Modelling of Environmental Impacts of Printed Self-healing Products

Akos Cseke[a], Merryn Haines-Gadd[b], Paul Mativenga*[a], Fiona Charnley[b], Bradley Thomas[c], Justin Perry [c]

[a]*Department of Mechanical, Aerospace & Civil Engineering, School of Engineering, The University of Manchester, United Kingdom*
[b]*Exeter Centre for Circular Economy, The Business School, University of Exeter, United Kingdom*
[c]*Department of Applied Sciences, Northumbria University Newcastle*

* Corresponding author. Tel.: +00 44 0161 306 3821; fax: +00 44 0161 306 3821. *E-mail address* P.Mativenga@manchester.ac.uk

---

Abstract

Products utilizing self-healing materials have the potential to restore some of their function following damage, thereby extending the product lifespan and contributing to waste prevention and increased product safety. Despite the growing interest in these products, there a lack of comprehensive studies on the environmental implications of self-healing products and the parameters that influence impacts. The study presented in this paper combined life cycle assessment combined with a Taguchi experimental design and analysis of variance to investigate the effect of various parameters across the life stages of a self-healing composite product manufactured by 3D printing using poly-lactic acid (PLA) and self-healing polyurethane (PU). The results of this study suggests that impacts are primarily affected by avoided production due to the increased service of the product, followed by electricity requirements and material deposition rate (efficiency) of 3D printing. In the case of water consumption raw material manufacturing of PLA and PU are the highest and hence should be a target for research on reducing their water footprint. When comparing self-healing vs. regular products it is evident that most of the impacts are dominated by the electricity consumption of the manufacturing process. These results suggest that maximizing avoided production can play a major role in reducing impacts of 3D printed products. The results are important for maximizing the circularity of additive manufacturing products while minimizing their life cycle impact.

Keywords: Environmental impacts, life cycle assessment, self-healing, product, 3D printing

Highlights:

- Novel design of experiments and life cycle assessment of self-healing products
- Global warming potential, marine ecotoxicity, ozone depletion and water consumption
- Optimum avoided production required to reduce overall product life cycle impact
- Dominant parameters for significantly reducing the life cycle impact

1. Introduction

Self-healing materials and products are of growing interest due to their ability to heal following damage and thereby restore some of their original material properties and functionality. Based on the mechanism, self-healing is usually categorised as either intrinsic or extrinsic. In brief, intrinsic self-healing can be described as the material's ability to reform broken bonds caused by damage within its structure. Because the mechanism is not dependent on a secondary material it can be considered as an inherent property of the self-healing material (Aïssa et al., 2012; Bekas et al., 2016; Blaiszik et al., 2010; Diesendruck et al., 2015; Hager et al., 2010). In the case of extrinsic self-healing, the healing agent is a secondary material incorporated in the substrate, usually in the form of microcapsules or vascular system. The healing mechanism is activated by the damage thereby limiting further damage and restoring some of the material's properties (Cohades et al., 2018; Diesendruck et al., 2015; Mauldin and Kessler, 2010; Wang et al., 2013). Self-healing ability has been developed in materials such as concrete (Biesiekierski et al., 2012; Herbert and Li, 2013; Lv et al., 2016), glasses (Coillot et al., 2010; Zhang et al., 2018) and polymers (Kim et al., 2017; Mauldin and Kessler, 2010; Xia et al., 2017). Recently, intrinsically self-healing polyurethane (PU) in particular seems to be the focus of research resulting in over 200 publications in the topic during the past year alone (see Figure 1) (Web of Science, 2021). While the research output about self-healing materials in material science has been steadily increasing, there seems to be little information available about the environmental implications of self-healing products.

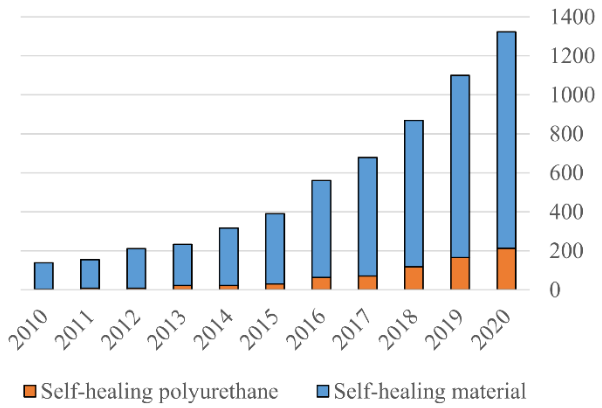


Figure 1 – Number of search results for "self-healing material" (blue) and “self-healing polyurethane” (orange) in topic (Searches title, abstract, author keywords, and Keywords Plus.), between 2010 and 2020 (Web of Science, 2021)

From the perspective of sustainability, the main advantage of self-healing lies in product service life extension, resulting in fewer products to fulfil the same function over a period of time, this in turn requires fewer products to be manufactured. In addition, self-healing products have great potential in contributing to waste prevention by providing the basis for long-term reliability (Bekas et al., 2016) and improving safety (Aïssa et al., 2012; Guadagno et al., 2014; Herbert and Li, 2013). Self-healing materials and products also have the potential in avoiding or reducing the costs of repair by reducing or completely avoiding the resource requirements and the environmental burden associated with the collection, transport, redistribution and repair or reprocessing of used products (see: Figure 2). An increase in the safety and reliability of a self-healing product or its parts can result in a higher value product and a higher level of compliance with standards and safety regulations, while increased safety factors of components through self-healing can aid in higher material utilisation and increased functionality. These indicators seem to suggest that self-healing products have the potential to aid in reducing environmental impacts through waste prevention and decreased production. From Figure 2, it can be postulated that self-healing should help maximise the product retained value compared to other circular economy options.

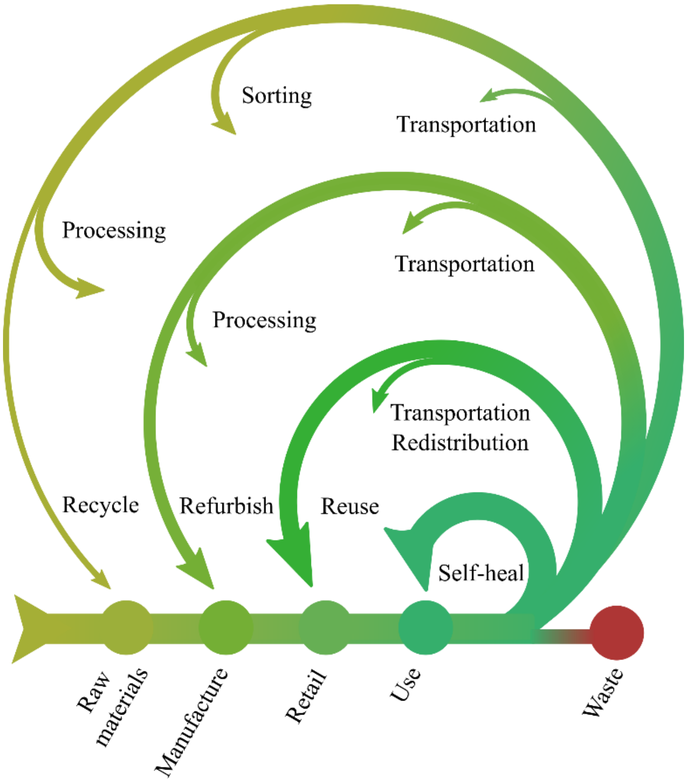


Figure 2 – Self-healing in the context of waste prevention and circular economy indicating main losses (not to scale)

