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Life cycle analysis of engineering polymer joining methods using adhesive bonding: fatigue performance and environmental implications

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Abstract

Traditional assembly processes such as screw fastening and riveting are increasingly being replaced by new processes such as adhesive bonding. Life cycle performance including fatigue and durability are critical, for which surface activation techniques are often used with the aim of improving both mechanical and life cycle performance. Within this context, the present paper aims to investigate the life cycle performance of adhesive bonding in relation to engineering polymers considering four surface pre-treatments: mechanical, chemical, plasma, and laser activation. The work focuses on two key aspects: (i) mechanical characterization of fatigue performance by assessing the useful life of joints, and (ii) environmental analysis through Life Cycle Assessment (LCA). The outcome of this study provides important insight into the development of laser and plasma technologies as sustainable surface activation methods for polymer joining methods. The substitution of traditional joining methods (i.e., bolting, riveting) with adhesive bonding will allow reductions in overall product weight to be achieved.

Keywords: assembly; design for assembly; adhesive bonding; fatigue; polymers; surface activation; environmental impact; LCA.

1. Introduction

Metal components are being replaced by engineering polymers in numerous applications such as structural elements due to the advantages of polymers in terms of wear resistance, density, corrosion resistance, flexibility, and cost [1]. Joining methods for these materials are still being studied, with adhesive bonding one of the most important techniques, along with welding and mechanical fasteners [2]. Adhesive bonding of engineering polymers has good growth outlook over the next few years [3]. Some of the main advantages of adhesive bonding include ease of manufacturing, better stress distribution over the joint area, the ability to join dissimilar materials, the possibility to couple thin materials, greater fatigue resistance and weight reduction [4]. However, this type of joint tends to present drawbacks such as the requirement for surface preparation [5], and environmental concerns (i.e., material separation) [6][7]. Several works have focused on reducing the environmental impact of adhesive use. The work of Attaf [8] provides new strategies for designing bonding processes in a sustainable way, guaranteeing protection of the environment and human health from potential impacts (i.e., toxic chemicals, harmful substances, carcinogens, VOCs, etc.). Studies such as that of McDevitt and Grigsby [9] have shown that the use of bio-based adhesives instead of those with petrochemical origins can reduce the life cycle impact by up to 22%. On the other hand, when adhesive bonding is used to couple engineering polymers, surface preparation is usually required to modify the surface chemistry and/or morphology, increasing the surface energy of low wettability polymeric substrates. These activation techniques can use different approaches such as chemical activation (i.e., primer or acids), which leads to important environmental impacts, or mechanical processes (i.e., abrasion, plasma or laser irradiation), which

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lead to greenhouse gas emissions due to energy consumption [10]. The present authors [11] recently conducted a Life Cycle Assessment (LCA) analysis where the environmental impact of different surface activation methods used for adhesive joints was studied in relation to their mechanical behavior. In that work it was demonstrated how the technique characterized by the greatest environmental impact was use of primer, while plasma and laser treatments achieved best results. However, mechanical characterization of these pre-treatments was only performed with static strength tests, which did not consider the service life of the adhesive bond. There is therefore scope to enlarge the results of the previous study by carrying out life cycle characterization of the adhesive bonded joints in terms of fatigue behavior.

The aim of the present work was therefore to investigate the life cycle performance of adhesive bonded polymer joints while considering four different surface pre-treatments: mechanical (i.e., abrasion), chemical (i.e., primer), plasma, and laser activation. The work focused on two aspects of pre-treated adhesive bonded joints: (i) mechanical characterization of fatigue performance by assessing the useful life of the joint, and (ii) environmental analysis through LCA. The employed method was divided into three phases. The first comprised definition of assembly procedure specifications for each pre-treatment. The second included mechanical characterization of each pre-treatment through experiments. The third comprised establishing the environmental load of each adhesive pre-treatment using LCA. Through analysis of the fatigue tests, pre-treatments can be compared based on the useful life of each, with the method leading to lowest impact to be deployed for mechanical applications where low weight and good environmental performance must be coupled. End-of-Life concerns fall outside the scope of this study, for which future works relating to this aspect will provide a holistic analysis of environmental performance.

