time in adhesive bonding, also known as open time or pot life, which refers to the period during which the adhesive remains workable and can form a bond after being applied to the surfaces being joined. This is a crucial parameter for production scheduling and can become a significant cost and environmental factor if large quantities of adhesive need to be discarded due to expired working time (Petrie, 2000); (ii) handling time, which is the time required for the adhesive to develop enough strength so that the bonded materials can be handled or moved without damaging the bond. This is typically shorter than the full cure time.

The techno-economic comparison highlights the trade-offs between cost, working time and handling time for different adhesive scenarios. Selecting the appropriate adhesive depends on specific project requirements, including budget constraints, production speed and the need for precision in component alignment.

To facilitate comparison of adhesives in terms of these different technical, economic and environmental aspects, the results obtained from TSS optimisation, LCA, cost analysis and timing evaluations have been normalised on a scale from 1 to 5, assigning to each dimensions analysed a score proportional to the algebraic value assumed by the corresponding indicator if the dimension leads to a positive impact on the process (i.e. Tensile Shear Strength) otherwise if the dimension has a negative impact on the process an inversely proportional score (i.e. Purchasing Cost).

Results and discussion

Optimisation of TSS response in the different scenarios

First, for each adhesive, the reference values obtained with traditional surface treatments were established. The purpose was not to evaluate the absolute effectiveness of the plasma treatment, as this is an expected result (Pizzorni et al., 2019; Lin et al., 2020; Dighton et al., 2019), but to find the process parameters that provide the best results in terms of improving the mechanical performance of the adhesive bonded joints.

ANOVA project for Polyurethane Adhesive resulted in a significant second-order regression model. Table 4, which refers to PU-scenario results, highlights a great significance of power, both at first and quadratic order but also the significance of the interaction term.

The Lack of Fit F-test is not significant, so the second-order linear regression model, described by the related Coded Equation (Eq. (1)), fits the experimental responses:

$$TSS = 3.12 - 1.19\,P + 0.14\,t - 0.40\,Pt + 0.83\,P^2 + 0.40\,t^2 \quad (1)$$

where P is power (W) and t is exposure time (s).

This equation is representative of the expected mean value of TSS (MPa) in each point of the investigated domain and, consequently, can be exploited to make predictions of the bonded-joint behaviour at

Table 4
ANOVA results of PU-scenario.

Source	Sum of squares	df	Mean Square	F-value	*p*-value	
Model	32.74	5	6.55	38.28	<0.0001	significant
A-Power	25.37	1	25.37	148.33	<0.0001	
B-Time	0.36	1	0.36	2.13	0.1594	
AB	1.94	1	1.94	11.37	0.0029	
A^2	4.09	1	4.09	23.94	<0.0001	
B^2	0.97	1	0.97	5.64	0.0271	
Residual	3.59	21	0.17			
Lack of Fit	0.63	3	0.21	1.27	0.3130	not significant
Pure Error	2.96	18	0.16			
Cor Total	36.33	26				

different combinations of the process factors. The process variables are all coded between −1 (low factor level) and +1 (high factor level).

Analysing the signs of these coefficients, TSS increases as power decreases and time increases; the interaction plot confirms the significance of the interaction effect between the operating parameters. For the low power level, a longer exposure time is preferable, whereas for maximum power, a minimal exposure time is preferable. (Fig. 2). Then, RSM was used to determine the combination of factors that provides the optimum TSS achievable and to predict the behaviour of joints under experimentally untested process conditions.

ANOVA of EP-scenario (Table 5) points out a similar behaviour for this type of bonded joint, whereby there is again a clear predominant effect of power as a parameter affecting TSS, also in the squared term accompanied by a significant interaction effect. However, in this second scenario, time is also significant.

This equation is representative of the expected value of TSS (MPa) at different levels of each factor:

$$TSS = 2.34 - 1.02\,P - 0.13\,t + 0.22\,Pt + 0.42\,P^2 \tag{2}$$

In this second model, the signs of the interaction effect change compared to the PU-scenario because the TSS is maximised at a low power level with a short exposure time, while for maximum power values, a higher exposure time is preferable. This is also visible from response surface and interaction plot in Fig. 3.

ANOVA of HY-scenario resulted in a not significant regression model (p-value of 0.1565, Table 6). Based on the assumptions made, the model terms are statistically significant if the p-value is lower than the significance level α=0.05. Hence, the differences in TSS values recorded are attributable to experimental error rather than the effect of the factors in the range investigated.

In this case the insignificance of the model means that each point of the investigated domain could be considered statistically equivalent in terms of response (TSS). In particular, the predicted value in this situation corresponds to the mean value of the experimental responses and the regression results in a mean-only model:

$$TSS = 2.11 \tag{3}$$

Looking at the interaction plot in Fig. 4, where the control joint reference value is reported (blue line), it can be concluded that plasma treatment can provide slightly higher TSS values (green line) compared to traditional treatment, regardless of the parameters used. Subsequently, for this type of joint, the most favourable process conditions (minimum power and exposure time) were selected.

The ANOVA analysis confirms the experimental results of the mechanical characterization campaign conducted on bonded joints. More details on the load-displacement curves obtained by testing the various configurations, as well as the typical fracture surfaces of significant cases, can be found in Appendix A.

Table 5
ANOVA results of EP-scenario.

