A Comprehensive Methodology to Support Decision-Making Towards Sustainability on Nanocomposite Materials in Additive Manufacturing Sector

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Abstract

Innovative nanocomposite materials and resultant additive manufacturing products are necessary to be assessed for their carbon footprint towards top priorities of EU for plastics, including the European Green Deal principles and the Action Plan for Circular Economy. Life Cycle Assessment (LCA) is widely applied standardized methodology that aims to study potential environmental impacts of novel products. Nano-scale materials (NM) are usually dispersed in polymer to enhance their limited functional properties resulting in a spectrum of end-products for multiple applications. However, little information exists on their environmental impact. Within this context, this study presents a 'cradle-to-gate plus end-of-life' LCA approach, studying different types of 3D printing nanocomposite filaments across the supply chain. Three different types of polymer matrices were examined: polyamide (PA), polypropylene (PP) and polylactic acid (PLA), additivated with three different types of nanomaterial additives: multiwall carbon nanotubes (MWCNTs), graphene oxide (GO) forms and graphene nanoplatelets (GNPs), considering lab-scale production. In addition, several different EoL scenarios have been examined for the materials. Finally, LCA findings are coupled with the performance (taken here as conductivity) of these new materials to assist the decision-making process for selecting efficient scenarios with the least environmental impact. The outputs of this examination enable identification of potential sustainability issues for novel nanocomposite materials at an early design stage, while also assisting in the definition of actions to mitigate such issues. Thus, LCA studies can generate knowledge on the environmental impacts of nano-enabled materials, while also serving as a valuable decision support tool towards optimizing material sustainability aspects.

Introduction

(Tofail SAM, Koumoulos EP, Bandyopadhyay A, Bose S, O'Donoghue L, Charitidis C (2018)). The increased demands on more complex products and designs in a more efficient and sustainable way reflect the advantages of AM (Gardan J (2019)). Last decades, Fused Filament Fabrication (FFF) or 3D printing, the most broad AM technique, has evolved and has displayed extensive application within industries from various sectors, including aerospace, and automotive, but also expanding to medical applications and use in the construction industry (Ngo TD, Kashani A, Imbalzano G, Nguyen KTQ, Hui D (2018)), (Wang X, Jiang M, Zhou Z, Gou J, Hui D (2017)). Mechanical and rheology property benefits, as well as ease of processing and possibility of recycling, render thermoplastic polymers as highly favorable for use as 3D printing feedstock. Various thermoplastic polymers are used in the FFF technique, including the widely applied and studied Polylactic Acid (PLA) and Acrylonitrile butadiene styrene (ABS), as well as other polymers, such as polyamide (PA), polypropylene (PP), polystyrene and polyester, which offer a different set of properties. The use of reinforcement fillers of various types and sizes enables overcoming the limitations of thermoplastic polymers as well as introducing new properties that can expand their field of application. Through incorporation of Carbon -based nanomaterials (CNMs) in thermoplastic filaments, high electrical conductivity, thermal conductivity and good mechanical strength properties are achieved, due to the intrinsic properties of the nanoadditives (Acquah SFA, Leonhardt BE, Nowotarski MS,
It is considered that carbonaceous nanomaterial species such as Carbon Nano Tubes (CNTs), Graphene Oxide (GO) and Graphene nanoplatelets (GNPs) will enable the emergence of a new group of nanocomposites with multi-functional and 'smart' properties (Farahani RD, Dubé M, Therriault D (2016)) (Spinelli G, Lambert P, Tucci V, Kotsilkova R, Ivanov E, Menseidov D, Naddeo C, Romano V, Guadagno L, Adami R, Meisak D, Bychanok D, Kuzhir P (2019)) (Kwon YJ, Park JB, Jeon YP, Hong JY, Park HS, Lee JU (2021)) (Bardot M, Schulz MD (2020)) (Tirado-Garcia I, Garcia-Gonzalez D, Garzon-Hernandez S, Rusinek A, Robles G, Martínez-Tarifa JM, Arias A (2021)). There has been extensive research work (Sandler JKW, Kirk JE, Kinloch IA, Shaffer MSP, Windle AH (2003)) (Arif MF, Alhashmi H, Varadarajan KM, Koo JH, Hart AJ, Kumar S (2020)) (Battisti A, Skordos AA, Partridge IK (2020)) (Capezza A, Andersson RL, Ström V, Wu Q, Sacchi B, Ferris S, Hedenqvist MK, Olsson RT (2019)) focused on the electrical properties of thermoplastic nanocomposites, measuring their resistivity and conductivity.

