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LCA FOR MANUFACTURING AND NANOTECHNOLOGY

Environmental assessment of an innovative adhesive for the footwear industry: road map for product development

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Abstract

Purpose This work presents an environmental assessment of two adhesives to evaluate the replacement of the traditional adhesive (PU) used in the footwear industry by a novel adhesive based on a microencapsulation approach (PUMC) which is more favorable from the safety and technical perspectives. The PU adhesive is polyurethane-based, while the PUMC adhesive is polyurethane-based but with its isocyanate compounds microencapsulated, increasing storage life and reducing risks related to the exposure of workers with the adhesives.

Methods The potential environmental impact of the adhesives was evaluated using the life cycle assessment (LCA) methodology. A detailed process model was developed (both for laboratory and for a pilot-scale implementation) to investigate the environmental impacts associated with these processes. The functional unit was one kilogram of adhesive (PU and PUMC adhesives) produced. A cradle-to-customers' gate approach was defined. The system boundary starts from extraction of resources, through material production, until adhesive use. This includes microcapsule production in the case of the PUMC adhesive. This study investigates the important drivers behind the environmental impacts to help guide commercialization efforts. A scenarios study/sensitivity analysis was conducted to determine the response of the PUMC adhesive system to the variability of the model, scenarios, and parameters.

Results The results show that the PU adhesive environmental impact is due to acetone and polyol consumption in the production stage. In the PUMC adhesive system, acetone consumption and microcapsule production are the major factors responsible for the environmental impact. Polybutylene adipate terephthalate (PBAT), dichloromethane (DCM), and isophorone diisocyanate (IPDI) consumption are the major factors responsible for the environmental impact of the microcapsules' production. A sensitivity analysis was conducted using three alternative scenarios focused on the reduction in material consumption and increase in material recuperation, as well as using an alternative renewable energy source. Although the traditional PU adhesive has a lower impact, it was found that the three alternative PUMC adhesive systems can become comparable to the traditional PU adhesive.

Conclusions This study shows the advance and development of a new technology for microencapsulation of isocyanate in adhesives and its environmental advantages and disadvantages with respect to a traditional product that uses non-encapsulated isocyanate. Finally, it was shown that there is significant potential for minimizing some environmental impacts of the PUMC adhesive, such as optimizing the microcapsules' production stage, increasing the production efficiency to decrease the required material consumption.

Keywords Life cycle assessment · Cradle-to-customer · Adhesives · Microencapsulation · Isocyanate · Polyurethane · Occupational Health and Safety

1 Introduction

The world footwear production in 2020 was 20.5 billion pairs, representing a relevant sector of the commercial market, even though the COVID-19 pandemic hit the footwear business severely, leading to a reduction in world's production by almost 4 billion pairs compared to the previous year. The impact of the pandemic was transversal, and at an aggregate level, it did not significantly change the geographical distribution of footwear production. Asia continues to be responsible for nearly nine out of ten pairs of footwear produced and has even increased its share by 0.1% points. Africa and Europe also achieved slight increases in share,

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Extended author information available on the last page of the article at the expense of North and South America, with Oceania playing a minor role in the sector (APICCAPS 2021a).

Portugal accounts for 0.3% points of the world's production (66 million pairs), being the vigesimal footwear producer of the world. However, Portugal has a strong tendency to export, exporting 93.2% points of its production (61 million pairs) and charging an average export price of more than 20 dollars per pair (APICCAPS 2021a). Despite the numerous challenges faced in 2020, the Portuguese footwear industry managed to export nearly 1.5 billion euros. The following year, in 2021, the industry experienced significant growth of 12% in foreign markets (APICCAPS 2021b). Furthermore, this industry has been an example and a reference for the Portuguese economy (DGAE 2017), as it has established itself in markets worldwide such as France, German, Netherlands, Spain, and UK (APICCAPS 2021a), is responsible for a significant part of exports, and is one of the most dynamic in the business sector. In addition, as stated by the Portuguese Agency for Investment and Foreign Trade (AICEP), the footwear industry in Portugal is currently recognized as one of the most innovative and competitive sectors within the country's economy. More than 1500 Portuguese companies operate in the footwear, components, and leather goods sectors, employing 40,000 people (APICCAPS 2021b). For these reasons, it has become essential to increase the degree of sustainability of this sector.

Footwear manufacturing involves transformation and assembly of various components made up of several materials where different adhesives play a key role, because, without them, the shoe would lack shape and structure (Orgilés-Calpena et al. 2019; Paiva et al. 2016a, b). A single running shoe can contain 65 discrete parts that require 360 processing steps for assembly (Cheah et al. 2013). In spite of the important function of the adhesives, their actual content is normally very low in a bonded product and is in most cases less than 1% of the final product weight (Cheah et al. 2013; Industrieverband Klebstoffe e.V. 2014). Nevertheless, footwear workers are routinely exposed to complex mixtures of solvents in them, such as toluene, n-hexane, and acetone (Gargouri et al. 2016; Heuser et al. 2005; Mayan et al. 2010; Staikos et al. 2006).

Like most industrial processes, the production and use of adhesives generates pollutants which can have an adverse health and environmental effect. The European adhesive market in 2022 was 4 million tonnes and is forecasted that its demand will increase in volume between 2021 and 2028 (FEICA 2024). Adhesives are a major industrial source of volatile organic compounds (Gargouri et al. 2016; Metzger and Eissen 2004; Packham 2009; Staikos et al. 2006). Much attention has been given in recent years to reducing their impact. It is now generally recognized that the emission of any volatile organic compound to the atmosphere is undesirable, as they contribute to the formation of photochemical smog, absorb infra-red radiation, and therefore act as greenhouse gases, and many are implicated in the aggravation of lung diseases such as asthma (Packham 2009). Isocyanates are regarded as one of the main causes of occupational asthma (Baur et al. 1994; Ameille et al. 2003; Lefkowitz et al. 2015; Gomez-Lopez et al. 2021; Karlsson et al. 2022). The large number of workers who are exposed to these chemicals has a concentration-dependent risk of developing chronic airway disorders, especially bronchial asthma (Baur et al. 1994; Coureau et al. 2021). Several studies describe the isocyanate emission potential of polyurethane adhesives (Heuser et al. 2005; Wirts et al. 2002) that may spread in aerosolized or gaseous form when heating or when vapors escape to the workplace air from open vessels at room temperature (Coureau et al. 2021; Heuser et al. 2005; Paal et al. 2002; Zhong and Siegel 2000). Respiratory disorders associated with isocyanate exposure (Collins 2002; Heuser et al. 2005; Paal et al. 2002; Skarping et al. 1996), and toxicity and/or genotoxicity, even in polymerized form, are described in several publications (Andersen et al. 1980; Bilban 2004; Collins 2002; Heuser et al. 2005; Coureau et al. 2021; Kligerman et al. 1987; Maki-paakkanen and Norppa 1987; Mori et al. 1988; Zhong and Siegel 2000) and occupational exposures (Heuser et al. 2005; Karlsson et al. 2022; Leng 2016; Marczynski et al. 1992). Isocyanate has been classified as a carcinogen in animals (Heuser et al. 2005; IARC 1999; NTP 1986; Senthilkumar et al. 2012) and is a suspected carcinogen in humans (Heuser et al. 2005; IARC 1999; Senthilkumar et al. 2012).