2. Materials and Methods

The overall methodology consisted of three phases: (i) definition of specifications for adhesive bonded joint preparation; (ii) fatigue tests for mechanical characterization of each pre-treatment; and (iii) LCA analysis.

2.1. Definition of specifications for adhesive bonded joint preparation

Several pre-treatments can be adopted as activation processes for adhesive bonded joints. Based on a previous investigation by the present authors [11], the following pre-treatments were investigated:

- Mechanical activation – abrasion: aluminum oxide 320 grit sandpaper was used in line with ASTM D 2093 [12]. To remove residual particles, samples were cleaned with Henkel Loctite 7063.
- Chemical activation – primer: samples were initially abraded and then treated with the selected primer, Teroson 150 (supplied by Henkel - Italy). Before applying the adhesive, the primer was left to evaporate for 10 minutes.
- Plasma treatment: this activation process was performed following the methodology described by Moroni et al. [13], where PA 66 plates were subject to atmospheric pressure plasma treatment. The translational speed employed was 100 mm/s while the nozzle-to-substrate distance was 5 mm. The main characteristics of the plasma source (Manufacturer: Diener Plasma GmbH & Co. KG; Model: PlasmaBeam) are shown in Table 1.

Table 1. Characteristics of the plasma setup utilized for experiments.

Parameter	Symbol	Value
Generator power (capacity)	P	300 W
Process gas	G_p	Air supplied at 6 bar
Cooling gas	G_c	Air supplied at 6 bar

- Laser irradiation: this activation process was carried out using a nanosecond pulsed fiber laser. The main characteristics of the laser source (Manufacturer: LaserPoint; Model: YFL 20P) are shown in Table 2.

Table 2. Characteristics of the laser setup utilized for experiments.

Parameter	Symbol	Value
Average power (capacity)	P	1 – 17 W
Wavelength	λ	1064 nm
Pulse duration	τ	104 ns
Pulse energy	E_p	50 – 850 µJ
Focal length	f	160 mm
Maximum scanning velocity	v	2500 mm/s

The strength of adhesive bonded joints subject to laser pre-treatment depends on the process parameters employed, including scanning strategy, pulse energy (E_p), scanning velocity (v) and transverse hatch spacing (Th). To determine the optimal value of each parameter, a Design-of-Experiments (DoE) approach was adopted. This method had previously been applied by Lutey and Moroni [14] to calculate optimal process parameters for laser pre-treatment of adhesive-bonded polyethylene (PE) joints. In addition, Lutey et al. [15] verified that such an approach could be employed for optimizing the static joint strength of adhesive-bonded PA 66 joints. Ten specimens were prepared for fatigue tests using the combination of laser parameters achieving the highest static strength, shown in Table 3.

Table 3. Laser parameters employed for fatigue test.

Parameter	Symbol	Value
Scanning strategy	PL	Parallel line
Average power	P	3 W
Scanning velocity (capacity)	v	700 mm/s
Longitudinal pulse spacing	Lp	35 µm
Transverse (hatch) pulse spacing	Th	50 µm
Peak pulse fluence	F	10.6 J/cm^2
Total energy dose	E	8.6 J/cm^2

Energy consumption of each pre-treatment was measured for use as an input in the Life Cycle Inventory (LCI) during the environmental impact analysis phase. The engineering polymer used for the investigation was polyamide PA 66, supplied by Ensinger (Italy), with dimensions 100 mm (L – length) x 25 mm (W – width) x 6.6 mm (T – thickness). The adhesive selected was Teroson PU 9225, supplied by Henkel (Italy). Joints were manufactured with an adhesive thickness (Ta) of 0.3 mm and an overlap length (OL) equal to 18 mm. The configuration of the bonded joint is shown in Fig. 1. Experimental conditions and joint geometry were in line with ASTM D3163 [16]. Production of the adhesive joints included the following steps: (i) washing and degreasing the plates to remove dirt; (ii) surface pre-treatment; (iii) assembly of the plates with the adhesive, and (iv) curing at room temperature.