Source	Sum of squares	df	Mean Square	F-value	*p*-value	
Model	20.86	4	5.22	106.38	<0.0001	significant
A-Power	18.91	1	18.91	385.71	<0.0001	
B-Time	0.31	1	0.31	6.26	0.0203	
AB	0.59	1	0.59	12.12	0.0021	
A^2	1.05	1	1.05	21.42	0.0001	
Residual	1.08	22	0.05			
Lack of fit	0.21	4	0.05	1.07	0.3992	not significant
Pure error	0.87	18	0.05			
Cor Total	21.94	26				

On the basis of the results obtained, Life Cycle Inventory could be finalised. Table 7 displays the percentage increase in TSS of LPP-treated joints compared to CJ across the different adhesives. The percentage increment values of TSS – which were obtained by comparing the optimum predicted value of the domain, determined from the fitted second-order regression model, and the mean value of control joints – are strongly dependent on the adhesive used. Significant improvement could be reached for the Polyurethane adhesive, where the TSS of plasma-treated joints was 129% higher than joints pre-treated with mechanical abrasion.

Environmental impact assessment through LCA methodology

This phase of the experimental campaign aimed to utilize the LCA methodology to compare the environmental impact of three optimized scenarios for GFRTPU LPP-treated bonded joints, as determined through RSM.

As previously mentioned in section "Life Cycle Assessment", to assess the environmental sustainability of the three different joint configurations, optimal TSS values achievable for the three types of bonded joints was established as the functional unit.

Subsequently, for each type of joint, the most favourable process conditions (minimising power and exposure time within the range investigated) were selected to ensure that each LPP-treated joint would achieve the maximum increment in term of TSS compared to the results

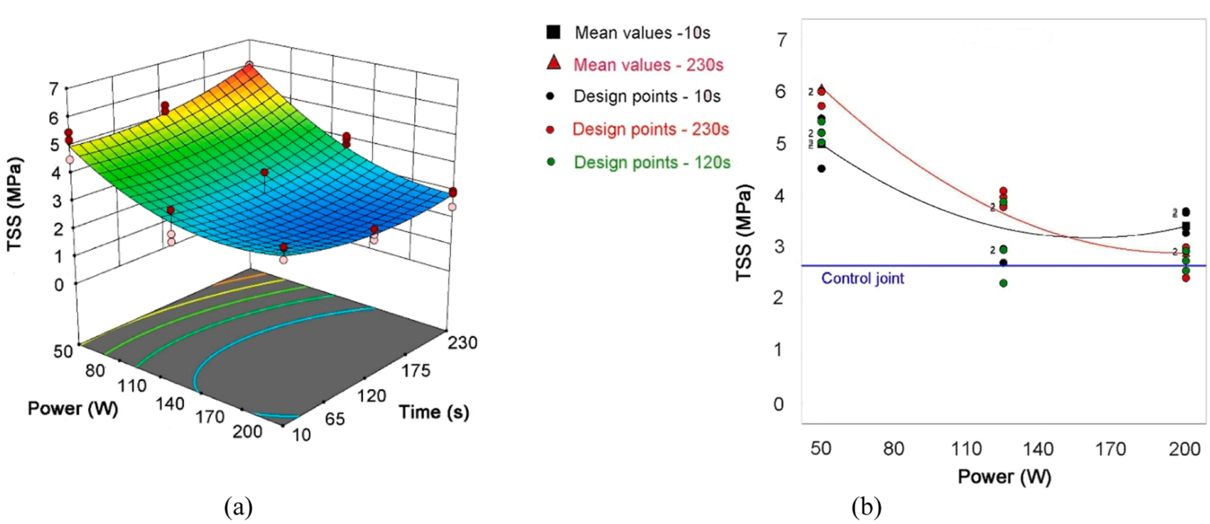

Fig. 2. (a) Response surface of PU-scenario and (b) interaction graph obtained through RSM.

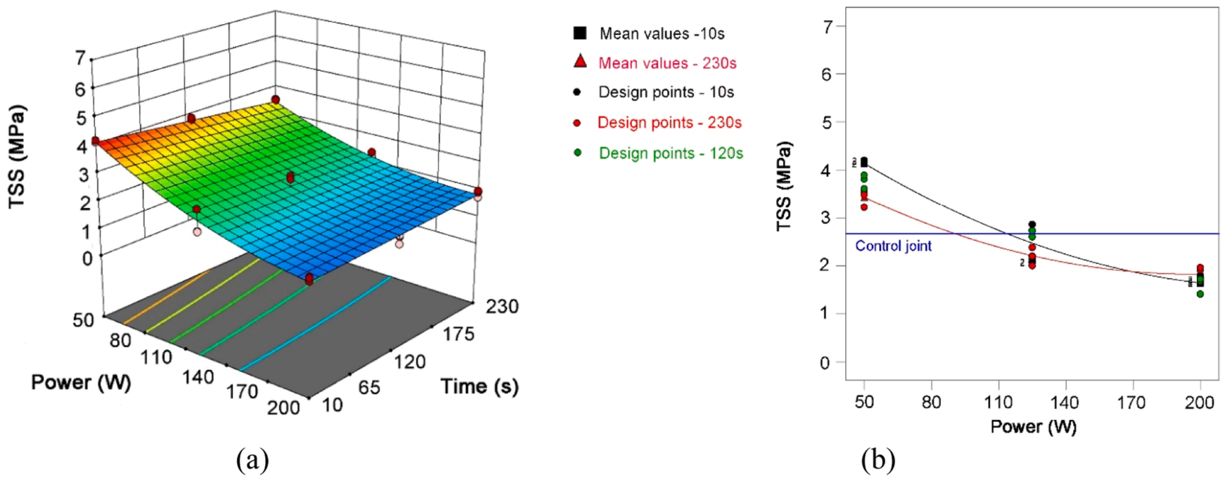

Fig. 3. (a) Response surface of EP-scenario and (b) interaction graph obtained through RSM.