Based on the recommendations of the European Council on waste hierarchy, prevention is the preferred scenario followed by reuse, recycling, energy recovery and finally disposal (European Council, 2008). While reusability depends on the type of product, the reduction in environmental impact is affected by the processes associated with reuse such as collection and preparation (Blanca-Alcubilla et al., 2020; Bovea et al., 2020; Farrant et al., 2010; Gasol et al., 2008; Unger and Landis, 2014; Zink et al., 2016). If the impacts associated with reuse outweigh the benefits of the avoided new production, replacement of the product may be more environmentally beneficial (Nußholz, 2017). While embodied impacts may become more significant with increasing energy efficiency of the product (Cooper and Gutowski, 2017), self-healing materials need to be applied with the aim to reduce the overall impacts through avoided production. This requires knowledge about self-healing materials that extend beyond material science into product design and sustainable and economic considerations.

While self-healing has the potential to reduce waste through avoided production, one of the main challenges of manufacturing remains the transition to low environmental impact on manufacturing. Additive manufacturing and fused deposition modelling (FDM) or 3D printing in particular, has been regarded as a key technology for reducing environmental impacts of manufacturing. The flexibility and customisation 3D printing can support sustainable design approaches such as reduced product weight, better utilisation of raw material, improved functionality as well as reduced impacts from transportation (DePalma et al., 2020; Faludi et al., 2019; Godina et al., 2020). In addition, 3D printing also has the potential to play a key role in a circular economy by facilitating reuse, remanufacture and refurbishment of products supported by “do-it-yourself” (DIY) and distributed manufacturing (Böckin and Tillman, 2019; Sauerwein et al., 2019; Turner et al., 2019). To date additive manufacturing and 3D printing seem to have higher impact compared to conventional manufacturing methods due to energy (electricity) requirements of 3D printing (Faludi et al., 2019; Khalid and Peng, 2021; Saade et al., 2020). There is need to understand if self-healing materials and improved manufacturing processes could turn 3D printing into an enabler and viable pathway to meeting zero carbon emission targets.

While self-healing materials used in 3D printing have the potential to further its cause in low environmental impact manufacturing, detailed assessment will be necessary to determine the technology’s complex effect on the overall life cycle of the product, from design and resource requirements, through to manufacturing including end of life. For this reason, the study presented in this paper was aimed to investigate the effect of self-healing and 3D printing process parameters and end of life (EoL) options to identify the most critical parameters that affect the overall environmental impact of a self-healing product.

2. Materials and methods

Taguchi or robust design method was used in order to estimate the effects of factors (in this case the life cycle parameters) on the response (environmental impacts). Taguchi experimental design is based on fractional factorial experiments, and compared to full factorial design allowed the number of calculations/experiments to be minimised, while at the same time determining the optimal parameters leading to improved environmental performance. By reducing the number of necessary calculations, research time and resources could be allocated to extend the number of variables and their levels to capture various aspects of the product's life cycle, such as transportation and recycling, and to increase the quality (resolution) of the results.

2.1. Life cycle goal and scope

The aim of this life cycle assessment study was to investigate the effect of embedded self-healing material and 3D printing parameters on the overall impact of a self-healing product. Parameters and levels were specifically selected to enable the data to be further analysed through Taguchi method.

A cradle to grave approach was chosen in order to capture the full life cycle impacts. Because variables used in the study affect both material and energy requirements of the product, a self-healing product manufactured from the combination of PLA and self-healing PU by 3D printing was used. Variables affected by product size, such as printing time were chosen to be within reasonable limits in both size and complexity for 3D printing manufacturing technology. Further consideration was to facilitate an easy interpretation of the results, therefore the functional unit was chosen to be one kg of self-healing composite product printed from a two filament 3D printer.

2.2. Life cycle inventory

The model of the life cycle of the self-healing product used in the study is shown in Figure 3. The two-component product is manufactured by 3D printing using both conventional PLA and self-healing material as a filament. These were selected for their suitability in 3D printing for example by fused deposition modelling using a multiple nozzle machine. Data available in Ecoinvent 3 was used to account for the impacts for both PLA and PU raw material manufacturing. While PLA was assumed to come in the form of granulates, self-healing PU was approximated using a modified version of the dataset for flexible PU foam manufacturing which was the closest material available in SimaPro's databases. The dataset was modified based on the information available in literature about the materials used for specific self-healing PU synthesis (see: Table 1). It is important to note that the exact composition of self-healing PUs are highly dependent on the requirements of the final product and the specific application. Additionally one or more of the key ingredients or their ratios are usually not disclosed for commercial reasons. Generally, the most commonly used building blocks for self-healing PUs include three main components; a diol terminated prepolymer (long diol), an isocyanate and a chain extender (Willocq et al., 2020). Based on the information in literature and consultation with polymer experts involved in the study, a closest approximation was compiled utilising the available material production data in SimaPro's databases, which is summarised in Table 1.

Table 1 – Main components of self-healing PU and the closest equivalents used in the life cycle assessment [a](Ritzen et al., 2021), [b](Imato et al., 2021), [c](Lee et al., 2019)

Component type	Example of material used in literature	Closest equivalent available in SimaPro databases	Ratio used for calculation
Diol terminated prepolymer (long diol)	CroHeal™2000[a] poly(tetra- methylene glycol) (PTMG)[b] poly(tetramethylene ether glycol) (PTMEG)[c]	Polybutadiene	60%
Isocyanate	4,4′-diphe- nylmethane diisocyanate (MDI)[a,b] isophorone diisocyanate (IPDI)[c]	Methylenediphenyl diisocyanate (MDI)	30%
Chain extender	2-Ethyl-1,3-Hexanediol (EHD)[a] "Py-diol"[b] Ethylenediamine/Chitosan[c]	Butane-1,4-diol	10%

In order to account for the uncertainty in the impacts brought about by the unknown components and variations of their ratios in the self-healing PU, it was assumed that the material requirements for 1 kg of self-healing PU filament follows a triangular distribution between 0.7 and 1.3 kg of self-healing PU.

Both PLA and PU manufacturing were modelled using market processes in Ecoinvent 3, therefore the emissions also included infrastructure such as buildings and machinery, as well as transportation.