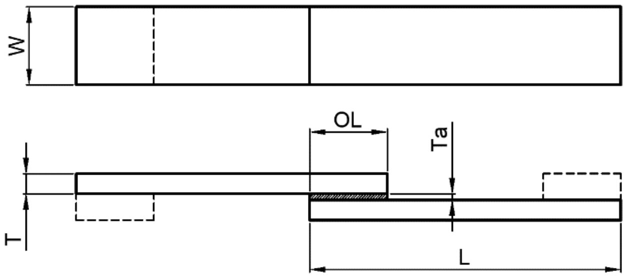

Fig. 1. Joint geometry and dimensions (based on ASTM D3163).

2.2. Fatigue tests for mechanical characterization of pre-treatments

The adhesive bonded joints described in the previous section were subject to fatigue tests to obtain the number of cycles to failure for different levels of applied load amplitude. Tests were carried out in line with ASTM D3166 [17]. This standard refers to metal adherends; however, its use was justified as there is no specific standard for polymeric bonded joints. A servo-hydraulic MTS 810 testing machine equipped with a 5 kN load cell was used. Tests were conducted under load control by imposing a sinusoidal wave with a load ratio (R) of 0.1 and a frequency of 6 Hz until final failure. For each pre-treatment that was considered, a set of ten samples was manufactured in line with the aforementioned specifications and tested under fatigue loading. The levels of applied load amplitude were selected to cover a range of cycles to failure between approximately 10^3 and 10^6. Fatigue tests were performed under constant environmental conditions (temperature and humidity) to ensure repeatability.

2.3. Life Cycle Assessment

In this phase, the methodology described in the ISO and ILCD Handbooks [18][19] was followed to compare the environmental performance of each pre-treatment. The functional unit was defined as: *"the process of surface activation for adhesive bonding of PA 66 polymers that guarantees a load amplitude of at least 380 [N] ± 10% and withstands at least 1000 cycles during fatigue tests"*. The minimum load amplitude and the number of cycles were chosen by considering that the pre-treatment with the worst results (chemical activation by primer). The adhesive-bonded joint employing chemical activation achieved approximately 1000 cycles with a load amplitude of 380 [N]. System boundaries included: (i) material extraction, (ii) joint manufacturing, and (iii) use. The joint manufacturing phase took into account the amount of adhesive used, as well as specific consumptions for each pre-treatment (i.e., electrical energy, cleaner, and primer). It is worth noting that adhesive bonding is a "clean process" when automated and no process waste is produced (i.e., excess adhesive). The only process producing by-products was the primer method, which was included in the analysis. Other treatments such as degreasing and cleaning were performed on all substrates and could therefore be excluded from the analysis. Maintenance and transportation phases were neglected since it was considered that they were the same for all studied configurations. The end-of-life (EoL) was also not contemplated as this aspect will be addressed in future works comparing not only adhesive joint pre-treatments, but also mechanical joining methods (i.e., bolted). Bolted joints present greater impacts during material and manufacturing phases due to the amount of material required; however, they allow disassembly of components and facilitate reuse, for which they are considered less problematic solution [20]. In the LCI, all materials and sources of energy consumption required for preparation of the adhesive bonded joints were considered. Table 4 shows the inventory for each pre-treatment.

Table 4. Data used in the LCI for adhesive joints.

Pre-treatment technique	Item	Material	Quantity
Abrasion	Adhesive PU 9225 (18 mm length)	PU 9225	0.21 [g]
	Cleaner Henkel Loctite 7063	Naphtha, ethanol and methylal	1.11 [g]
Primer	Adhesive PU 9225 (18 mm length)	PU 9225	0.21 [g]
	Cleaner Henkel Loctite 7063	Naphtha, ethanol and methylal	1.11 [g]
	Primer Teroson 150	Xylene and ethylbenzene	1.08 [g]
Plasma	Adhesive PU 9225 (18 mm length)	PU 9225	0.21 [g]
	Energy consumption		0.13 [Wh]
Laser	Adhesive PU 9225 (18 mm length)	PU 9225	0.21 [g]
	Energy consumption		0.45 [Wh]

Energy consumption was measured with a power meter, considering only the source and excluding peripheral devices. Tests were carried out in Italy for which the Italian grid mix was used as the energy source. The Ecoinvent database was utilized for secondary data and ReCiPe midpoint (H) was adopted as the Life Cycle Impact Assessment (LCIA) method. Midpoints shown in Table 5 were analyzed, together with endpoint impact categories: (i) human health (HH), ii) ecosystem (ED), and (iii) resources (RA).