The recognition of the potential health-hazards and environmental impacts of solvent-based adhesives has led to the development of adhesives with no organic solvents. Many adhesive systems formerly based on organic solvents are now produced as aqueous emulsions. Polyurethanes (PU) have become one of the most widely used classes of polymers and today are found in many high-performance materials, such as adhesives (Nasar et al. 1998; Paiva et al. 2016b). Nonetheless, guided by environmental concerns and legal obligations, greener and safer alternatives to conventional PUs are now being sought to avoid the use of toxic isocyanates that are one of the primary components used in their formulation. Despite all these developments, volatile emissions associated with adhesive application remain high (Coureau et al. 2021; Metzger and Eissen 2004; Packham 2009). Furthermore, some studies emphasize that the performance of many newly developed products in this field is not benchmarked to those of commercial analogues, which makes direct comparison difficult (Gomez-Lopez et al. 2021).

Several authors already see the life cycle assessment (LCA) methodology as a very useful tool for evaluating the environmental impact of footwear, and in particular of adhesives (Eisen et al. 2020; Maciel et al. 2017; Packham

2009; Yang and Rosentrater 2019). According to Milà et al. (1998) the manufacturing stage showed the main environmental burdens that contribute to the environmental impact of the footwear product system life cycle. This occurs mainly due to energy requirements in many shoe manufacturing steps, including drying both adhesives and primers, which leads to another environmental burden: organic emissions (Borchardt et al. 2011; Cheah et al. 2013; Maciel et al. 2017). However, this study did not include the adhesive production because they considered the weight of this component to be negligible. Albers et al. (2008) analyzed 4 different models of shoes; however, they exclude the adhesives as they considered the weight of this component to be negligible in the analysis. The same can be said for the case of the study by Milà et al. (1998), which also excluded the adhesives from the analysis. Packham (2009) discussed the significant environmental impacts of adhesive technology based on a qualitative assessment. However, he stresses the need of the application of a holistic life-cycle analysis already during adhesive development. Also, it is pointed out that there are already improvements, but also in the future, there is still a great need for research on renewable raw material sources, energy savings and the avoidance of emissions. Cheah et al. (2013), performed a carbon footprint study from cradle to grave, or life cycle Global Warming Potential (GWP), of a pair of running shoes and suggested strategies to reduce the product's impact. The results indicated that most of the emissions are released during shoes' material processing (29%) and manufacturing phase (68%). Concluding that the polypropylene glycol (PPG) adhesive component contributes only 1% to the total environmental impact. Muñoz (2008) applied the LCA methodology (analyzing water consumption, energy consumption and GWP indicators) to a pair of leather shoes, considering the entire life cycle of the shoes including the bonding of the different components. This study considered that a pair of leather shoes uses 168 g of adhesive and that this contributes to the GWP by 0.13456 kg CO_2 eq/pair of leather shoes. According to Paiva et al. (2016b), the footwear industry has a close association with the adhesive industry, using bonding techniques to join the variety of materials employed in assembling shoes. However, this study did not perform a full life cycle assessment. Yang and Rosentrater (2019) also complain that there are insufficient comparisons from other studies between petrochemical-based adhesives and bio-based adhesives that could be used for comparisons, which is a limitation in the interpretation of LCA results. Maciel et al. (2017) studied three polyurethane adhesive technologies used in the footwear industry: a solvent-based adhesive (SBA), a water-based adhesive (WBA), and a powder-based adhesive (PBA), using the LCA methodology. The analysis showed that any actions that seek to minimize environmental impacts should begin in "the footwear industry," more precisely, in the stage of adhesive use due to the electricity required during the adhesive application. Therefore, a better management of the energy expended during the application step is suggested from renewable energy sources, improvement of equipment energy efficiency, and development of new formulations are potential alternatives for solutions seeking to reduce environmental impacts involving all adhesive technologies and consequently shoe production. Also, the studies analyzed in the literature review by Eisen et al. (2020), in which adhesives are considered, show that adhesives in particular usually have a very large influence on the LCA of the product system. Nevertheless, the environmental impacts were often only examined in relation to the entire product system, which meant that the environmental impacts of the adhesive technologies were always in relation to the entire product system and the adhesive itself was therefore difficult to assess. On the whole, it is positive to note that the number of relevant studies has increased in recent years and thus indicates an emerging relevance for the topic.

Nowadays, the LCA methodology is a well-established and widespread, though still evolving, tool and the only internationally standardized environmental assessment method (ISO 2006a; b). These and other considerations led to an increasing number of LCA-related publications, namely, those referring to adhesive production and application (Eisen et al. 2020; Gonzalezet al. 2017; Liu et al. 2018; Maciel et al. 2017; Packham 2009; Yang and Rosentrater 2019).

For all these reasons, there is a need of increasing the use of technical ingenuity in order to develop ways of addressing all or most of these problems. In view of this conjuncture, an advanced technology (microencapsulation) was applied to develop a new adhesive, with microencapsulation of the isocyanate compounds. The microencapsulation of the isocyanate eliminates the risks associated with their direct handling, protects the isocyanate species from air moisture, increases the storage life, and at the same time offers control over its triggered release (Aguiar et al. 2023a; Loureiro et al. 2023). It must be emphasized that a broad view of environmental impact must be taken. For this reason, it has become essential to assess the life cycle environmental impact of the newly developed adhesive when compared to commercial adhesive for the footwear industry. The main objective was to evaluate the possible replacement of the traditional adhesive used in the footwear industry (PU adhesive) by other safely and technically more favorable (PUMC adhesive). It is important to state that a simultaneous technological validation study demonstrated the feasibility of replacing the traditional adhesive by the novel one, ensuring at least a similar technical performance of the adhesives.

2 Methodology–life cycle assessment

The potential environmental impact of the adhesives (PU and PUMC adhesives) was evaluated using the LCA methodology, which includes all stages of a product's life (Bauman and Tillman 2004; Finnveden et al. 2009; Guinée, 2002; Pennington et al. 2004; Rebitzer et al. 2004). The methodology was performed in accordance with the standards from the ISO 14040 series (ISO 2006a; b). It comprises four major stages, the definition of goal and scope, life cycle inventory analysis (LCI), life cycle impact assessment (LCIA), and results' interpretation.

2.1 Goal and scope definition

The PU adhesive is a polyurethane-based adhesive, which is blended with isocyanate compounds to speed up the curing process, increase the temperature resistance, and improve the endurance of the adhesive joint. These sorts of adhesives are often classified as bicomponent adhesives since they are composed of two components: polyol adhesive and isocyanate cross-linker. They have to be provided separately because when they are blended the lifetime of the mixture is low (Aguiar et al. 2023a). The PUMC adhesive is also a polyurethane-based adhesive, but in this case, the isocyanate compounds are microencapsulated, reducing the risks related to isocyanates' direct handling by workers and increasing their storage life. The components of this adhesive have to be also provided separately because when they are blended the quality of the mixture is reduced; therefore, blending should be performed when the adhesive is going to be used. The main objective of the LCA study is to evaluate the potential environmental impact of the PU adhesive in comparison with the PUMC adhesive and also to investigate the important drivers behind these environmental impacts, in order to guide commercialization efforts. A detailed process model was developed (based on laboratory and pilot-scale data) to investigate the environmental impacts associated with these processes. A scenarios study/sensitivity analysis was conducted to determine the response of the PUMC adhesive system to model, scenario, and parameter variability.