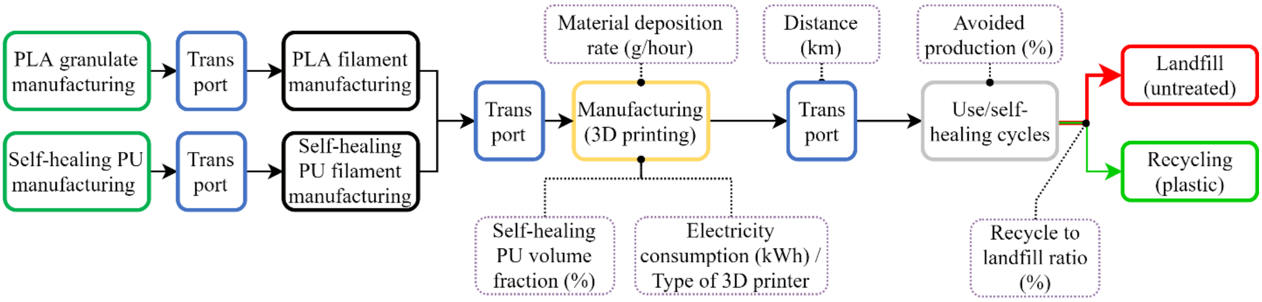


Figure 3 - Overview of the model used in the LCA study indicating variables (dashed) used in the study

Following raw material manufacturing the PLA and PU were transported by lorry (200±50 km) to the filament manufacturing facility where the pellets were extruded, cooled and spooled on drums. The facilities such as buildings, machinery and infrastructure for manufacturing were excluded from the study as they were deemed negligible compared to the impact coming from the energy requirement of other processes, particularly 3D printing. In order to estimate the energy requirements of the filament manufacturing, data from the manufacturer of several filament making systems were collected and an average electricity requirement was determined (see Table 2). Where average power consumption was not available, half of the maximum nominal power consumption was used (see: Table 2: [c]calculated).

Table 2 – Electricity requirement of filament manufacturing used in LCA study, [a,b]manufacturer's specifications (Felfil Evo specs., Filabot specs., Filabot spooler specs.), [c]calculated

Component and type	Extrusion rate (g/h)	Max power consumption (average) (W)	Power requirement per kg filament (Wh/kg)	Average power requirement per kg filament (Wh/kg)
Felfil Evo (complete system)	1.15 m/minute[a] (PLA: 816.5 g/h)[c]	240 (110)[a]	135[c]	321[c]
Anonymous manufacturer (compact)	500	1300 (300-400)	700[c]	

Filabot Ex2 (Incl. spooler and cooler)	910+[b]	470[b] (235)[c]	258[c]
Filabot Ex6 (Incl. spooler and cooler)	4500+[b]	1720[b] (860)[c]	191[c]

Following manufacturing the filaments were transported by lorry to the 3D printing facility (100±20 km) where the fabrication of the self-healing product took place. In order to estimate the range of material deposition rate of the 3D printer, a 3D model of a one cm^3 (unit) cube was loaded to two 3D printing software (Slic3r V1.3.0 and Stacker Run V2.1.2) to calculate an estimate of the required printing time. Using the printing time and the amount of material required for a one unit cube, the material deposition rate was chosen between 6 and 18 g/hour (see Table 3).

Table 3 - Life cycle inventory data used in the LCA, [a]Ecoinvent 3 database, [c]calculated, *value used for average self-healing product

Variable			**Level**			**Comments**
	1	2	**3***	4	5	
Self-healing material volume fraction (SR) (%)	20%	40%	**60%**	80%	100%	Related to healing efficiency of the self-healing material
3D printer electricity consumption (3DE) (Wh)	100	350	**600**	850	1100	Basic Prusa consumes about 120 Wh, Dimension SST up to and over 1 kW.
Material deposition rate (MD) (g/hour)	6	9	**12**	15	18	Affected by various 3D printing parameters (see: Figure 4)
Avoided production (AP) (%)	19%	38%	**57%**	76%	95%	As a result of product life extension.
Transportation of self-healing product (TR) (km)	100	200	**300**	400	500	From 3D printing facility to retail by lorry
Recycling ratio (RE) (%)	10%	30%	**50%**	70%	90%	Ratio of recycle and landfill. Affected by the recycle ability of materials

Constant	**Value**	**Comments**
Filament maker electricity consumption (Wh)	234	Average value based on manufacturer's specifications
Transportation of raw materials (km)	200	From raw materials to filament manufacturing facility by lorry[c]
Transportation of filaments (km)	100	From filament manufacturing to 3D printing facility by lorry[c]
Self-healing PU raw material ratio (by weight)	Polybutadiene: 60% Methylenediphenyl diisocyanate (MDI): 30% Butane-1,4-diol: 10%	See: Table 1

The overall energy requirement of 3D printing is affected by numerous factors such as printing speed (mm/s), model orientation, infill density, printing temperature, just to name a few (Khalid and Peng, 2021). The relationship of most important 3D printing variables and their effect on electricity consumption is summarised in Figure 4.

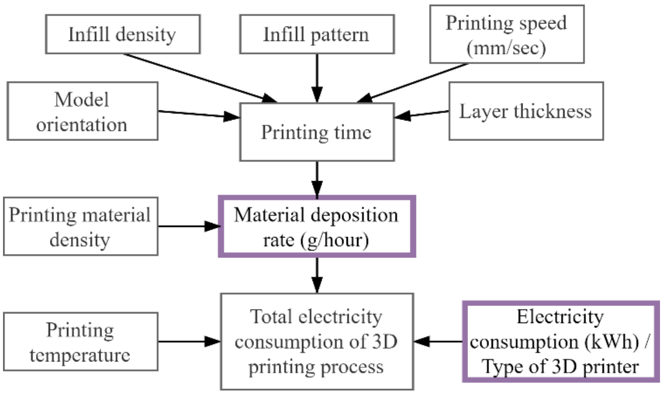


Figure 4 - Parameters and their effect on total electricity consumption a 3D printing process: Purple - used as a parameter (see: Table 3 for details)

Although the material deposition rate ultimately affects the energy consumption of the 3D printing process, overall electricity consumption was still chosen as a variable as it also reflects the type of 3D printer being used and therefore was essential to include as a separate variable in the study. Electricity consumption of small commercial 3D printers such as the Prusa typically start at around 120 Wh (Prusa FAQ) and can reach as high as 1.1 – 1.5 kWh for printers with heated building chambers such as the Dimension SST (Balogun et al., 2014). For this reason the electricity consumption of the 3D printer was chosen to be a variable between 100 and 1100 Wh. Following 3D printing the self-healing product were transported to retail by lorry, the distance was varied between 100 and 500 km in order to investigate its effect on the overall impacts.

In order to distinguish self-healing from other restoration techniques such as repair, self-healing in this study refers to "*a product's capability to restore/maintain its function and user acceptance within its operating environment*" (Cseke et al., 2020). Thus self-healing ability is linked to the product's function rather than a restoration of a material properties, such as tensile strength (Aïssa et al., 2012; Bekas et al., 2016; Blaiszik et al., 2010; Diesendruck et al., 2015; Guadagno et al., 2014; Hager et al., 2010; Herbert and Li, 2013), which do not on their own describe the self-healing capability and life extension of a product. Depending on the self-healing ability of the product, if the product's service life is extended by a factor N during the product's life time, the manufacture of a total of N-1 products can be avoided. For example, if a self-healing product lasts five times (N=5) as long as its conventional counterpart, during its life time the manufacturing of four (5-1=4) conventional products can be avoided. Expressing the avoided production as a percentage of the total therefore equates to $\frac{5-1}{5} \cdot 100\% = 80\%$ avoided production. Similarly, levels of 19, 38, 57, 76 and 95% avoided production used in the study therefore corresponds respectively to a life extension of 1.24, 1.61, 2.33, 4.17 and 20 times the original lifespan of the product.