Table 5. Midpoint impact category [21].

Name	Acronym	Unit
Global warming potential	GWP	[kg CO2 eq]
Ozone depletion potential	ODP	[kg CFC11 eq]
Terrestrial acidification potential	TAP	[kg SO2 eq]
Freshwater eutrophication potential	FEP	[kg P eq]
Human toxicity potential	HTP	[kg 1.4-DCB]
Particulate matter formation potential	PMFP	[kg PM2.5 eq]
Terrestrial ecotoxicity potential	TETP	[kg 1.4-DCB]
Freshwater ecotoxicity potential	FETP	[kg 1.4-DCB]
Marine ecotoxicity potential	METP	[kg 1.4-DCB]
Photochemical oxidant formation potential	OFP	[kg NOx eq]
Fossil fuel potential	FFP	[kg oil eq]
Water consumption potential	WCP	[m^3]

3. Results and discussion

In this section, results obtained for the fatigue tests and LCA analysis are reported.

3.1. Fatigue test results

Fig. 2 shows the results of fatigue tests performed on PA 66 adhesive bonded joints for all activation methods outlined above.

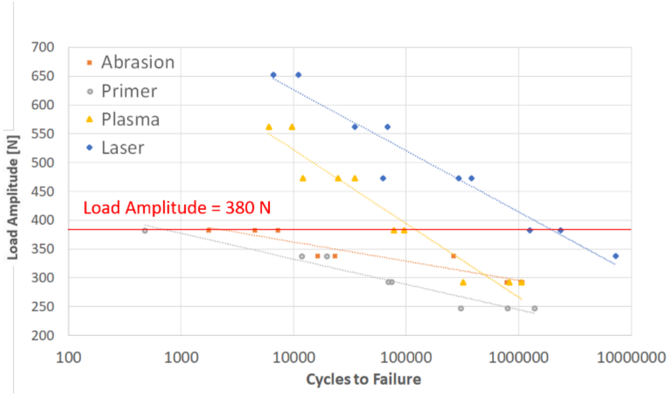

Fig. 2. Load amplitude vs Cycles to failure.

Best results were observed with laser irradiation, which achieved 7.3×10^6 cycles with a load amplitude of 0.34 kN and 6.7×10^3-1.1×10^4 cycles with a load amplitude of 0.65 kN. Plasma performed very well at high load amplitude (achieving 6.1×10^3-9.7×10^3 cycles with a load amplitude of 0.56 kN) but was outperformed at low load amplitude by abrasion (achieving 3.2×10^5-1.1×10^6 cycles with a load amplitude of 0.29 kN) due to the low surface roughness of plasma treatment. Abrasion achieved 7.8×10^5-1.1×10^6 cycles with a load amplitude of 0.29 kN and 1.7×10^3-7.3×10^3 cycles with a load amplitude of 0.38 kN. Finally, worst performance under cyclic loading was observed with the primer, which achieved 3.1×10^5-1.4×10^6 cycles with a load amplitude of 0.25 kN and 4.8×10^2 cycles with a load amplitude of 0.38 kN. These results prove that laser irradiation is the best pre-treatment for adhesive-bonded polymer joints in terms of fatigue resistance at both high and low load amplitude, representing a surface activation method that increases the useful life over a range of load amplitudes.