Table 6
ANOVA results of HY-scenario.

Source	Sum of squares	df	Mean square	F-value	*p*-value	
Model	2.01	3	0.67	2.11	0.1565	not significant
A-Power	1.33	1	1.33	4.19	0.0653	
B-Time	0.55	1	0.55	1.74	0.2141	
AB	0.13	1	0.13	0.41	0.5344	
Residual	3.48	11	0.32			
Lack of Fit	0.57	1	0.57	1.96	0.1919	not significant
Pure Error	291	10	0.29			
Cor Total	5.49	14				

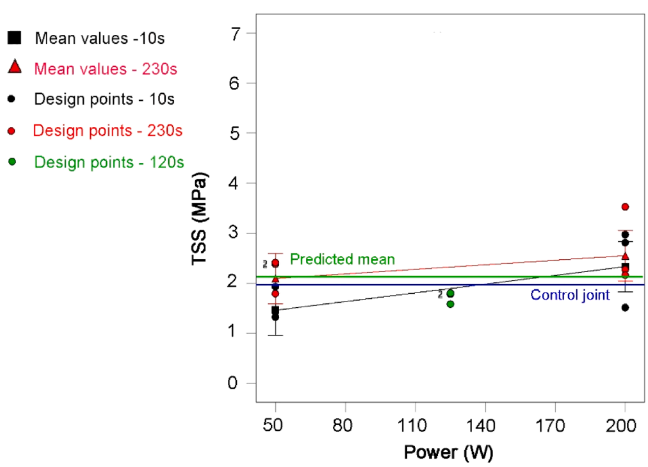

Fig. 4. Interaction graph of HY-scenario obtained through RSM.

obtained with a mechanical abrasion treatment, which was considered a threshold value.

Table 7, in addition to the predicted values for the three joints and the percentage increases compared to the CJ, provides the data used in the LCI phase regarding the energy consumption related to the optimised plasma treatment. As can be observed, to achieve the maximum increase, very long times are required for the joint made using PU adhesive, which leads to energy consumption tripling compared to the other joints.

The analysis carried out using SimaPro software allowed the definition of a comparison between the endpoint-impact categories for the different joints considered (Fig. 5). In particular, the endpoint method provides a single environmental score (in millipoints – mPt) for each joint, considering normalisation and weighting factors defined by the LCIA method selected, in this case Recipe2016 Endpoint (H).

From this, we can make a few observations: (i) the bonding process primarily affects the Human Health impact category, while the impact

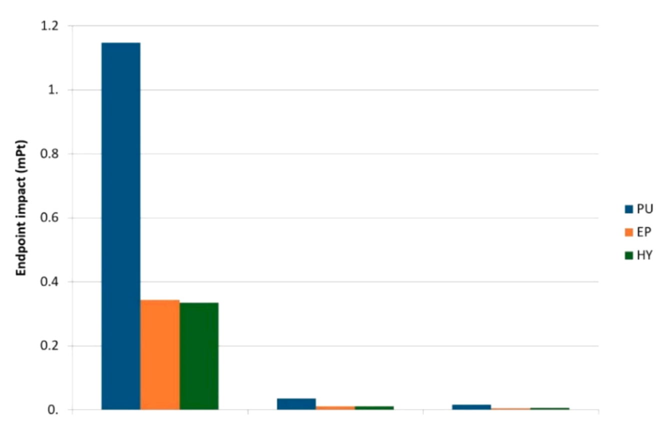

Fig. 5. Impact assessment for each endpoint category of the three scenarios.

Table 7
Plasma process parameters used for LCA with related mean predicted values.

Project	TSS of CJ (MPa)	Optimum TSS value LPP treatment (MPa)	TSS increase of LPP joints vs CJ (%)	Set-up parameters @ optimum value	Energy Consumption (kWh)
PU-Project	2.65 ± 0.64	6.07 ± 0.41	129	50 W – 230 s	0.055
EP-Project	2.67 ± 0.22	4.14 ± 0.22	55	50 W – 10 s	0.017
HY-Project	1.97 ± 0.02	2.11 ± 0.56	7	50 W – 10 s	0.017

on Ecosystem and Resource exploitation is nearly negligible. (ii) PU-scenario has the highest impact, while EP and HY have almost the same impact, approximately one-third compared to PU. This could be explained by adhesive main constituents and most of all the higher energy consumption.

Evaluating the elements that contribute most significantly to this impact category (Fig. 6), it is noticeable that the electricity used to optimise surface treatment is the primary factor in terms of impact for all three types of joints. The PU joint also has a substantial impact in terms of the main adhesive constituent.