Gonçalves et al. (Gonçalves J, Lima P, Krause B, Pötschke P, Lafont U, Gomes JR, Abreu CS, Paiva MC, Covas JA (2018)) studied the development of electrically conductive polyetherketone (PEEK-based) filaments with a range of weight ratios of MWCNTs (1-6%) and GNPs (1-6%) suitable for FFF. It was demonstrated that the PEEK/MWCNTs material displays an electrical percolation threshold between 2 and 3% loading filler. Further increase in electrical conductivity could be attained by incorporating a percentage of GNP to the nanocomposite, while the addition of the nanoadditives offered improved properties in terms of Young's modulus and yield strength for the filament material. In a study by Ivanov et al. (Ivanov E, Kotsilkova R, Xia H, Chen Y, Donato RK, Donato K, Godoy AP, Di Maio R, Silvestre C, Ciminino S, Angelov V (2019)), ten nanocomposite polymer formulations were investigated in terms of structure and electrical and thermal properties. The thermoplastic matrix of the nanocomposites was polylactic acid (PLA), and 0-6% additivation for three types of carbon nanofillers (MWCNTs, GNPs and MWCNTs/GNPs) was examined. The authors observed a synergetic action between the nanoadditives in particular cases, such as the addition of GNPs in the nanocomposite that included MWCNTs, at additivation ratios of 3% GNP:3% CNT and 1.5% GNP:4.5% CNT, attributing this to a more MWCNTs-efficient network. A maximum filler content of 6% wt for the bi-filler composite led to a 7-8 order of magnitude increase in electrical conductivity compared to pure PLA. Based on the results, conductivity values of $8.4 \times 10^{-3}$ (S/m) and $2.1 \times 10^{-2}$ (S/m) for GNPs and CNTs, respectively were reached, while higher electrical conductivity values were displayed for the mono-filler nanocomposites incorporating MWCNTs, reaching a percolation threshold at 3 wt. % additivation. Podsiadły et al. (Podsiadły B, Matuszewski P, Skalski A, Sloma M (2021)) fabricated acrylonitrile butadiene styrene (ABS)-based nanocomposite filaments, through additivation with CNTs with various loadings. The mechanical and electrical properties of printed specimens were examined as well. Results indicated that printed components with below 1.96% CNTs were treated as non-conductive, while at 4.76% the electrical resistivity reduced at 2.5 Ωm. Further increase at 9.09 wt% of CNTs, reduced further the value at 0.15 Ωm. Gomez et al. (Gomez J, Villaro E, Perez J, Haidar B A (2020)) applied the process of melt compounding to produce PLA/rGO nanocomposite filaments. The electrical conductivity properties were measured for the nanocomposite incorporating 2 wt% of rGO as $1 \times 10^{-3}$ S/m. An adjusted Hummers' method was applied...
to prepare the GO material. Socher et al. (2011) prepared electrical conductive thermoplastic composites of PA12 by incorporating carbon nanotubes (CNTs) by melt compounding and demonstrated an electrical percolation threshold between 2.0 and 2.25 wt%. for the nanocomposite material.

Most of the CNTs used in different applications are MWCNTs produced by chemical vapour deposition (CVD) (Socher R, Krause B, Hermasch S, Wursche R, Pötschke P (2011) ) (Socher R, Krause B, Pötschke P (2017) ). LCAs studies on CNTs production are, however, scarce (Cossutta M, McKechnie J (2021) ) (Wu F, Zhou Z, Temizel-Sekeryan S, Ghamkhar R, Hicks AL (2020) ) (Arvidsson R (2017) ) (Gavankar S, Suh S, Keller AA (2014)). Teah et al. (Teah HY, Sato T, Namiki K, Asaka M, Feng K, Noda S (2020) ) used three lab-scale CVD synthesis methods to study the Greenhouse Gas (GHG) emissions of developed CNTs. Using LCA technique, the GHG emissions of CNT manufacturing production process was calculated, and the relevant hotspots were found supporting technological improvements.