3.2. LCA results

Table 6 reports the results of the midpoint indicators of the LCA, while Table 7 shows the normalized endpoints. In both cases, results refer to the material and manufacturing phases. It is possible to appreciate how plasma treatment has the lowest impact in all categories, followed by laser irradiation, abrasion and lastly primer. Results relating to plasma and laser treatments are very similar, with a maximum difference of 9% in GWP and 8% in TAP, due to the fact that both methods use the same amount of adhesive. The only difference between them is energy consumption, which is slightly higher for laser irradiation. Abrasion and primer are characterized by greater environmental impact due to the use of chemical compounds such as primer and cleaner. The average difference between the technique with lowest impact (plasma) and that with the highest impact (primer) is 50%, with a maximum of 82% in the FFP midpoint due to the chemical composition of the cleaner.

Table 6. Midpoint impact category results – Material and manufacturing phase.

Midpoint	Abrasion	Primer	Plasma	Laser
GWP	2.13×10^{-3}	4.24×10^{-3}	1.48×10^{-3}	1.62×10^{-3}
ODP	3.80×10^{-9}	3.90×10^{-9}	3.07×10^{-9}	3.19×10^{-9}
TAP	1.01×10^{-5}	1.51×10^{-5}	6.21×10^{-6}	6.72×10^{-6}
FEP	8.10×10^{-7}	1.09×10^{-6}	6.40×10^{-7}	6.76×10^{-7}
HTP	2.40×10^{-3}	3.04×10^{-3}	1.93×10^{-3}	2.01×10^{-3}
PMFP	4.30×10^{-6}	6.38×10^{-6}	2.97×10^{-6}	3.14×10^{-6}
TETP	7.36×10^{-3}	1.04×10^{-2}	5.27×10^{-3}	5.42×10^{-3}
FETP	1.42×10^{-4}	1.69×10^{-4}	1.15×10^{-4}	1.19×10^{-4}
METP	1.56×10^{-4}	1.93×10^{-4}	1.20×10^{-4}	1.25×10^{-4}
OFP	1.29×10^{-5}	2.11×10^{-5}	8.11×10^{-6}	8.62×10^{-6}
FFP	1.77×10^{-3}	3.37×10^{-3}	6.05×10^{-4}	6.48×10^{-4}
WCP	4.93×10^{-5}	8.31×10^{-5}	3.35×10^{-5}	3.57×10^{-5}

Table 7. Endpoint impact category results – Material and manufacturing phase.

Endpoint	Abrasion	Primer	Plasma	Laser
HH	2.39×10^{-7}	3.92×10^{-7}	1.70×10^{-7}	1.82×10^{-7}
ED	1.50×10^{-8}	2.66×10^{-8}	1.01×10^{-8}	1.10×10^{-8}
RA	2.47×10^{-8}	4.69×10^{-8}	6.38×10^{-9}	6.83×10^{-9}

Fig. 3 shows how most of the GWP impact in almost all treatments is due to the adhesive, except for chemical activation pre-treatments where the contribution of the primer is slightly higher. In relation to the contribution of electrical energy for plasma and laser treatments, this is not significant and accounts for 5% and 13% of the overall impact, respectively. The Italian grid mix employed is mostly characterized by non-renewable resources. The adoption of other grid mixes mainly characterized by renewable energy would therefore be beneficial to the final results, reducing the impact of both pre-treatments.

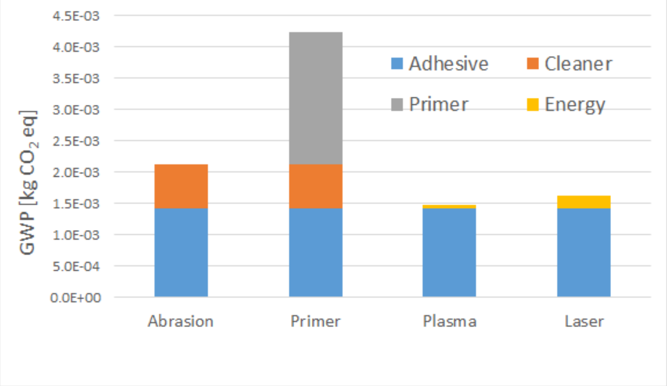

Fig. 3. Contribution of each item to GWP indicator – Material and manufacturing phase.