The functional unit (FU) defined in this study was one kilogram (1 kg) of adhesive (PU and PUMC adhesives) produced. This FU was defined, because it was considered that the adhesives under consideration have similar technical performance that was verified in the technological validation study (Aguiar et al 2023a).

A cradle-to-customers' gate approach was defined. The life cycle stages of using the adhesive in the footwear industry, shoe use, and shoe End of Life (EoL) were not considered in the LCA analysis, because it is assumed that these processes are similar in both systems (PU and PUMC adhesives). The system boundary starts from extraction of resources, through material production, until adhesive use. This includes the raw material extraction and material production, material transportation to the adhesive plant, adhesive production (including microcapsule production in the case of the PUMC adhesive), adhesive packaging production and transport to the adhesive plant, and final adhesive transport to the customer (Fig. 1). In the case of the PUMC adhesive, it is assumed that microcapsules are produced in the adhesive production plant, and therefore there is no additional transport of this component.

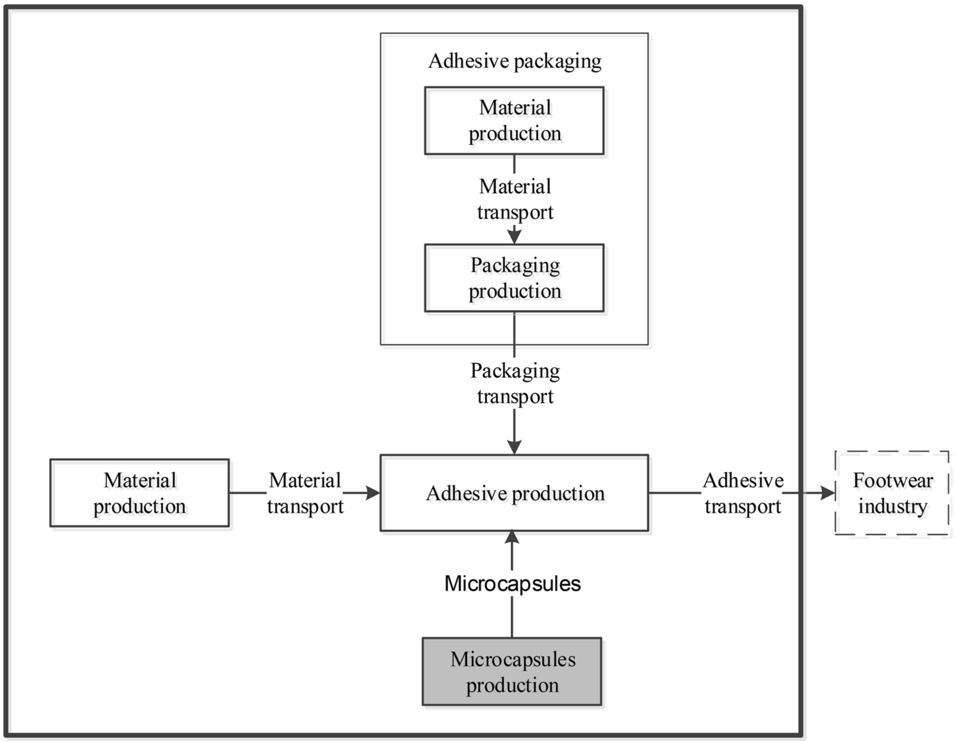
In the present work, the same polymer base, polyurethane, was used in the production of the adhesives (PU and PUMC adhesives). The solvent-based adhesive works with PU dissolved in organic solvents, and its production is simple. The process begins with a solvent mixture, and after that polymers and additives are introduced and blended. Last, the final product is packaged, usually into metal (tinplate) containers. The isocyanate cross-linker is also mixed with an organic solvent and packaged separately, generally into plastic (polypropylene, PP) containers. Therefore, this adhesive is supplied in two packages and only mixed at the final user (footwear industry).

The PUMC adhesive production, beyond the PU component that is packed as usual, requires the production of microcapsules using an innovative technology developed by companies involved in this study (Aguiar et al. 2023a, b). These microcapsules are used to microencapsulate the isocyanate compounds (used in the curing process of the adhesive). In order to determine the optimal protocol for achieving similar technical performance to PU adhesive, laboratory-scale experiments were conducted to gather information on the microencapsulation process sequence and microcapsule yields. Various compounds and procedures were tested in these experiments to assess their effectiveness. A pilot-scale installation was designed based on laboratory-scale experiments and built (fully functional) at the adhesive plant company. The isocyanate microencapsulation process consists of a two-stage process where material components are fed to a first reactor, with the resulting solution forwarded to a second reactor where more material components are added, and the microcapsules are formed. Finally, the microcapsule solution is filtrated, and the microcapsules are washed, dried and packed into plastic (polyethylene terephthalate, PET) containers (Fig. 2). As the PU adhesive system, this adhesive is also supplied in two packages and only mixed at the final user (footwear industry).

2.2 Life cycle inventory

The LCI was completed using primary data collected in an adhesive production plant. All flows associated with

Fig. 1 System boundary of the two adhesive (PU and PUMC adhesives) technologies



equipment and capital goods employed during the PU and PUMC adhesives technology life cycle were left out of this study, since it is not usual to consider the infrastructure in LCA studies (Guinée 2002), and also there is a lack of available LCA data on infrastructure related to adhesives. The LCIs attributed to the two systems in this study are presented in Tables 1 and 2, and were built following the methodology and considerations already discussed. The input and output flows for each technology are linked with enough amounts to produce 1 kg of adhesive (FU). As is well known, the selected LCI database influences the results of the LCA study, being one of its limitations. The Ecoinvent database was selected since it is a globally leading database and has a long history of gathering data (Kalverkamp et al. 2020). All the systems have been modeled by means of the commercial Ecoinvent v3.4 database (The Swiss Centre for Life Cycle Inventories 2017) and, whenever possible, using field data from the companies involved in the study, which were ultimately summarized in the LCI that was performed in SimaPro 8.5 (Pré Consultants 2017). Some data did not exist in the Ecoinvent database, and their inventory was collected from other sources, namely, European Life cycle Database (ELCD) (European Commission 2017) and Industry data 2.0 (Pré Consultants 2017).