Following the product's disposal some of the material is recycled while other materials go through a waste scenario for household waste in England (available in SimaPro 9.1.0.8, not reviewed by Ecoinvent), including municipal waste and waste separation in advance. The waste stream going into recycling appears as avoided production of PLA and self-healing PU, therefore primary production have both the burdens and benefits of recycling. Depending on the type of self-healing material used in a product, the embedded impacts may affect its end-of life and recycle ability. For this reason the ratio of the waste stream going into recycling was used as a variable to express the varying degree at which self-healing materials are likely to be recycled.

Altogether six input variables at five levels were selected (see: Table 3). If this was a full factorial design then a total of $5^6 = 15{,}625$ combinations and calculations would have been required. The Taguchi fractional experimental design corresponding to six variables at five levels (L25) enables the same investigation with only 25 experimental runs. The aim of the Taguchi approach was to use the signal to noise ratio to determine which of the six variables have the most effect on the life cycle impacts of the self-healing material (for details see: Appendix A). Results for the 25 observations were calculated using SimaPro version 9.1.0.8 and impact categories of global warming potential (GWP) (kg CO_2 eq), Marine ecotoxicity (ME) (kg 1,4-DCB), Stratospheric ozone depletion (SOD) (kg CFC11 eq) and Water consumption (WC) (m^3) were examined. These were selected based on their relevance to a low carbon economy and the environmental damage pathways linked to polymer materials.

In Taguchi analysis, the output or responses are classified as nominal-the-best, smaller-the-best or larger-the-best. Since the aim was to minimise the environmental impact, smaller-the-best was selected. The L25 Taguchi array and the corresponding impacts from LCA for each run from SimaPro modelling were entered in Minitab statistical software. Through Analysis of Variance (ANOVA), corresponding levels for each input variable required to minimise the environmental footprint were determined.

Initially, a comparative study was conducted to determine how impacts are shared across the product's life cycle stages and to point out the main differences between a non-self-healing (100% PLA) and an "average" self-healing product using level 3 variables (see: Table 3). Life cycle stages of raw material manufacturing (PLA and PU), filament manufacturing, 3D printing, transportation (from all stages) and waste were used. This was then followed by a study to identify of dominant parameters.

3. Results and Discussions

The results for the comparative study of regular and self-healing products are presented in Table 4, for one functional unit in each impact category contributing to the main processes during the product's life cycle. The numbers in brackets for each total show the standard deviations due to self-healing PU raw materials. The largest life cycle impact stage is shown in bold. Since regular product does not contain self-healing PU and uncertainties arising from transportation were negligible only the uncertainty of additional self-healing PU impact are indicated. The category transportation includes all three stages from raw materials to retail while waste includes the impacts arising from the non-recycled content. The negative impact of recycled materials, i.e. the benefits of recycling that come from the avoided raw material manufacturing were already included in other life cycle stages.

Table 4 - Characterisation of impact categories for one kg of regular (SR=0 %, 3DE=600 Wh, MD=12 %, AP=0 %, TR=300 km, RE=50 %) and self-healing product (SR=60 %, 3DE=600 Wh, MD=12 %, AP=57 %, TR=300 km, RE=50 %) (see: Table 3) using calculation in SimaPro, SD due to self-healing PU shown in brackets

Impact category	**Material type**	**Total**	**Raw materials manuf.**	**Filament manuf.**	**3D printing**	**Transport**	**Waste**
Global warming (kg CO_2 eq)	Regular	30.438	1.657	0.105	**28.230**	0.208	0.238
	Self-healing	13.246 (0.124)	0.784	0.045	**12.139**	0.176	0.103
Stratospheric ozone depletion (kg CFC11 eq)	Regular	1.543E-05	4.744E-06	5.763E-08	**1.046E-05**	1.410E-07	3.002E-08
	Self-healing	5.773E-06 (7.589E-08)	1.121E-06	2.478E-08	**4.498E-06**	1.163E-07	1.291E-08
Marine ecotoxicity (kg 1,4-DCB)	Regular	2.347	0.072	0.011	**2.166**	0.007	0.090
	Self-healing	1.001 (0.002)	0.020	0.005	**0.931**	0.006	0.039
Water consumption (m^3)	Regular	0.241	**0.1342**	0.0017	0.1042	0.0006	0.0002
	Self-healing	0.074 (0.0012)	0.0279	0.0007	**0.0448**	0.0005	0.0001

Based on the results for regular 3D printed material it is evident that majority of the impacts are dominated by 3D printing arising from the electricity consumption of the process, which seem to agree with the findings of other authors (Faludi et al., 2019; Khalid and Peng, 2021; Saade et al., 2020). The second most influential product stage is raw material manufacturing, which in the case of water consumption carries most of the impacts (0.134 m^3) followed by 3D printing (0.104 m^3). The higher proportion of water consumption arises from the current technological requirements of PLA and PU chemical processing. Together with 3D printing, raw material manufacturing constitute over 95% of the total impacts in all categories. Negligible impact arises from transportation between processing stages and retail. These results seem to suggest that manufacturing processes for both raw materials and products (3D printing), cleaner energy sources and sustainable design strategies for material saving can play a crucial role in reducing the impacts of 3D printed products.

End of life stage (waste) seem to be most influential in marine ecotoxicity resulting in higher impact than raw material manufacturing (0.072 vs 0.09 kg 1,4-DCB). This points to the significance of the impact of plastic waste in marine ecology. Therefore plastic waste prevention methods, such as product life extension through self-healing can play significant role in reducing marine ecotoxicity.

Transportation and filament manufacturing stages account for less than 2% of the total emissions in all impact categories suggesting that attempts to reduce the impacts of these categories should be lowest priority in the strategy for reducing the overall impacts of 3D printed products. The data presented in Table 2 is further illustrated in Figure 5 as the percentage of contribution of the main process stages to the total maximum emission in each impact category.