Thus, the impact related to the energy consumption in laser and plasma methods is much lower than that of the adhesive, with a difference of one order of magnitude for the laser and up to two orders of magnitude for the plasma. Similar trends can be observed for the other midpoints (i.e., HTP), which are even less sensitive to the influence of electrical energy and thus to the type of the grid mix. However, due to the nature of fatigue tests, it is possible to calculate the environmental impact considering the useful life of the adhesive bonded polymer joint. For this, the joint must withstand a load of at least 380 N (as outlined in the functional unit). Using the equations of the trendlines in Fig. 2, the number of cycles that each pre-treatment technique withstands can be calculated. The fatigue performance of all pre-treatments is reported in Table 8.

Table 8. Fatigue performance of pre-treatments 380 N load

Fatigue performance	Abrasion	Primer	Plasma	Laser
Number of cycle [cycles]	2473	4744	123759	2005130

Equation (1) can be used to calculate the environmental impact considering the useful life by applying a corrective coefficient.

$$I_{use,i} = \frac{N_1}{N_i} \times I_{manufacturing,i} \tag{1}$$

- N_1: cycles to failure of pre-treatment achieving shortest lifespan (i.e., the primer).
- N_i: cycles to failure of the *i-th* type of pre-treatment.
- $I_{manufacturing,i}$: environmental impact during material and manufacturing phase of the *i-th* type of pre-treatment.
- $I_{use,i}$: environmental impact considering the use phase of the *i-th* type of pre-treatment.

Results obtained in this manner, including the use phase, are shown in Table 9 and Table 10.

Table 9. Midpoint impact category results – whole life cycle.

Midpoint	Abrasion	Primer	Plasma	Laser
GWP	6.41×10^{-4}	4.24×10^{-3}	8.88×10^{-6}	6.01×10^{-7}
ODP	1.14×10^{-9}	3.90×10^{-9}	1.85×10^{-11}	1.18×10^{-12}
TAP	3.04×10^{-6}	1.51×10^{-5}	$3.73\text{E}\times10^{-8}$	2.49×10^{-9}
FEP	2.44×10^{-7}	1.09×10^{-6}	3.85×10^{-9}	2.51×10^{-10}
HTP	7.22×10^{-4}	3.04×10^{-3}	1.16×10^{-5}	7.46×10^{-7}
PMFP	1.29×10^{-6}	6.38×10^{-6}	1.79×10^{-8}	1.17×10^{-9}
TETP	2.21×10^{-3}	1.04×10^{-2}	3.17×10^{-5}	2.01×10^{-6}
FETP	4.26×10^{-5}	1.69×10^{-4}	6.92×10^{-7}	4.40×10^{-8}
METP	4.69×10^{-5}	1.93×10^{-4}	7.22×10^{-7}	4.63×10^{-8}
OFP	3.89×10^{-6}	2.11×10^{-5}	4.88×10^{-8}	3.20×10^{-9}
FFP	5.33×10^{-4}	3.37×10^{-3}	3.64×10^{-6}	2.40×10^{-7}
WCP	1.48×10^{-5}	8.31×10^{-5}	2.02×10^{-7}	1.33×10^{-8}

Table 10. Endpoint impact category results – whole life cycle.

Endpoint	Abrasion	Primer	Plasma	Laser
HH	7.21×10^{-8}	3.92×10^{-7}	1.02×10^{-9}	6.75×10^{-11}
ED	4.52×10^{-9}	2.66×10^{-8}	6.10×10^{-11}	4.09×10^{-12}
RA	7.45×10^{-9}	4.69×10^{-8}	3.84×10^{-11}	2.53×10^{-12}

By taking service life into account, laser irradiation becomes the technique with lowest environmental impact, approximately one order of magnitude lower than plasma treatment in almost all categories. Fig. 4 displays the results for the normalized endpoints, allowing the very large difference between abrasion and primer pre-treatments with plasma or laser treatments to be appreciated (the latter having a negligible contribution).