This is due to the presence of isocyanates, which are chemicals commonly used in producing polyurethane adhesives. Despite their industrial utility, they pose significant health risks primarily through inhalation or skin contact. Indeed, isocyanates are potent respiratory irritants and can trigger allergic reactions, leading to occupational asthma and skin rashes upon exposure. Also, in terms of environmental impact, they can contribute to air pollution. Proper disposal and handling are necessary to minimise environmental contamination (Delavarde et al., 2024; Kemona and Piotrowska, 2020).

As further confirmation, an evaluation of the contribution of surface treatment and adhesive to the overall impact was carried out, always within the three scenarios. As can be seen from the bar chart in Fig. 7, the impact of surface treatment is always predominant, regardless of the adhesive considered. However, the impact of the polyurethane adhesive is still greater compared to the energy consumption envisaged in the scenarios using epoxy and hybrid adhesives.

To understand why electricity consumption has such a significant impact on Human Health, it was useful to add a drill down analysis of this impact category for the PU-scenario in terms of midpoint indicators, in particular distinguishing the contribution of electricity, isocyanates and other processes (Fig. 8). As can be seen, the electricity used to optimise the surface treatment significantly impacts several midpoint indicators within the Human Health category.

As previously mentioned, isocyanates, although less impactful than electricity, still affect many midpoint indicators, including the fine-particulate-matter-formation indicator, which is related to air pollution and as expected, human health toxicity. This substance is indeed highly carcinogenic (Qin et al., 2024).

Regarding the most impacted categories, the prominence of the fine particulate matter formation and global warming indicators is largely due to the substances identified as outputs by SimaPro associated with electricity production in the EU area. This is tied to the specific energy production mix, which in some EU countries is still heavily reliant on coal and fossil fuels, as illustrated in the pie charts of Fig. 9.

Comprehensive techno-economic-environmental comparison

To gain a thorough understanding that effectively guides the

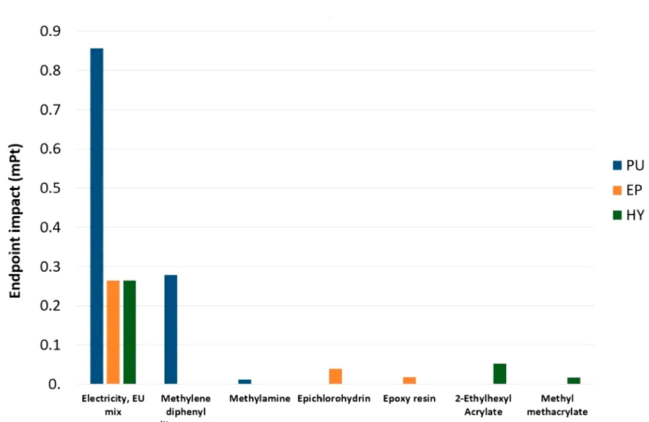

Fig. 6. Processes contribution on the Human Health endpoint category.

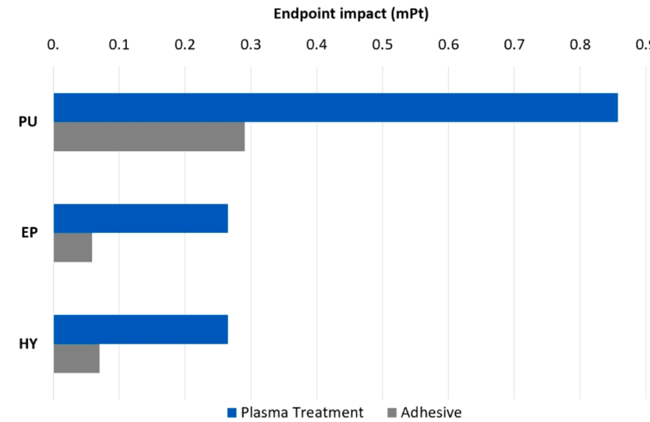

Fig. 7. Process impact assessment.

applicability of one adhesive system compared to another, it is important to recognise that mechanical and environmental impact considerations offer only a limited perspective. This prompted us to proceed towards a more comprehensive view and motivate analysis, in which we delve into a technical, economic and environmental comparison of using three different adhesives for plasma-treated thermoplastic composites.

A radar graph (Fig. 10) is used as an effective and fast way to compare the different scenarios, merging multiple variables to assess different aspects, and making a multidimensional analysis possible. In this case, the comparison is set between the use of PU, EP and HY adhesives. Metrics include purchasing cost, working time, handling time, TSS and environmental impact, considering data reported in Table 8 as reference values. For the sake of comparison between cases, the latter have been normalised, assigning a value on an upward scale from 1 to 5 to the five dimensions analysed in relation to both the numerical value assumed by the corresponding indicator and its inherent meaning and influence in the process: it is indeed worth noting that benefits in technical, economic and environmental aspects can be associated to greater values of working time and TSS, as well as lower values of handing time, environmental impact and adhesive cost. For example, a normalized value equal to 1 was assigned to the lowest value of working-time parameter (in this case, 1 min for HY adhesive) since this situation generally involve greater difficulty in bonding (e.g., related to positioning or waste) due to rapid reticulation of the adhesive (even inside the nozzle); consequently, a normalized value equal to 5 was assigned to the value corresponding to EP adhesive (90 min). In contrast, shorter handling time is generally beneficial for production time: therefore, the value 1 corresponded to 36 h of the PU adhesive, while the value 5 corresponded to 0.08 h of the HY one.