2.3 Life cycle impact assessment

The Impact 2002 + V2.14 method was applied to assess the midpoint impacts (Humbert et al. 2012). This method allows the analysis of 15 environmental categories, as shown in Table 3. The Impact 2002 + model is well adapted to European conditions, which is consistent with the geographical scope of this LCA (Humbert et al. 2012). This analysis is used seeking to identify which substances or processes are responsible for substantial contributions to the environmental interventions. These results are shown in percentage over total, since the objective is to identify which substance or process present the biggest environmental impact contribution among the two systems in study. Also, the results were normalized, in order to better understand the magnitude of the category indicator results relative to the environmental impact of a European person. The normalization is achieved by dividing the environmental impact results for the system under study, by the environmental impact

Fig. 2 System boundary of the microcapsule production stage

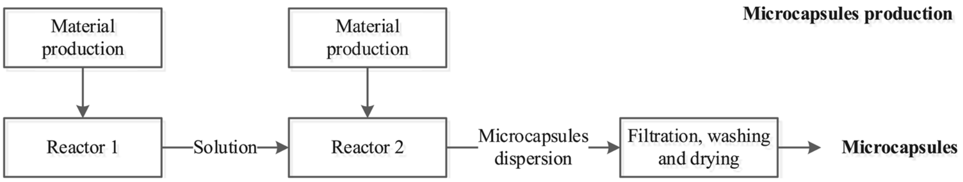


Table 1 LCI results of PU adhesive system to 1 kg of adhesive (FU)

Flows	Amount/FU	Unit	Data source/database
Materials			
Hydrophilic fumed silica	1.91E – 02	kg	Primary data/Ecoinvent
Fumaric acid	1.15E – 03	kg	Primary data/Ecoinvent
Linear hydroxyl polyurethane	1.37E – 01	kg	Primary data/Ecoinvent
High crystallization polyurethane	1.59E – 02	kg	Primary data/Ecoinvent
Acetone	7.81E – 01	kg	Primary data/Ecoinvent
Monopropylene glycol	9.55E – 04	kg	Primary data/Ecoinvent
TDI isocyanate	3.60E – 02	kg	Primary data/Ecoinvent
Ethyl acetate	9.01E – 03	kg	Primary data/Ecoinvent
PU adhesive production			
Transport of materials	879.253	kg·km	Estimated/Ecoinvent
Energy (electricity)	0.085	kWh	Primary data/Ecoinvent
PU adhesive packaging			
Tinplate	7.12E – 02	kg	Primary data/ELCD
Polypropylene (PP)	4.95E – 03	kg	Primary data/Ecoinvent
Production of tinplate packaging	7.12E – 02	kg	Estimated/Ecoinvent
Production of PP packaging	4.95E – 03	kg	Estimated/Ecoinvent
Packing transport	9.57E – 01	kg·km	Primary data/Ecoinvent
PU adhesive transport			
PU adhesive transport	96.851	kg·km	Estimated/Ecoinvent

of the normalization reference. Finally, the environmental category indicator results were also weighted and the addition of categories into one single score was done, allowing a direct comparison of the environmental impact of the two adhesive systems and different scenarios (Pizzol et al. 2017). The Impact 2002 + method performs normalization and weighting at damage category level. Table 3 presents the normalization and weighting factors.

Table 2 LCI results of PUMC adhesive system to 1 kg of adhesive (FU)

Flows	Amount/FU	Unit	Data source/database
Materials			
Hydrophilic fumed silica	1.85E – 02	kg	Primary data/Ecoinvent
Fumaric acid	1.11E – 03	kg	Primary data/Ecoinvent
Linear hydroxyl polyurethane	1.33E – 01	kg	Primary data/Ecoinvent
High crystallization polyurethane	1.54E – 02	kg	Primary data/Ecoinvent
Acetone	7.56E – 01	kg	Primary data/Ecoinvent
Monopropylene glycol	9.25E – 04	kg	Primary data/Ecoinvent
Polybutylene adipate terephthalate (PBAT)	3.25E – 02	kg	Primary data/Ecoinvent
Dichloromethane (DCM)	8.79E – 02	kg	Primary data/Ecoinvent
Isophorone diisocyanate (IPDI)	6.88E – 02	kg	Primary data/Industry data 2.0
Poly (vinyl alcohol) (PVA)	5.00E – 03	kg	Primary data/Ecoinvent
Water	5.00E – 01	kg	Primary data/Ecoinvent
Anti – foam	2.50E – 03	kg	Primary data/Ecoinvent
Washing water	5.00E – 01	kg	Primary data/Ecoinvent
PUMC adhesive production			
Transport of materials	776.353	kg·km	Estimated/Ecoinvent
Energy (electricity)	0.2	kWh	Primary data/Ecoinvent
PUMC adhesive packaging			
Tinplate	6.89E – 02	kg	Primary data/ELCD
Polyethylene terephthalate (PET)	9.60E – 03	kg	Primary data/Ecoinvent
Production of tinplate packaging	6.89E – 02	kg	Estimated/Ecoinvent
Production of PET packaging	9.60E – 03	kg	Estimated/Ecoinvent
Packing transport	1.65	kg·km	Primary data/Ecoinvent
PUMC adhesive transport			
PUMC adhesive transport	97.068	kg·km	Estimated/Ecoinvent

Table 3 Impact assessment categories and normalization and weighting factors

Impact categories	Damage categories	Normalization factors	Weighting factors
Carcinogens (kg C_2H_3Cl eq)	Human health (DALY)	141	1
Non-carcinogens (kg C_2H_3Cl eq)			
Respiratory inorganics (kg $PM_{2.5}$ eq)			
Ionizing radiation (Bq C-14 eq)			
Ozone layer depletion (kg CFC-11 eq)			
Respiratory organics (kg C_2H_4 eq)			
Aquatic ecotoxicity (kg TEG water)	Ecosystem quality (PDF*m^2 year)	7.3E−5	1
Terrestrial ecotoxicity (kg TEG soil)			
Terrestrial acid/nutri (kg SO_2 eq)			
Land occupation (m^{2}org.arable)			
Aquatic acidification (kg SO_2 eq)	–	–	–
Aquatic eutrophication (kg PO_4 P-lim)	–	–	–
Global warming (kg CO_2 eq)	Climate change (kg CO_2 eq)	0.000101	1
Non-renewable energy (MJ primary)	Resources (MJ surplus)	0.00000658	1
Mineral extraction (MJ surplus)			

DALY disability adjusted life years (years of disabled living or years of life lost due to the impacts), *PDF* potentially disappeared fraction of species (species that disappear as result of the impacts), *MJ surplus* surplus energy (MJ) (extra energy that future generations must use to extract scarce resources)

3 Results and discussion

3.1 Traditional adhesive: PU adhesive

An overview of the relative contribution to the environmental impact categories of Impact 2002 + method for the PU adhesive system is shown in Fig. 3. The pattern of the burden distribution is similar in many of the impact categories. The PU adhesive production stage is the main contributor to the environmental impact in all impact categories, except in the Land occupation, and Mineral extraction, where the main contributor is the PU packaging production stage. The PU packaging production's main contribution is due mainly to tinplate consumption. Figure 4 shows the relative contribution to the environmental impact categories of the PU adhesive production stage, where it is possible to claim that acetone and polyol consumption are the major responsible for the environmental impact of the PU adhesive, except for the environmental impact categories Ionizing radiation, Ozone layer depletion, Terrestrial ecotoxicity, Land occupation, and Mineral extraction. The transport of materials and energy (electricity) requirements are the main contributors to these environmental impact categories. Based on the gathered data, the total GWP and non-renewable energy environmental impacts are estimated, respectively, at 2.59 kg CO_2 eq/kg PU adhesive, and 74.93 MJ primary/kg PU adhesive.