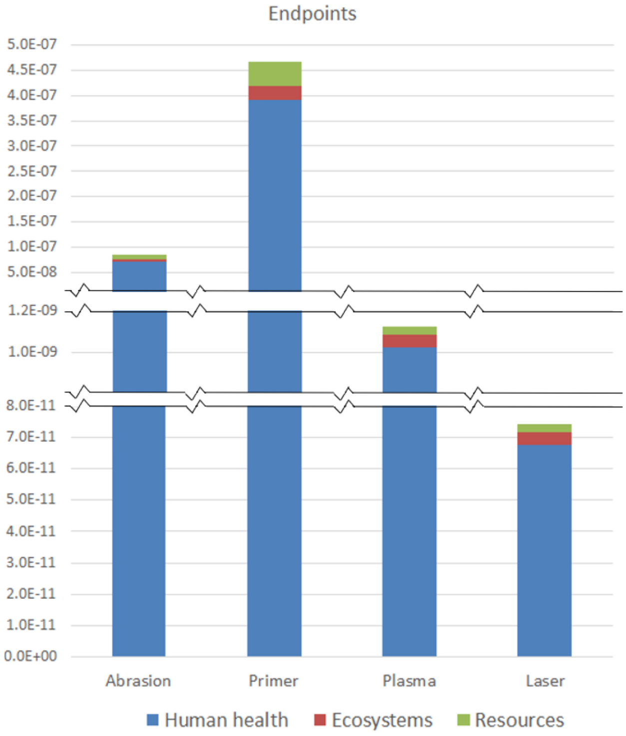

Fig. 4. Impact assessment (endpoints) considering service life.

Comparing primer with abrasion, it can be seen how primer has an impact five times higher in HH and six times higher in

ED and RA. In addition, Fig. 4, together with Table 10, also highlights the fact that the HH category is most affected by all pre-treatments (an order of magnitude higher than the ED and RA categories).

4. Conclusion

Through an LCA analysis it has been possible to compare the environmental impact of different pre-treatments (abrasion, primer, plasma and laser) performed on PA 66 joints bonded with Teroson PU 9225 adhesive. Mechanical characterization of the joints was conducted via fatigue tests, allowing the behavior of joints prepared with each pre-treatment to be determined under dynamic loading. These tests sought to determine trends relating to the mechanical behavior of each activation method to correct environmental impact results by considering the service life of bonded joints when subject to a cyclical load of 380 N. Results showed how laser and plasma treatments were the techniques with lowest environmental impact, with a large difference observed between these and abrasion and primer pretreatments. It was found that the impact of the primer component was 1.5 times that of the adhesive itself, while the impact of the cleaner was half that of the adhesive. The impact of energy consumption was much lower than that of the adhesive, about 7 times less for laser irradiation and almost 24 times less for plasma treatment. The outcomes of this study show how plasma and laser can be considered sustainable surface activation technologies. Nanosecond pulsed laser irradiation with optimized parameters was shown to be an effective technique for improving the fatigue strength of PA 66 adhesive bonded joints, leading to lowest environmental impact of all tested pretreatments when service life was taken into account. This outcome will stimulate deployment of this technology for the production of adhesive bonded joints within assembled mechanical products. Environmental benefits, in terms of impact reduction for life cycle indicators, will be observed in mechanical products that substitute the use of traditional joints (i.e., bolt, rivets) with adhesive bonded joints providing lightweight solutions (reduction of energy/fuel consumption during product use). While joining processes may have major environmental impacts when compared to one another, at a system level (e.g., as noted previously in relation to disassembly) the LCA results may be very different with the necessity to enlarge the system boundaries. A set of case studies (products) using the different joining systems would be helpful for interpretation of results. A trolley wheel made of PA, subject to a high number of cycles, and a charge-air duct made of PA, subject to a low number of cycles (thermal stress), will be analyzed in subsequent works to achieve this aim. Thus, future works will be focused on system application of these assembly methods, calculating environmental benefits in different fields of application (i.e., automotive, naval transport, aeronautical field, industrial machinery). Additional research will be performed to consider end-of-life aspects for this type of joint. In particular, novel technologies will be investigated (e.g., reversible adhesives, active debonding, etc.) to couple mechanical performance with suitable solutions for correct management of these joints at the end-of-life.

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