PU-scenario is characterised by low cost, making it an attractive option for projects with tight budgets. Its working time is relatively short (15 min), but it has a significantly longer handling time (36 h), which might delay the overall project timeline. PU adhesive exhibits the highest TSS of 6.07 MPa with cohesive failure, indicating it forms the strongest bond among the three adhesives, making it suitable for applications requiring high strength. However, the main drawback is its environmental impact, which is the highest among the three adhesives compared (1.2 mPt), due to the energy consumption and chemical constituents used in its formulation.

EP-scenario shows that the adhesive is moderately expensive, with the longest working time (90 min), which might slow down the application process. Its handling time is considerably shorter than PU adhesive (5 h), offering a balance between preparation and curing time. EP adhesive provides a TSS of 4.14 MPa, which is lower than PU but still sufficient for many applications requiring good bond strength (failure mode was predominantly cohesive). The environmental impact of EP adhesive is moderate (0.36 mPt), striking a balance between

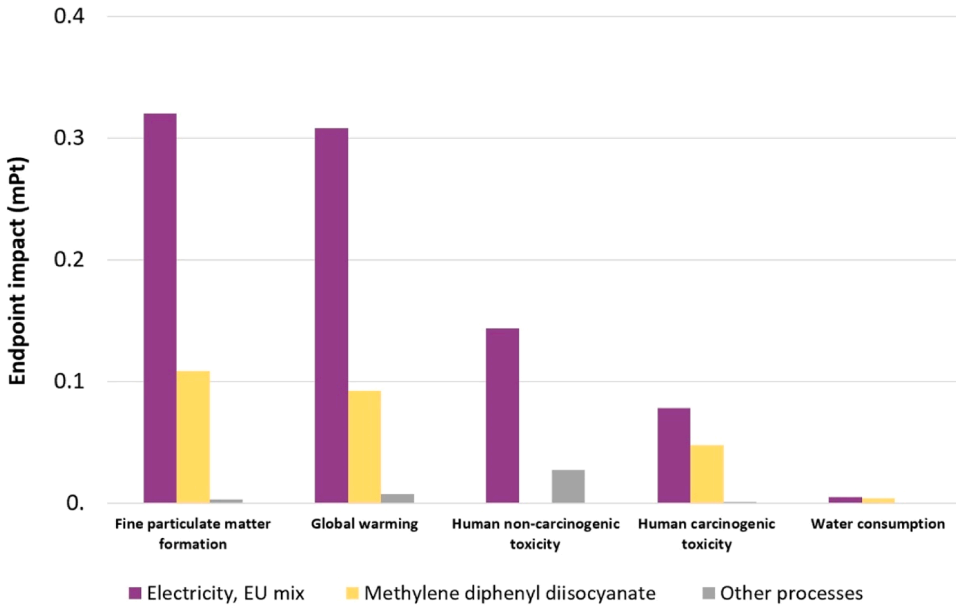

Fig. 8. Process contribution of PU on midpoint impact indicators of the Human Health category.

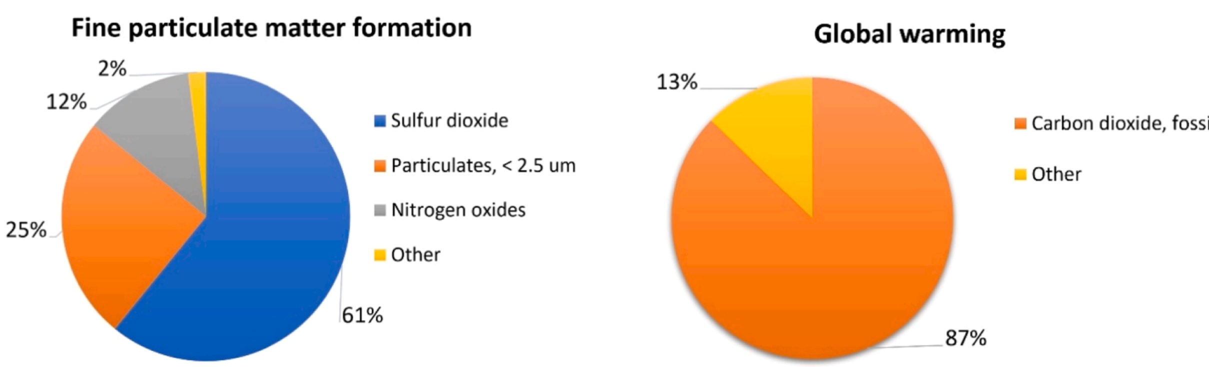

Fig. 9. Substances impacting midpoint indicators for PU-scenario.

performance and sustainability, making it a viable option for projects needing to consider environmental factors without compromising joint strength.