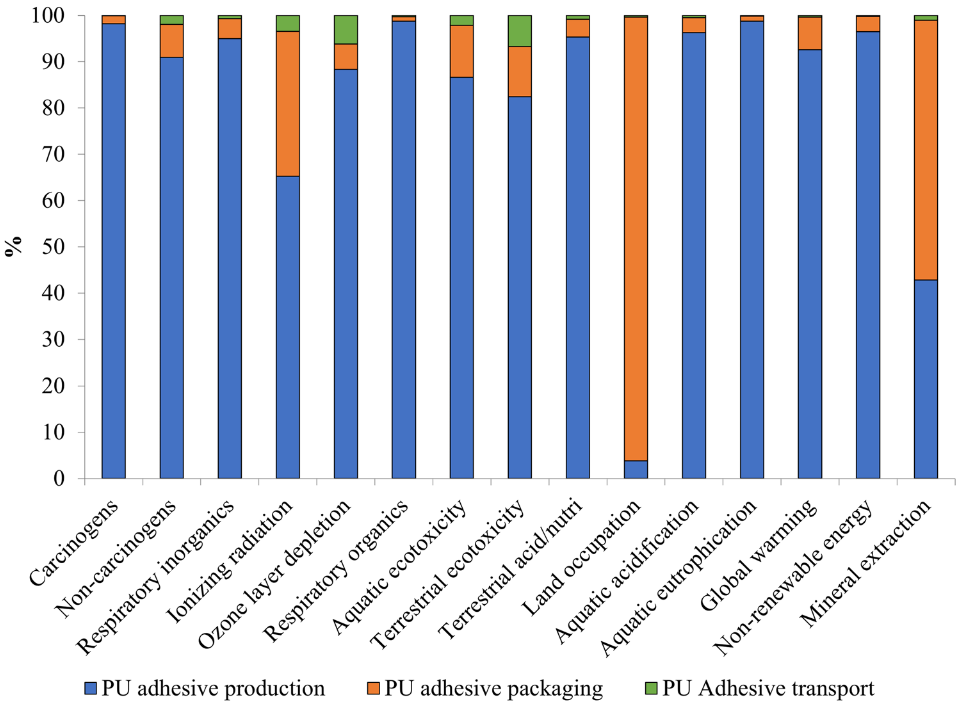
3.2 New developed adhesive: PUMC adhesive

Similarly, to what is observed for the PU adhesive system, the PUMC adhesive system shows a distribution pattern of the burdens alike in many of the impact categories. The PUMC adhesive production stage is the main contributor to the environmental impact in all impact categories, except in the Land occupation, where the PUMC packaging production stage is the main contributor. The PUMC packaging production main contribution is again due mainly to tinplate consumption. Figure 5 shows the relative contribution to the environmental impact categories of the PUMC adhesive production stage, where it is possible to claim that the acetone consumption and microcapsule production are the major responsible for the environmental impact of the PUMC adhesive. The polyol consumption shows some relevance for this system as well. The microcapsule production stage results (Fig. 6) indicate that the polybutylene adipate terephthalate (PBAT), dichloromethane (DCM), and isophorone diisocyanate (IPDI) consumption are the major responsible for the environmental impact of this stage. Based on the gathered data, the total GWP and non-renewable energy environmental impacts are estimated, respectively, at 3.09 kg CO_2 eq/kg PUMC adhesive and 83.02 MJ primary/kg PUMC adhesive.

3.3 Comparatives LCA between adhesives

Figure 7 shows the relative results of the comparison between the PU and PUMC adhesive systems, having as reference the adhesive technology with the biggest impact. In general, the PUMC adhesive has the biggest impact in all categories, except the Carcinogens environmental impact category. In this case, the PU adhesive system has a higher environmental impact due to a higher acetone consumption. Considering model, scenario, and parameter

Fig. 3 Relative contributions (in %) for all midpoint impact categories for the PU adhesive system



uncertainty (Bamber et al. 2020; Bjorklund 2002), it is considered a rule of thumb that differences up to 10% are irrelevant. Therefore, both systems are considered similar, regarding respiratory organics, land occupation, aquatic eutrophication, and non-renewable energy.

To better understand the magnitude of the category indicator results relative to the environmental impact of an European person, the previous results were normalized. The normalized results (Fig. 8) of the environmental impacts of the two adhesive systems reveal that the highest environmental impact corresponded to non-renewable energy and global warming, followed by respiratory inorganics and also carcinogens.

It is also important to highlight some available studies that have evaluated the environmental impact of footwear and, in specific, of adhesives (Albers et al. 2008; Cheah

Fig. 4 Relative contributions (in %) for all midpoint impact categories for the PU adhesive production stage

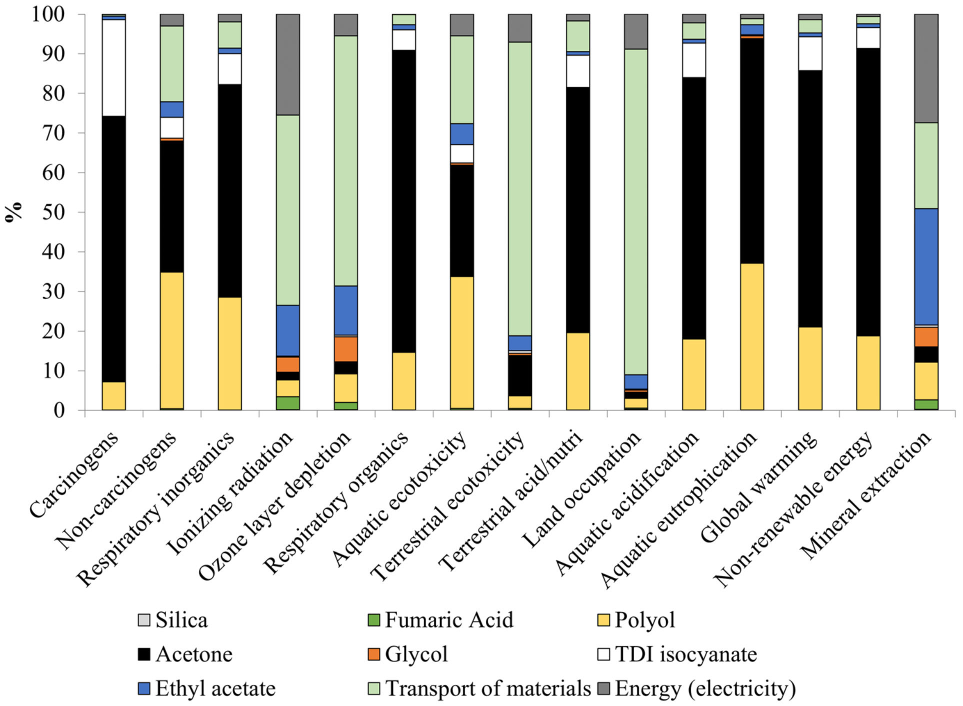
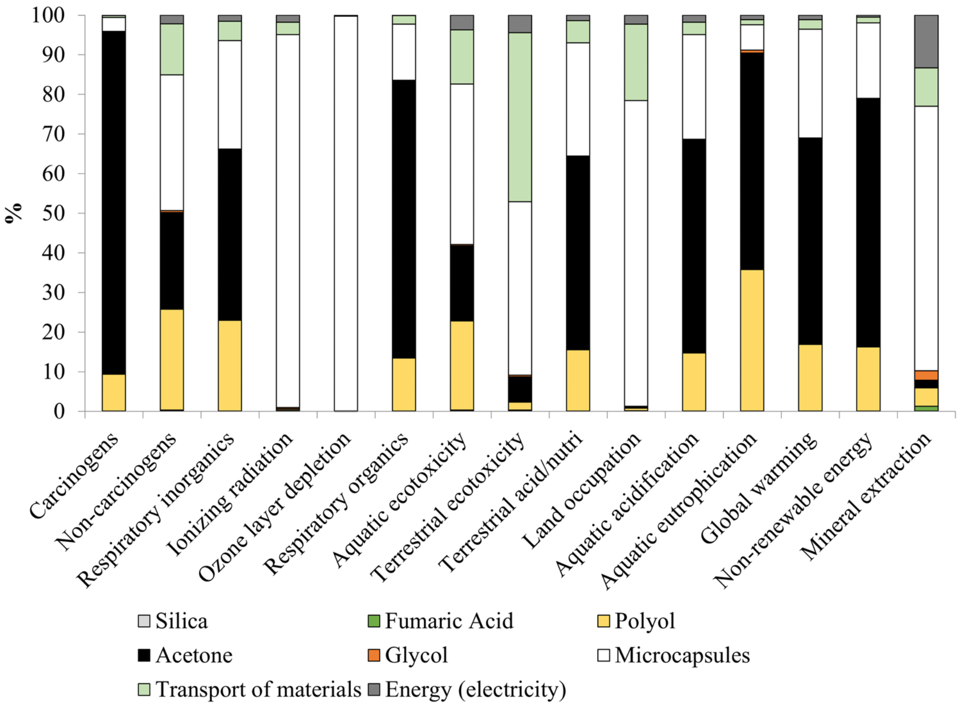