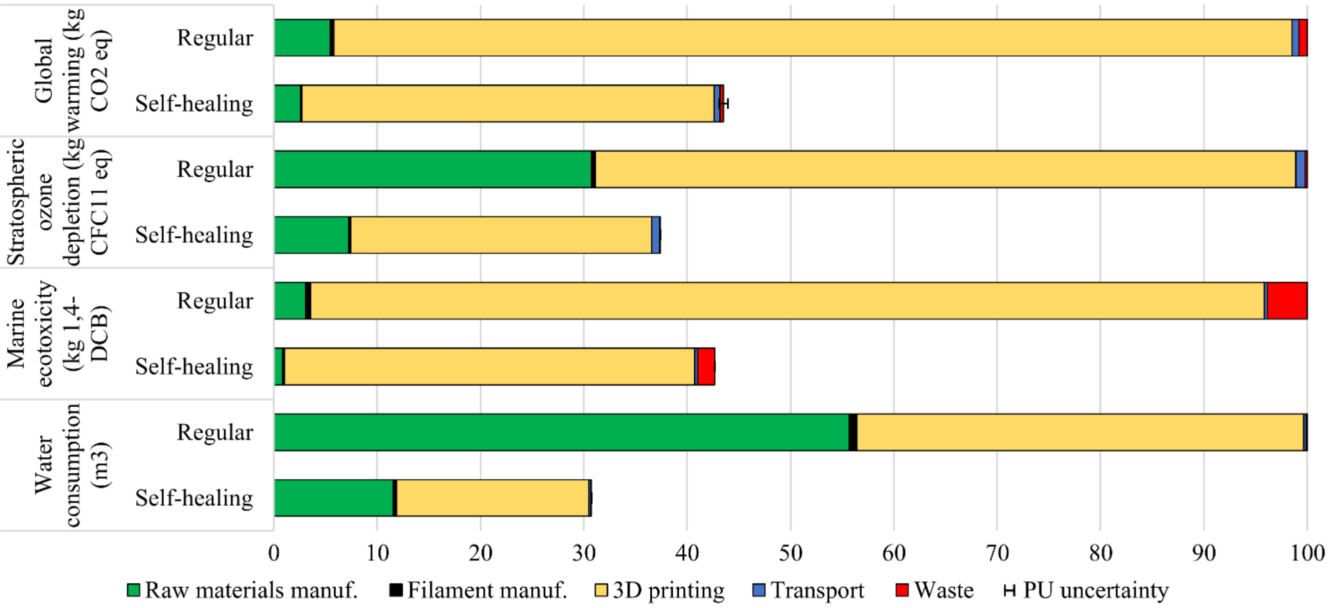
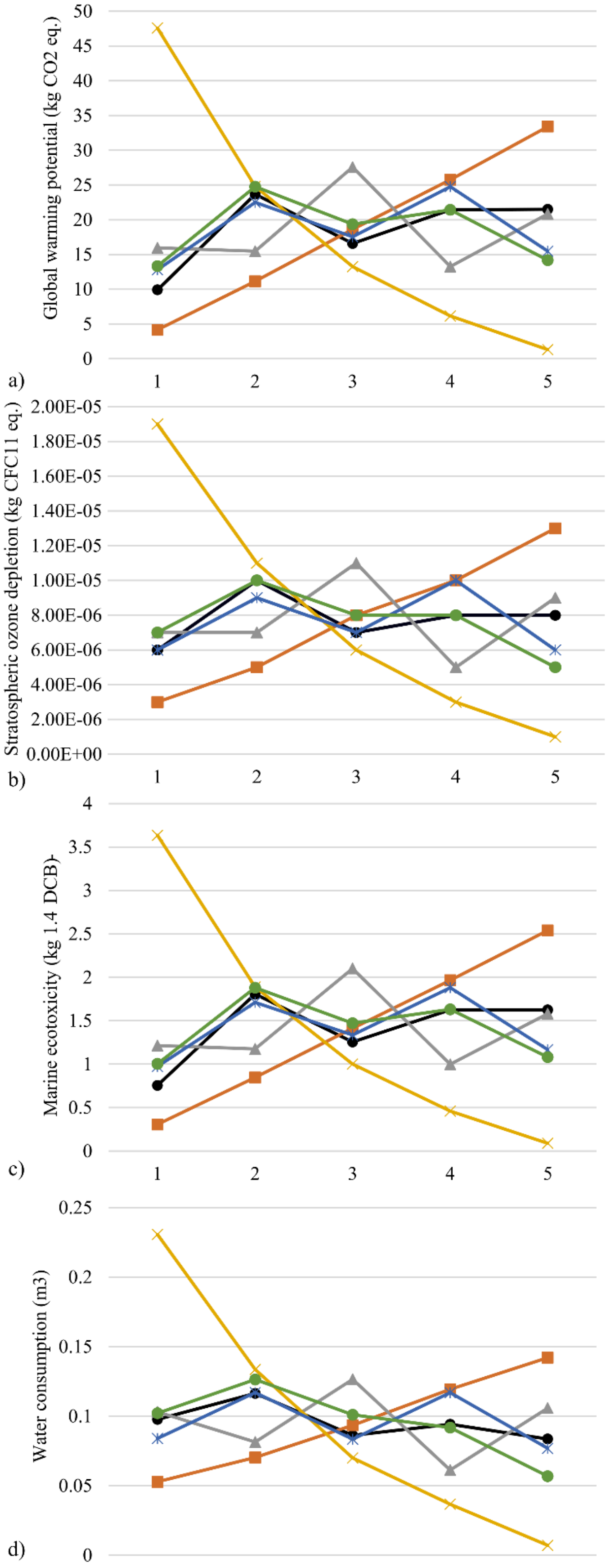


Figure 5 - Process contribution to impact categories relative to their maximum for one kg of regular (SR=0 %, 3DE=600 Wh, MD=12 %, AP=0 %, TR=300 km, RE=50 %) and self-healing product (SR=60 %, 3DE=600 Wh, MD=12 %, AP=57 %, TR=300 km, RE=50 %) (see: Table 3) using SimaPro calculation. Error bar: uncertainty from self-healing PU manufacturing

When comparing the regular product to self-healing, the overall emissions of the self-healing product are lower in all categories, indicating that the reduction in the impacts brought about by the avoided production outweigh the embedded impacts of the self-healing technology. As expected, from all of the categories transportation is the least affected. This is because avoided production only affects the transportation processes related to manufacturing, due to less raw materials needed to be transported, while the weight and transport distance of the finished self-healing products are the same (functional unit: one kg) in both scenarios.

The largest impact of avoided production can be observed in the case of water consumption where in the case of the self-healing product it is reduced by 68%. Similar results can be observed across the product stages with 79% less water consumption during raw material manufacturing, 57% for 3D printing and 58% in the case of filament manufacturing. The smallest reduction in overall emission between the two products can be observed in the case of global warming where an overall reduction by 56% is achieved with only 53% decrease in raw material manufacturing. In the case of stratospheric ozone depletion emissions are reduced by 63%, while in the case of marine ecotoxicity, the impact is reduced by 57%. These results correspond to the 57% avoided production while deviations can be attributed to the impacts arising from infrastructure, such as manufacturing facilities and transportation. The sensitivity of most of these parameters on the resulting impacts is further explored using Taguchi method.

Figure 6 shows the results of the Taguchi mean plots for each impact category as derived from the L25 design of experiments. The vertical axis indicate emission while the horizontal axis the parameters levels used for the study as shown in Table 3 as well as in the legend at the bottom of Figure 6 graph d). For each impact category the difference between lowest and highest values corresponding to each variable was calculated (Δ) and used for ranking (r) of the variable's effect (sensitivity) on each impact category. The results are summarised in Table 5.