HY-scenario indicates that this adhesive comes with the highest cost, which could be a significant limit for budget-conscious projects. It compensates for this with the shortest working time (1 min) and the shortest handling time (5 min), features that are advantageous in operations that need to be completed rapidly (like positioning or repairs). However, it should be noted that although plasma treatment resulted in improved TSS of HY joints with different parameter combinations, the optimal TSS value is the lowest (2.11 MPa) among the three scenarios. As shown in Appendix A, unlike the PU and EP scenarios, plasma treatment does not change the failure mode of HY joints, which always remains adhesive as the process parameters change. In this regard, it should be considered that the criteria defining joint quality depend on the criticality class assigned to the specific joint based on its function and field of application. Consequently, the applicability of such a bonding solution must be evaluated in relation to the defined performance requirements and the benefits brought in terms of process. Proving to be the most environmentally friendly choice by virtue of its low environmental impact (0.353 mPt), the HY adhesive system analyzed here can be considered appropriate to address those projects that aim to minimize their ecological footprint. Considering its observed mechanical behavior, this is valid for applications characterized by less stringent performance requirements. Conversely, it may not be the preferred solution for many more critical structural bonding applications, since plasma treatment has proven to be – in this specific case – only a compromise way to improve the strength of the joint, but not its reliability. In such contexts, alternative pre-bonding treatments should therefore be considered and their effectiveness in shifting the failure mode from adhesion to cohesion should be evaluated, while considering that each alternative surface treatment will presumably have different economic and environmental implications than those highlighted here in relation to plasma treatment.

Conclusions

This study addresses the critical need for a comprehensive techno-economic-environmental comparison of adhesive bonded joints in thermoplastic composites. The methodology encompasses five key phases: preparing thermoplastic composite substrates with various plasma surface treatment setups; preparing adhesive-bonded joints using three distinct adhesives; conducting statistical analysis using the RSM to optimise process parameters for each joint configuration; performing an LCA of the different optimised scenarios, using the plasma parameters that maximise TSS; and conducting a comprehensive techno-economic-environmental comparison of the bonded joints.

Summing up, integrating techno-economic evaluation with

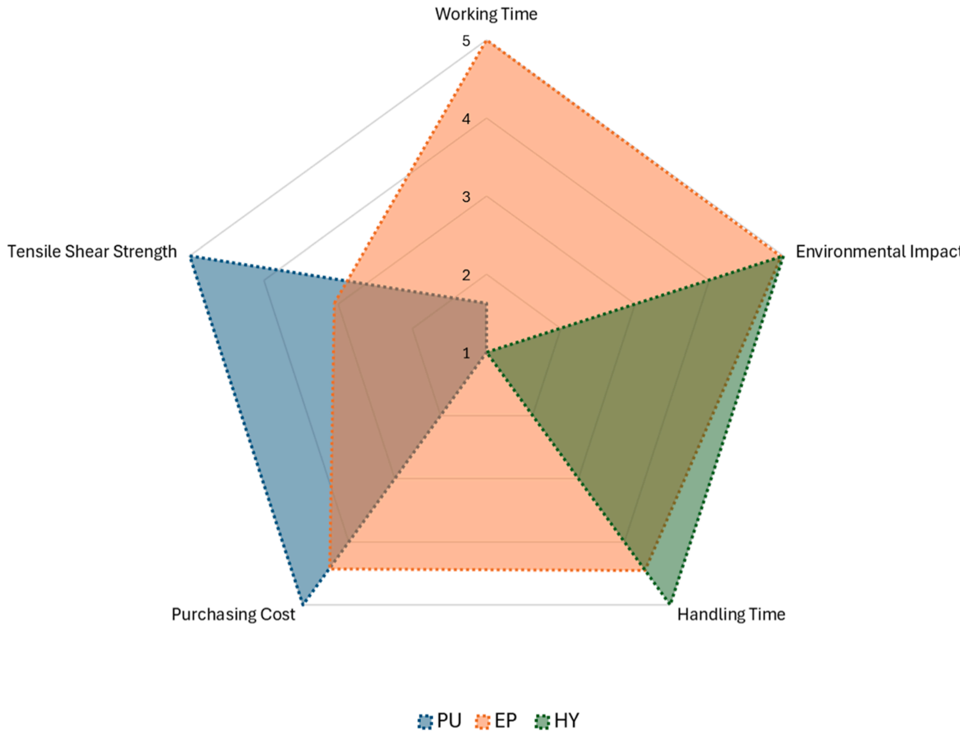

Fig. 10. Radar graph comparing techno-economic-environmental aspects of three adhesives.

Table 8
Data used to conduct the techno-economic-environmental comparison between the various adhesives.

	PU		EP		HY	
	Actual value	Normalized value	Actual value	Normalized value	Actual value	Normalized value
Working time (min)	15.00	1.63	90.00	5.00	1.00	1.00
Environmental impact (mPt)	1.20	1.00	0.36	4.97	0.35	5.00
Handling time (hours)	36.00	1.00	5.00	4.45	0.08	5.00
Purchasing cost (€/50 ml)	18.00	5.00	35.00	4.42	135.00	1.00
Tensile shear strength (MPa)	6.07	5.00	4.14	3.05	2.11	1.00

mechanical characterisation and LCA provides a robust framework for evaluating and advancing sustainable innovations. It enhances decision-making by considering a broad spectrum of factors, ultimately fostering the development of products and processes that are economically viable, environmentally responsible and technically sound.