Fig. 5 Relative contributions (in %) for all midpoint impact categories for the PUMC adhesive production stage

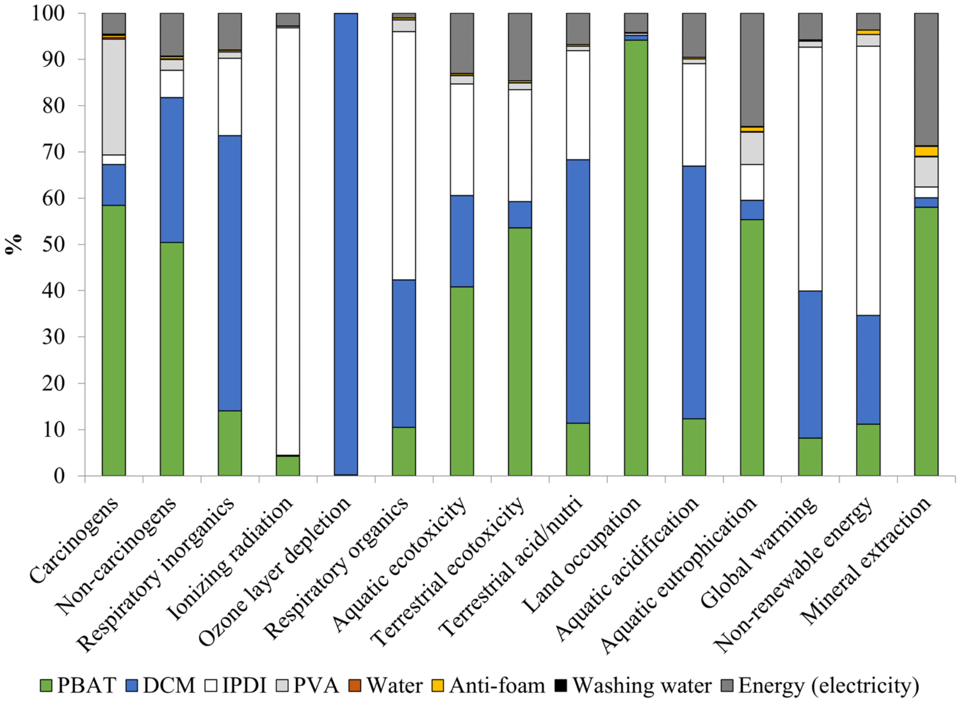


et al. 2013; Eisen et al. 2020; Maciel et al. 2017; Milà et al. 1998; Muñoz 2008; Packham 2009; Paiva et al. 2016b; Yang and Rosentrater 2019). However, for reasons of scope, those cannot be directly compared to the present study, as they do not consider the adhesive production or mention the contribution of the adhesive production system separately. For instance, Maciel et al. (2017) studied a SBA; however, their findings included also adhesive application. Therefore, the environmental impacts are higher comparatively with those reported in this study, as the adhesive application stage is energy intensive.

3.4 Sensitivity analysis

To better understand the environmental impact of the new adhesive (PUMC adhesive), a sensitivity analysis was

Fig. 6 Relative contributions (in %) for all midpoint impact categories for the PUMC adhesive microcapsule production stage



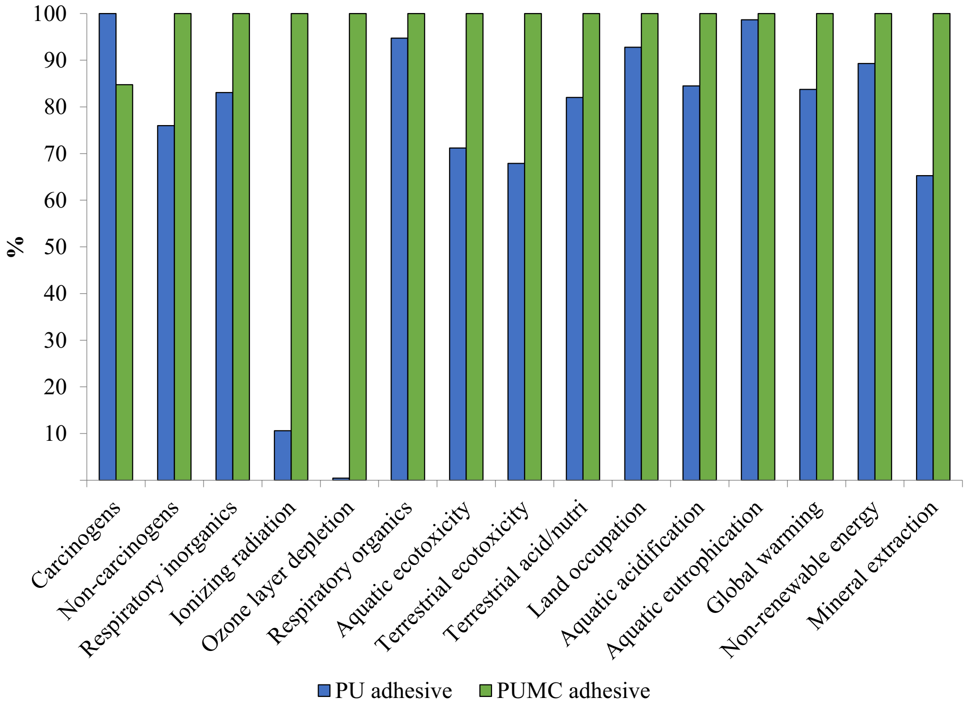


Fig. 7 Comparison of relative contributions (in %) for all midpoint impact categories of the PU and PUMC adhesive systems

conducted (using scenarios) to determine the response of the PUMC adhesive system to model, scenario, and parameter variability. Once the main hotspots of the production of the PUMC adhesive have been identified, alternative scenarios and a sensitivity analysis were developed to determine the degree of improvement achievable in the environmental profiles.