Level	1	2	3	4	5
SR	20%	40%	60%	80%	100%
3DE	100	350	600	850	1100
MD	6	9	12	15	18
AP	19%	38%	57%	76%	95%
TR	100	200	300	400	500
RE	10%	30%	50%	70%	90%

Figure 6 - The effect of LCI variables on impact categories. Figures a) – d): SR: Self-healing volume fraction (%), 3DE: Electricity requirement of 3D printing (Wh), MD: Material deposition rate (g/hour), AP: Avoided production (%), TR: Transportation by lorry (km), RE: Recycling ratio (%)

Table 5 - overall change caused by variable: Delta with rank (r); GWP: global warming potential (GWP) (kg CO2 eq), ME: Marine ecotoxicity (ME) (kg 1,4-DCB), SOD: Stratospheric ozone depletion (SOD) (kg CFC11 eq), WC: Water consumption (WC) (m3)

Impact category		**Self-healing volume fraction (SR) (%)**	**Electricity requirement of 3D printing (3DE) (Wh)**	**Material deposition rate (MD) (g/hour)**	**Avoided production (AP) (%)**	**Transportation by lorry (TR) (km)**	**Recycling ratio (RE) (%)**
Global warming (kg CO_2 eq)	Max-min (Δ)	13.759	29.233	14.321	46.256	11.951	11.422
	Rank (r)	4	2	3	1	5	6
Stratospheric ozone depletion (kg CFC11 eq)	Max-min (Δ)	4.E-06	1.E-05	6.E-06	2.E-05	4.E-06	5.E-06
	Rank (r)	6	2	3	1	5	4
Marine ecotoxicity (kg 1,4-DCB)	Max-min (Δ)	1.05	2.233	1.102	3.546	0.907	0.876
	Rank (r)	4	2	3	1	5	6
Water consumption (m^3)	Max-min (Δ)	0.033	0.089	0.065	0.224	0.04	0.07
	Rank (r)	6	2	4	1	5	3

From the results of the Taguchi analysis it is evident that the impacts in each category are affected mostly by avoided production (AP) which ranks number one in all impact categories (Figure 6 - c)). This is supported by the dominant share of raw material to manufacturing constituting over 95% of the overall impacts of the 3D printed products. The second most influential variable is the electricity consumption of the 3D printing (3DE) which is placed second in each impact category. When comparing the contribution from avoided production (AP) with energy requirements for 3D printer the difference in their impact are between 37 to 60% across all categories which seem to agree with the findings of the comparative study (see: Figure 5) where Electricity consumption of 3D printing is the highest source of emission in most impact categories. Electricity consumption is followed by material deposition rate (MD) which ranked third in all categories except water consumption where the effect of recycling seems to be 7% higher (0.07 vs. 0.065 m^3), indicating material deposition rate’s direct relation to 3D printing time and the electricity consumption (see: Figure 4). For variables SR, TR and RE the effect size was less than 30% compared to AP for GWP, with similar results in other impact categories. It can also be observed that for these variables there is a considerable fluctuation in impact values across levels 1-5 and no confident trend can be drawn observable in Figures 6 a) – d).

Based on the results of the sensitivity analysis the lowest impact for all categories is to be expected using the lowest (20%) self-healing PU fraction (SR), 3D printer with the lowest energy consumption (100 Wh) and highest depositions rate 18 (g/hour), highest avoided production 95%, shortest transportation (100 km) and 90% recycling ratio. The comparison between the average self-healing product (level 3 variables) and best case scenario based on the Taguchi analysis is shown in Table 6.

Table 6 - Comparison between average self-healing product (level 3 variables) and best case scenario (based on Taguchi analysis)

Impact category	**Material type**	**Total**	**Raw materials manuf.**	**Filament manuf.**	**3D printing**	**Transport**	**Waste**
Global warming (kg CO_2 eq)	Optimum	0.549	0.017	0.005	0.470	0.054	0.002
	Average	13.246 (0.124)	0.784	0.045	12.139	0.176	0.103
	Reduction	**96%**					
Stratospheric ozone depletion (kg CFC11 eq)	Optimum	2.534E-07	4.031E-08	2.882E-09	1.744E-07	3.552E-08	3.002E-10
	Average	5.773E-06 (7.589E-08)	1.121E-06	2.478E-08	4.498E-06	1.163E-07	1.291E-08
	Reduction	96%					
Marine ecotoxicity (kg 1,4-DCB)	Optimum	0.0402	0.0006	0.0006	0.0361	0.0020	0.0009
	Average	1.001 (0.002)	0.02	0.005	0.931	0.006	0.039
	Reduction	96%					
Water consumption (m^3)	Optimum	3.093E-03	1.111E-03	8.648E-05	1.737E-03	1.560E-04	2.092E-06
	Average	0.074 (0.0012)	0.0279	0.0007	0.0448	0.0005	0.0001
	Reduction	96%					

The results presented in Table 6 the study shows that by using combined Taguchi experimental design and seeking for an optimum solution through analysis of variance a drastic, 96% decrease in all impact categories was realised in the life cycle assessment. This high reduction is also enabled by the 95% avoided production.

4. Limitations and boundaries of the study

While the results indicate that avoided production is critical for making self-healing products feasible, the tipping point can change from product to product. There is a range of product function restoration for which self-healing can be applied for. For this reason product specific assessment may be required as early as conceptual product design, including an understanding of product failure and the mechanism driving the failure of critical life limiting components.

While self-healing in the case of this study was assumed to be fully autonomous, energy requirements and potential emission (leaching, reaction with environment etc.) of healing mechanism may need to be taken into consideration. Currently very little data is available for the energy requirements of different healing mechanisms, therefore developing such datasets would provide vital input for the life cycle assessment for self-healing materials. However, a fully autonomous self-healing products appears to be the ideal in terms of functionally and potential for reduced impact.

The impacts presented in this paper need to be updated to more accurately represent the impacts of specific self-healing PUs which may use different material ratios and various other additives depending on the specific application. The development of data sets for life cycle assessment of new self-healing materials is an area that needs addressing and the cooperation of commercial materials developers.

The model used is for a passive product, i.e. without energy requirement and emission during use phase, the case for active products needs to be investigated.

5. Conclusions

Self-healing products represent a new innovation in the circular economy and in waste reduction. They enable increasing of service lifetime and product safety. In the study presented in this paper a multi parameter analysis was conducted using SimaPro 9.1.0.8 and Taguchi analysis in order to determine the crucial parameters that affect the impact of self-healing products manufactured by 3D printing. The following conclusions were made.

- In the study, global warming potential, marine ecotoxicity, stratospheric ozone depletion and water consumption were considered, and for each of these categories avoided production had the greatest effect followed by the electricity consumption of 3D printing. Material deposition rate of the 3D printer had lower affect in all categories, while self-healing material volume fraction, transportation and recycling had the lowest affect in most categories.
- Due to the overall high impact of electricity associated with manufacturing, cleaner electricity sources can have major role in reducing impacts of 3D printing.
- Self-healing materials can play a major and important role in reducing impacts of 3D printed products due to avoided production. The focus for researchers should be on maximising the life extension enabled by autonomous self-healing mechanisms. This will enable more effective use of embedded energy in materials and realisation of avoided impacts.
- Product specific assessments are required as early as the conceptual product design stage in order to assess and consider alternative designs, materials and manufacture to promote reduced life cycle impacts.
- The use of renewable and near zero carbon footprint energy sources for 3D printing of self-healing materials will have significant positive impact on the net zero agenda and on maximising material circularity.
- More collaboration between commercial developers of self-healing materials and impact modellers will help in developing updated and refined datasets to support life cycle assessment of self-healing products.