The selection of the adhesive system to use ultimately depends on the specific priorities of the project and requires balancing multiple factors. Based on the experimental, statistical, and LCA findings, and considering the specific context in which LPP treatment is employed to prepare TPU-based composites, it can be concluded that PU adhesive is the best choice for an effective bonding process, offering high joint strength and low cost, although its significant environmental impact and long handling time may present notable drawbacks. In contrast, EP adhesive offers a more balanced solution when considering cost, joint strength and environmental impact, making it well-suited for projects needing good performance without compromising sustainability. Lastly, HY adhesive is particularly suitable for projects prioritising environmental sustainability and rapid application, despite its higher cost and lower performance. Specifically, low joint strength and the occurrence of deadhesive failure (at least in combination with plasma treatment) may restrict its use to less demanding applications. In this regard, the authors acknowledge that further research is needed to investigate the specific performance of HY adhesive, particularly with respect to alternative surface treatments and their effect on joint quality, as well as their techno-economic and environmental implications.

Overall, this analysis highlights the importance of considering not just the technical performance but also the economic and environmental implications when choosing adhesives for plasma-treated thermoplastic composites. By carefully weighing these factors, project managers can make informed decisions that align with their project goals and values.

CRediT authorship contribution statement

Chiara Mandolfino: Writing – original draft, Investigation, Conceptualization. **Lucia Cassettari:** Writing – original draft, Methodology, Conceptualization. **Marco Pizzorni:** Writing – review & editing, Validation, Data curation. **Luigi Benvenuto:** Writing – original draft, Validation, Investigation. **Enrico Lertora:** Writing – review & editing, Supervision, Project administration.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A

In Tables A1, A2 and A3, the failure modes exhibited by joints fabricated with polyurethane (PU), epoxy (EP), and hybrid (HY) adhesives, respectively, are indicated. Figs. A1–A3 illustrate the characteristic load-displacement curves obtained from tensile shear testing of joints, along with details of the typical fracture surfaces corresponding to the worst and the best plasma conditions.

Based on the results of the experimental tests and the statistical analysis, the influence of the power parameter appears to be predominant over the treatment time for both the PU and EP scenarios. Plasma treatment effectively increases joint strength, particularly at lower power level. Joints with surfaces treated at the minimum power level (50 W) – and, interestingly, with shorter treatment times (10 s) – exhibited the highest strength values, typically associated with failure modes that were almost fully cohesive in type, as shown in Figs. A1b and A2b. This situation suggests that low-power plasma treatment was effective in establishing strong and stable bonding conditions at the adhesive/substrate interface. However, as plasma power increases beyond 125 W, the fracture location shifts from within the adhesive to the interface with substrate (fully de-adhesive failure), resulting in reduced mechanical performance of joints.

Conversely, for the HY scenario, a clear influence of plasma treatment parameters cannot be determined. This is supported by Table A3 and Fig. A3: although some parameter combinations result in increased joint performance compared to control joints, the fracture consistently occurs at the adhesive interface across all cases.

Table A1
PU-scenario: Failure mode for different setup combinations.

Sample	Failure mode
Control Joint	Adhesive
PL_PU_50W_10s	Cohesive
PL_PU_50W_120s	Cohesive
PL_PU_50W_230s	Cohesive
PL_PU_125W_10s	Adhesive
PL_PU_125W_120s	Adhesive
PL_PU_125W_230s	Adhesive
PL_PU_200W_10s	Adhesive
PL_PU_200W_120s	Adhesive
PL_PU_200W_230s	Adhesive

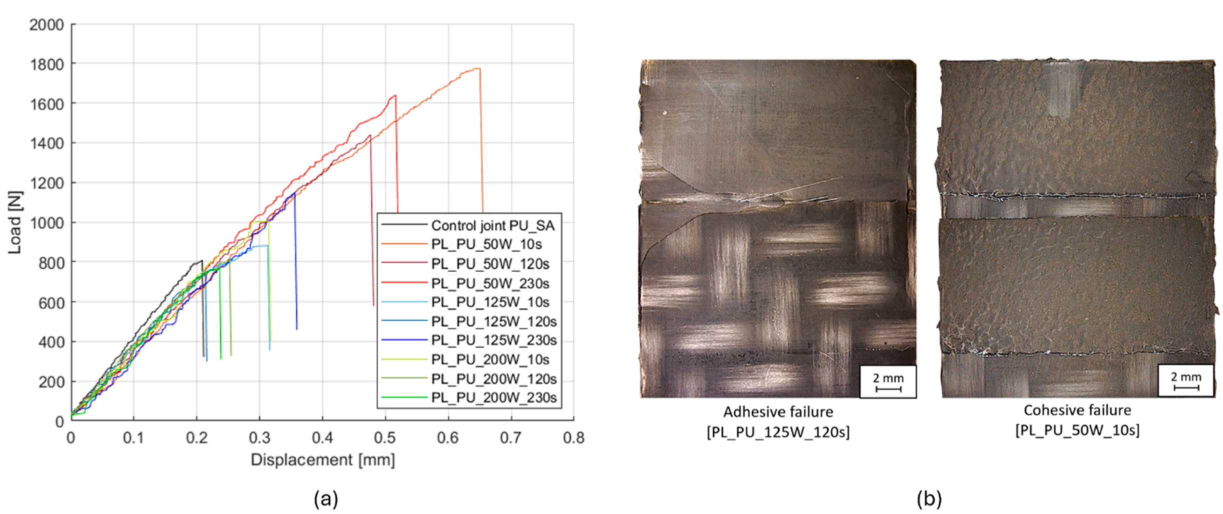

Fig. A1. PU-scenario: (a) load-displacement curves and (b) typical fracture surfaces corresponding to the worst (left) and the best (right) plasma conditions.