A possible aspect for future work is the possibility of enhancing the microcapsule production efficiency from 75 to 90%. Originally, the PUMC adhesive microcapsule production efficiency is at 75%, but from meetings with the technical staff of the company and the research team, the performance of this process is expected to reach 90%, due to the improvement of the microcapsule production

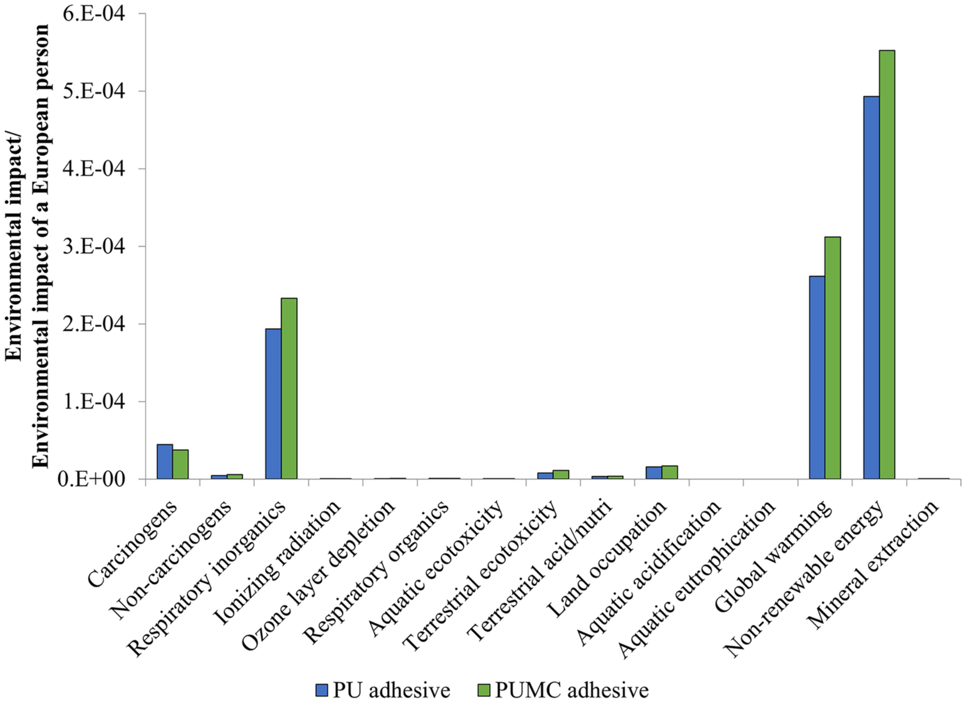


Fig. 8 Normalized results of the comparison of the PU and PUMC adhesive systems

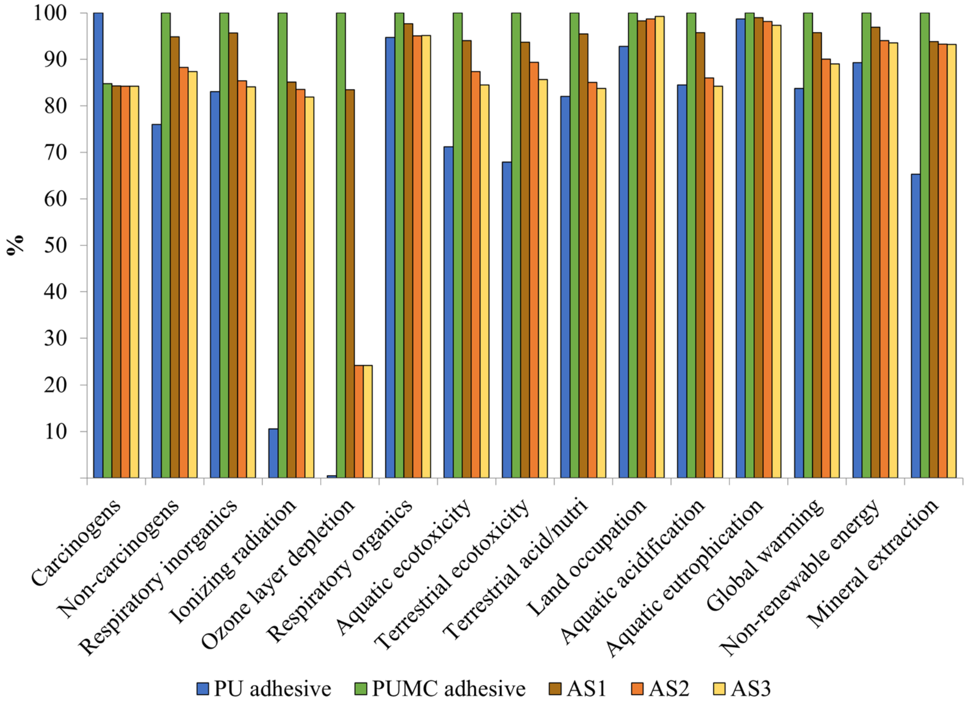


Fig. 9 Comparison of relative contributions (in %) for all midpoint impact categories of the PU and PUMC adhesive systems, and the three alternative scenarios of the PUMC adhesive systems

equipment efficiency (moving from a pilot scale to industrial equipment).

Also, another aspect for future improvement is the efficiency of recovery of the DCM in the production of the microcapsules, expected to increase from 65 to 90%. Originally, the PUMC adhesive microcapsule production DMC recuperation efficiency is at 65%, mostly as the DMC condensation is based on a small scale unit; however, from meetings with the technical staff of the company and the research team, using an industrial equipment the DCM recuperation efficiency is expected to reach 90%.

Yet another possibility for further performance improvement is that of using an alternative energy source for the PUMC adhesive system. The PUMC adhesive system assumed the Portuguese country's electricity mix as an energy source. However, based on the current focus on environmental and sustainability goals of the energy industry and the partner company strategy, it can be assumed that an alternative renewable energy source can be used. For the study, the renewable energy source was assumed as photovoltaic panels, which is a current trend in the industry and is being considered by the partner company.

Therefore, three cumulative alternative scenarios of PUMC adhesive systems were analyzed: alternative scenario 1 (AS1): PUMC adhesive microcapsule production efficiency of 90%; alternative scenario 2 (AS2): AS1 plus PUMC adhesive microcapsule production with a 90% DCM recuperation; and alternative scenario 3 (AS3): AS2 plus alternative PUMC adhesive energy source.