6. Data Statement

All data underlying data for this study is reported in the paper or supported by cited in the references.

Acknowledgements

This work was supported by the Engineering and Physical Sciences Research Council (EPSRC) funded project on New Industrial Systems: Manufacturing Immortality (EP/R020957/1). The authors are also grateful to the Manufacturing Immortality consortium.

References

Aïssa, B., Therriault, D., Haddad, E., Jamroz, W., 2012. Self-healing materials systems: Overview of major approaches and recent developed technologies. Adv. Mater. Sci. Eng. 2012, 1–17. https://doi.org/10.1155/2012/854203

Balogun, V.A., Kirkwood, N.D., Mativenga, P.T., 2014. Direct electrical energy demand in fused deposition modelling. Procedia CIRP 15, 38–43. https://doi.org/10.1016/j.procir.2014.06.029

Bekas, D.G., Tsirka, K., Baltzis, D., Paipetis, A.S., 2016. Self-healing materials: A review of advances in materials, evaluation, characterization and monitoring techniques. Compos. Part B Eng. 87, 92–119. https://doi.org/10.1016/j.compositesb.2015.09.057

Biesiekierski, A., Wang, J., Abdel-Hady Gepreel, M., Wen, C., 2012. A new look at biomedical Ti-based shape memory alloys. Acta Biomater. 8, 1661–1669. https://doi.org/10.1016/j.actbio.2012.01.018

Blaiszik, B.J., Kramer, S.L.B., Olugebefola, S.C., Moore, J.S., Sottos, N.R., White, S.R., 2010. Self-Healing Polymers and Composites. Annu. Rev. Mater. Res. 40, 179–211. https://doi.org/10.1146/annurev-matsci-070909-104532

Blanca-Alcubilla, G., Bala, A., de Castro, N., Colomé, R., Fullana-i-Palmer, P., 2020. Is the reusable tableware the best option? Analysis of the aviation catering sector with a life cycle approach. Sci. Total Environ. 708, 135121. https://doi.org/10.1016/j.scitotenv.2019.135121

Böckin, D., Tillman, A.M., 2019. Environmental assessment of additive manufacturing in the automotive industry. J. Clean. Prod. 226, 977–987. https://doi.org/10.1016/j.jclepro.2019.04.086

Bovea, M.D., Ibáñez-Forés, V., Pérez-Belis, V., 2020. Repair vs. replacement: Selection of the best end-of-life scenario for small household electric and electronic equipment based on life cycle assessment. J. Environ. Manage. 254. https://doi.org/10.1016/j.jenvman.2019.109679

Cohades, A., Hostettler, N., Pauchard, M., Plummer, C.J.G., Michaud, V., 2018. Stitched shape memory alloy wires enhance damage recovery in self-healing fibre-reinforced polymer composites. Compos. Sci. Technol. 161, 22–31. https://doi.org/10.1016/j.compscitech.2018.03.040

Coillot, D., Méar, F.O., Podor, R., Montagne, L., 2010. Autonomic self-repairing glassy materials. Adv. Funct. Mater. 20, 4371–4374. https://doi.org/10.1002/adfm.201000147

Cooper, D.R., Gutowski, T.G., 2017. The Environmental Impacts of Reuse: A Review. J. Ind. Ecol. 21, 38–56. https://doi.org/10.1111/jiec.12388

DePalma, K., Walluk, M.R., Murtaugh, A., Hilton, J., McConky, S., Hilton, B., 2020. Assessment of 3D printing using fused deposition modeling and selective laser sintering for a circular economy. J. Clean. Prod. 264, 121567. https://doi.org/10.1016/j.jclepro.2020.121567

Diesendruck, C.E., Sottos, N.R., Moore, J.S., White, S.R., 2015. Biomimetic Self-Healing. Angew. Chemie - Int. Ed. 54, 10428–10447. https://doi.org/10.1002/anie.201500484

European Council, 2008. Directive 2008/98/CE of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain Directives. Off. J. Eur. Union L312, 1–59.

Faludi, J., Van Sice, C.M., Shi, Y., Bower, J., Brooks, O.M.K., 2019. Novel materials can radically improve whole-system environmental impacts of additive manufacturing. J. Clean. Prod. 212, 1580–1590. https://doi.org/10.1016/j.jclepro.2018.12.017

Farrant, L., Olsen, S.I., Wangel, A., 2010. Environmental benefits from reusing clothes. Int. J. Life Cycle Assess. 15, 726–736. https://doi.org/10.1007/s11367-010-0197-y

Felfil Evo specifications, URL https://felfil.com/Felfil_Bundle_techincals.pdf?v=5ea34fa833a1 [Accessed: 13.07.2021]

Filabot specifications, URL https://www.filabot.com/collections/filabot-core/products [Accessed: 13.07.2021]

Filabot spooler specifications, URL https://cdn.shopify.com/s/files/1/0236/7897/files/FSMR1_Rev_3_03142019.pdf?107 [Accessed: 13.07.2021]

Gasol, C.M., Farreny, R., Gabarrell, X., Rieradevall, J., 2008. Life cycle assessment comparison among different reuse intensities for industrial wooden containers. Int. J. Life Cycle Assess. 13, 421–431. https://doi.org/10.1007/s11367-008-0005-0

Godina, R., Ribeiro, I., Matos, F., Ferreira, B.T., Carvalho, H., Peças, P., 2020. Impact assessment of additive manufacturing on sustainable business models in industry 4.0 context. Sustain. 12, 1–21. https://doi.org/10.3390/su12177066

Guadagno, L., Raimondo, M., Naddeo, C., Longo, P., Mariconda, A., 2014. Self-healing materials for structural applications. Polym. Eng. \& Sci. 54, 777–784. https://doi.org/https://doi.org/10.1002/pen.23621

Hager, M.D., Greil, P., Leyens, C., van der Zwaag, S., Schubert, U.S., 2010. Self-Healing Materials. Adv. Mater. 22, 5424–5430. https://doi.org/https://doi.org/10.1002/adma.201003036

Herbert, E.N., Li, V.C., 2013. Self-Healing of Microcracks in Engineered Cementitious Composites (ECC) Under a Natural Environment. Materials (Basel). 6, 2831–2845. https://doi.org/10.3390/ma6072831

Imato, K., Nakajima, H., Yamanaka, R., Takeda, N., 2021. Self-healing polyurethane elastomers based on charge-transfer interactions for biomedical applications. Polym. J. 53, 355–362. https://doi.org/10.1038/s41428-020-00432-4