Table A2
EP-scenario: Failure mode for different setup combinations.

Sample	Failure mode
Control Joint	Adhesive
PL_EP_50W_10s	Mixed cohesive-adhesive
PL_EP_50W_120s	Mixed cohesive-adhesive
PL_EP_50W_230s	Mixed cohesive-adhesive
PL_EP_125W_10s	Adhesive
PL_EP_125W_120s	Adhesive

(continued on next page)

Table A2 (*continued*)

Sample	Failure mode
PL_EP_125W_230s	Adhesive
PL_EP_200W_10s	Adhesive
PL_EP_200W_120s	Adhesive
PL_EP_200W_230s	Adhesive

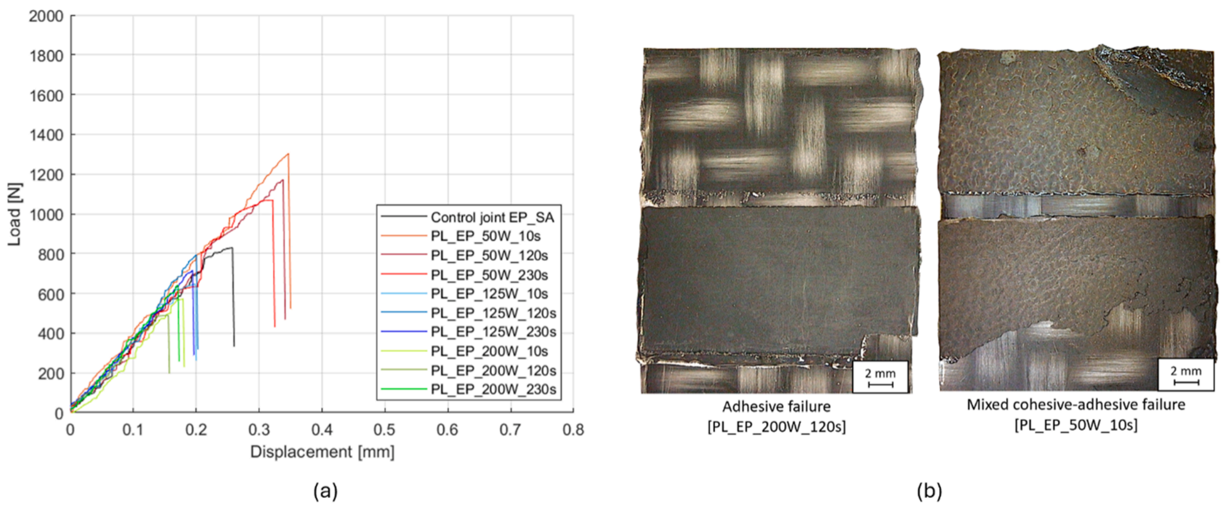

Fig. A2. EP-scenario: (a) load-displacement curves and (b) typical fracture surfaces corresponding to the worst (left) and the best (right) plasma conditions.

Table A3
HY-scenario: Failure mode for different setup combinations.

Sample	Failure mode
Control Joint	Adhesive
PL_HY_50W_10s	Adhesive
PL_HY_50W_120s	Adhesive
PL_HY_50W_230s	Adhesive
PL_HY_125W_10s	Adhesive
PL_HY_125W_120s	Adhesive
PL_HY_125W_230s	Adhesive
PL_HY_200W_10s	Adhesive
PL_HY_200W_120s	Adhesive
PL_HY_200W_230s	Adhesive

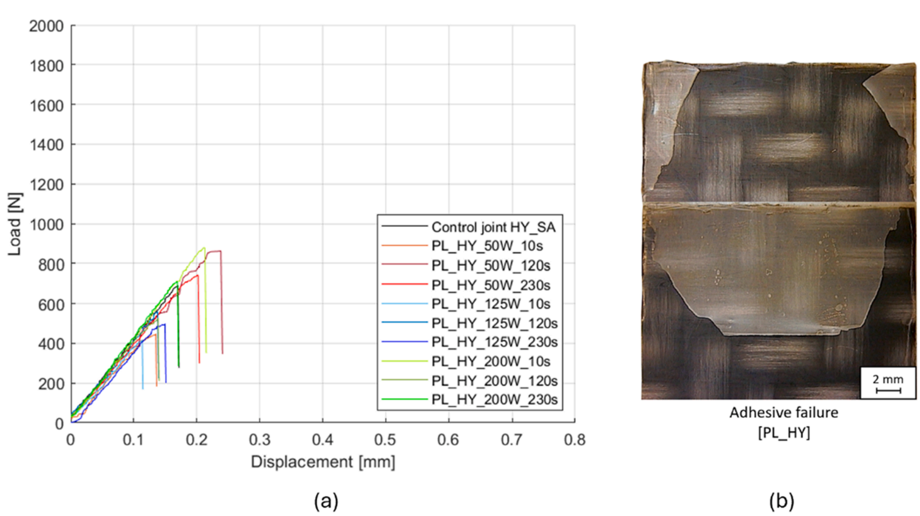

Fig. A3. HY-scenario: (a) load-displacement curves and (b) typical fracture surface: all joints failed de-adhesively.

Data availability

Data will be made available on request.

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