Figure 9 shows the relative results of the comparison between the PU and PUMC adhesive systems and the three alternative PUMC adhesive systems, having as reference the adhesive technology with the highest impact. In general, as expected, the three alternative PUMC adhesive systems have a lower environmental impact than PUMC adhesive, however always higher than PU adhesive except for the Carcinogens environmental impact category. However, the alternative PUMC adhesive scenarios results show in some environmental categories differences below 10%, therefore being considered similar to the PU adhesive system in these categories. The results also show that using an alternative renewable energy source (AS3) does not lead to as much decrease in the environmental impact as observed for the optimization of material consumption (AS1 and AS2).

To evaluate and compare the environmental impact of the five adhesive systems, a process of normalization, weighing, and combining the categories into a single score was carried out. Figure 10 shows the single score results of the PU adhesive, the PUMC adhesive, and the three alternative PUMC adhesive systems. From the single score results, it is possible to conclude that—globally—the PU adhesive is environmentally similar to the alternative PUMC adhesive systems (AS1, AS2, and AS3), since the results differ by less than 10%.

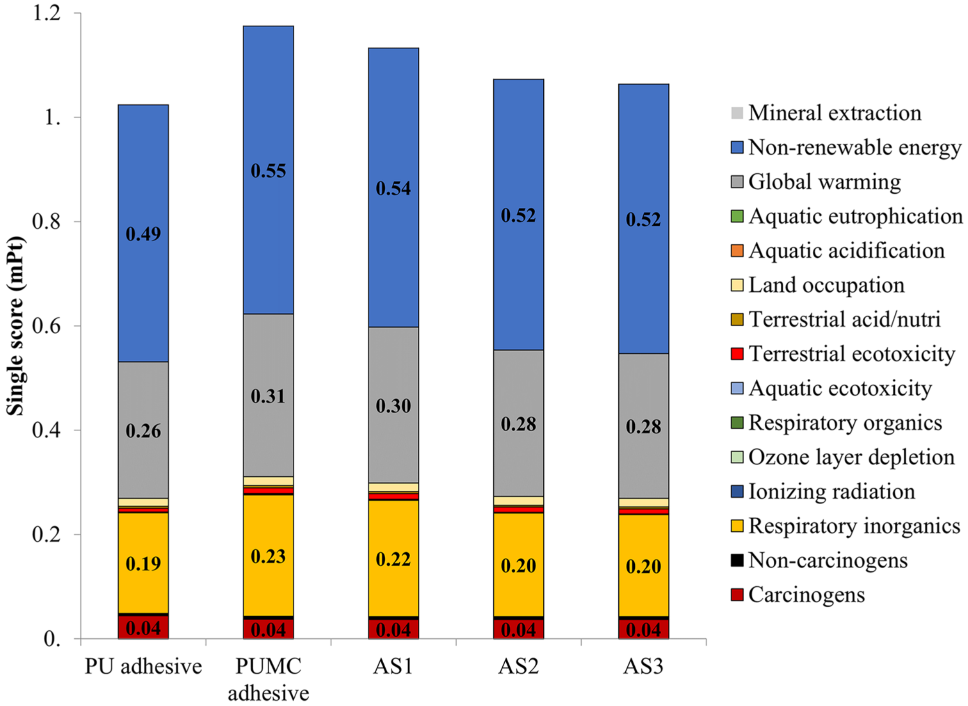


Fig. 10 Single score results of the comparison of the PU and PUMC adhesive systems and the three alternative scenarios of the PUMC adhesive systems (mPt: milli-point)

4 Conclusions

In this study, a LCA comparison was performed between a traditional adhesive used in the footwear industry (PU adhesive) with a novel microencapsulation approach which is safer and technically more favorable (PUMC adhesive). PUMC adhesive is a new adhesive prepared using a state-of-the-art technology (microencapsulation of isocyanates).

The findings indicate that the conventional PU adhesive's environmental footprint stems from its production phase, involving the consumption of acetone and polyol, as well as the transportation of materials and energy (specifically, electricity) requirements. For the PUMC adhesive, the production stage is also the main contributor to the environmental impact. However, in this case, the acetone consumption and microcapsule production are the major factors responsible for the environmental impact. Within the microcapsule production stage, the polybutylene adipate terephthalate (PBAT), dichloromethane (DCM), and isophorone diisocyanate (IPDI) consumption are the main responsible for the environmental impact. In general, PUMC adhesive has the biggest impact in all categories, except in the Carcinogens environmental impact category.

A sensitivity analysis was conducted using a scenarios study to determine the response of the PUMC adhesive system to model, scenario, and parameter variability. Three cumulative alternative scenarios of PUMC adhesive systems were assessed: AS1—PUMC adhesive microcapsule production efficiency of 90%; AS2—plus PUMC adhesive microcapsule production with a 90% DCM recuperation; and AS3—plus alternative PUMC adhesive energy source (photovoltaic panels). As expected, the three alternative PUMC adhesive systems have a lower environmental impact than the PUMC adhesive; however, their environmental impact remains higher than that of the traditional PU adhesive, except for the Carcinogens environmental impact category. The alternative PUMC adhesive scenarios results show differences equal to or less than 10% in some environmental categories, and therefore should be considered equivalent to the PU adhesive system in these categories.

Thus, future studies that aim to decrease the environmental impact of the PUMC adhesive should focus on the optimization of the microcapsule production stage, seeking to increase the production efficiency in order to decrease material consumption. The results show that using an alternative renewable energy source does not significantly improve the environmental impact (compared to a decrease in material consumption).

Despite the fact that the PUMC adhesive has a slightly higher environmental impact compared to the traditional PU adhesive, the increased safety of the novel approach gives its merit. The production and use of adhesives generate pollutants which can have an adverse health and environmental effect (Coureau et al. 2021; Metzger and Eissen 2004; Packham 2009; Staikos et al. 2006), and footwear workers, in particular, are routinely exposed to isocyanates that are regarded as one of the main causes of occupational asthma (Baur et al. 1994; Ameille et al. 2003; Lefkowitz et al. 2015; Gomez-Lopez et al. 2021; Karlsson et al. 2022). With the proposed microencapsulation approach for the isocyanate compounds, there are zero emissions of isocyanate within the footwear production factory, isolating the factory workers from those compounds.

Another aspect that should be assessed in the future is the adhesive waste resulting from the industry footwear production process. The authors believe that the use of the PUMC adhesive will lead to a decrease in leftover adhesive (the traditional adhesive has a short life span once blended for use, and some of it often ends up curing from being exposed to air and is then wasted). In the PUMC adhesive, the isocyanate compounds are microencapsulated and the curing process is controlled by the footwear worker, thus increasing the lifetime of the adhesive blend and consequently decreasing adhesive waste.

In conclusion, this study describes and analyses the development of a new technology for footwear adhesives based on microencapsulation of isocyanate and its environmental advantages and disadvantages concerning the traditional adhesive that uses non-enclosed isocyanate. Finally, the PUMC adhesive also shows potential for mitigating some environmental impacts.

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Data availability Data will be made available by the authors on reasonable request.

Declarations

Competing interests The authors declare no competing interests.

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