Khalid, M., Peng, Q., 2021. Investigation of Printing Parameters of Additive Manufacturing Process for Sustainability Using Design of Experiments. J. Mech. Des. 143. https://doi.org/10.1115/1.4049521

Kim, C., Ejima, H., Yoshie, N., 2017. Non-swellable self-healing polymer with long-term stability under seawater. RSC Adv. 7, 19288–19295. https://doi.org/10.1039/C7RA01778B

Lee, D. Il, Kim, S.H., Lee, D.S., 2019. Synthesis of self-healing waterborne polyurethane systems chain extended with chitosan. Polymers (Basel). 11. https://doi.org/10.3390/polym11030503

Lv, L., Yang, Z., Chen, G., Zhu, G., Han, N., Schlangen, E., Xing, F., 2016. Synthesis and characterization of a new polymeric microcapsule and feasibility investigation in self-healing cementitious materials. Constr. Build. Mater. 105, 487–495. https://doi.org/https://doi.org/10.1016/j.conbuildmat.2015.12.185

Mauldin, T.C., Kessler, M.R., 2010. Self-healing polymers and composites. Int. Mater. Rev. 55, 317–346. https://doi.org/10.1179/095066010x12646898728408

Nußholz, J.L.K., 2017. Circular business models: Defining a concept and framing an emerging research field. Sustain. 9, 14–17. https://doi.org/10.3390/su9101810

Prusa FAQ, URL https://help.prusa3d.com/en/article/faq-frequently-asked-questions_1932/ [Accessed: 13.07.2021]

Ritzen, L., Montano, V., Garcia, S.J., 2021. 3d printing of a self-healing thermoplastic polyurethane through fdm: From polymer slab to mechanical assessment. Polymers (Basel). 13, 1–21. https://doi.org/10.3390/polym13020305

Saade, M.R.M., Yahia, A., Amor, B., 2020. How has LCA been applied to 3D printing? A systematic literature review and recommendations for future studies. J. Clean. Prod. 244, 118803. https://doi.org/10.1016/j.jclepro.2019.118803

Sauerwein, M., Doubrovski, E., Balkenende, R., Bakker, C., 2019. Exploring the potential of additive manufacturing for product design in a circular economy. J. Clean. Prod. 226, 1138–1149. https://doi.org/10.1016/j.jclepro.2019.04.108

Turner, C., Moreno, M., Mondini, L., Salonitis, K., Charnley, F., Tiwari, A., Hutabarat, W., 2019. Sustainable production in a circular economy: A business model for re-distributed manufacturing. Sustain. 11, 1–19. https://doi.org/10.3390/su11164291

Unger, S.R., Landis, A.E., 2014. Comparative life cycle assessment of reused versus disposable dental burs. Int. J. Life Cycle Assess. 19, 1623–1631. https://doi.org/10.1007/s11367-014-0769-3

Wang, X., Zhao, J., Chen, M., Ma, L., Zhao, X., Dang, Z.M., Wang, Z., 2013. Improved self-healing of polyethylene/carbon black nanocomposites by their shape memory effect. J. Phys. Chem. B 117, 1467–1474. https://doi.org/10.1021/jp3098796

Willocq, B., Odent, J., Dubois, P., Raquez, J.M., 2020. Advances in intrinsic self-healing polyurethanes and related composites. RSC Adv. 10, 13766–13782. https://doi.org/10.1039/d0ra01394c

Xia, N.N., Xiong, X.M., Rong, M.Z., Zhang, M.Q., Kong, F., 2017. Self-Healing of Polymer in Acidic Water toward Strength Restoration through the Synergistic Effect of Hydrophilic and Hydrophobic Interactions. ACS Appl. Mater. \& Interfaces 9, 37300–37309. https://doi.org/10.1021/acsami.7b11230

Zhang, K., Hu, Z., Zhao, Z., Wei, B., Li, Y., Wei, Y., 2018. Whiskers growth and self-healing in Ti-based metallic glasses during ion irradiation. Appl. Surf. Sci. 437, 176–180. https://doi.org/https://doi.org/10.1016/j.apsusc.2017.12.166

Zink, T., Geyer, R., Startz, R., 2016. A Market-Based Framework for Quantifying Displaced Production from Recycling or Reuse. J. Ind. Ecol. 20, 719–729. https://doi.org/https://doi.org/10.1111/jiec.12317

Appendix A – Variables and level for the L25 Taguchi Orgonal Design used in the calculation

Table 7 - Variables and levels for the L25 Taguchi array used in the calculation

	Input variables						Results			
Run	Self-healing material volume fraction (%)	3D printer electricity consumption (Wh)	Material deposition rate (g/hour)	Avoided production (%)	Transportation of self-healing product (km)	Recycling ratio (%)	Global warming (kg CO_2 eq)	Stratospheric ozone depletion (kg CFC11 eq)	Marine ecotoxicity (kg 1,4-DCB)	Water consumption (m^3)
1	20%	100	6	19%	100	10%	10.646	8.858E-06	0.821	0.192
2	20%	350	9	38%	200	30%	15.507	8.697E-06	1.189	0.148
3	20%	600	12	57%	300	50%	13.199	6.387E-06	1.009	0.094
4	20%	850	15	76%	400	70%	8.202	3.588E-06	0.622	0.045
5	20%	1100	18	95%	500	90%	2.007	8.526E-07	0.144	0.008
6	40%	100	9	57%	400	90%	2.417	1.179E-06	0.179	0.016
7	40%	350	12	76%	500	10%	4.270	2.776E-06	0.316	0.051
8	40%	600	15	95%	100	30%	1.344	7.557E-07	0.092	0.010
9	40%	850	18	19%	200	50%	66.592	2.683E-05	5.101	0.312
10	40%	1100	6	38%	300	70%	43.731	1.723E-05	3.349	0.192
11	60%	100	12	95%	200	70%	0.481	3.097E-07	0.026	0.003
12	60%	350	15	19%	300	90%	27.191	1.042E-05	2.081	0.111
13	60%	600	18	38%	400	10%	25.837	1.172E-05	1.954	0.160
14	60%	850	6	57%	500	30%	18.659	8.101E-06	1.413	0.104
15	60%	1100	9	76%	100	50%	10.674	4.472E-06	0.806	0.053
16	80%	100	15	38%	500	50%	5.402	2.759E-06	0.389	0.042
17	80%	350	18	57%	100	70%	7.848	3.261E-06	0.587	0.038
18	80%	600	6	76%	200	90%	5.764	2.257E-06	0.433	0.023
19	80%	850	9	95%	300	10%	1.786	8.396E-07	0.124	0.010
20	80%	1100	12	19%	400	30%	86.421	3.336E-05	6.590	0.359
21	100%	100	18	76%	300	30%	1.875	8.979E-07	0.120	0.011
22	100%	350	6	95%	400	50%	0.920	4.364E-07	0.058	0.004
23	100%	600	9	19%	500	70%	46.967	1.764E-05	3.579	0.180
24	100%	850	12	38%	100	90%	33.530	1.253E-05	2.563	0.126
25	100%	1100	15	57%	200	10%	24.156	9.324E-06	1.811	0.098