Findings highlighted the significant impact that configuration adjustments display in the reduction of GWP, within setup parameters that include oxidative additive type (CO₂ or H₂O), reactor growth modes (2D flat-plate or 3D spherical), technique for catalyst deposition (sputtering or CVD), and purging gases selection (Ar or N₂). The highest potential and promise for achieving industrial scale of production of CNTs are considered to be displayed in continuous processes, such as fluidized bed CVD, as discussed by Healy et al. (Healy ML, Dahlben LJ, Isaacs JA (2008) ), in a study that evaluated an unspecified catalytic CVD method. For Graphene, the synthesis technologies of which are still developing, there are limited literature-based information available on the study of potential environmental impacts within the context of its synthesis methods. The most widely applied methods for Graphene synthesis are exfoliation, CVD and epitaxial growth (Sivudu KS, Mahajan Y (2012) ).

In a review study conducted by Arvidsson (Arvidsson R (2017) ) concerning the environmental life cycle assessment evaluation of different graphene production routes, five different routes were identified and taken into consideration: CVD, exfoliation (ultrasonication or thermal), chemical reduction of GO, and epitaxial growth. Significant challenges emerged in direct comparative analysis of the results, seeing that the functional unit considered in the reviewed studies was not the same. In another study, performed by Cossuta et al. (Cossutta M, McKechnie J and Pickering SJ (2017) ) a comparative cradle-to-gate LCA evaluation for three graphene production routes was conducted: CVD, electrochemical exfoliation, and chemical oxidation (followed by chemical or thermal reduction). The synthesis route displaying the lowest impacts for large quantity production of rGO was defined as the chemical oxidation process followed by thermal reduction within the LCA study. The definition of this synthesis route as the least impactful was confirmed by a complementary prospective LCA evaluation, in which impacts of a potential commercial scale were estimated.

Serrano-Lujan et al. (Serrano-Luján L, Víctor-Román S, Toledo C et al (2019)) studied the environmental impacts of the two techniques considered to present the highest efficiency towards producing rGO of high-performance properties, namely the Hummers and Marcano methods, studying in total, seven rGO production routes. Two functional units were proposed: In order to enable a direct comparative analysis
of production routes, a functional unit of 1 kg of rGO was used, while a functional unit normalized by conductivity enabled an analysis specific to the application studied. Hummer's method resulted in a decreased total energy consumption per kilogram of graphene produced. Pizza et al. (Pizza A, Renaud M, Mehrdad H, Jean-Louis B (2014)) studied the LCA of high-quality nanocomposites made of thermally conductive GNP, placing the focus on the study of the energy requirements for the stages of the nanomaterial production and nanocomposite manufacturing and excluding from the analysis the potential nanoparticle emissions and nano-waste generation. Based on the findings, the fabrication of GNP filler was found to be an energy-intensive procedure (1,879 MJ/kg), and thermal conductivity values of 1 W/mK are achieved by incorporating 5.8% wt% filler in the manufacturing process for 1 kg of epoxy composite. Currently, in terms of EoL scenarios for plastic waste generated in Europe, it has been presented that 75% is recycled, considering 32% recycling & 43% energy recovery and the 25% is landfilled (Crippa M, De Wilde B, Koopmans R, Leyssens J, Muncke J, Ritschkoff A-C, Van Doorselaer K, Velis C, & Wagner M (2019)). Therefore, it is reasonable to consider the potential for nanocomposite recycling when designing the production of nano-enabled products. The market concerning 3D printing filament feedstock presents considerable growth potential, as it is projected to display a compound annual growth rate (CAGR) of 28.1% up to 2025, estimated at USD 739 million in 2020 (3D Printing Filament Market by Type (Plastics, Metals, Ceramics), End-Use Industry (Aerospace & Defense, Medical & Dental, Automotive, Electronics), Region (North America, Europe, Asia Pacific, MEA, South America) - Global Forecast to 2025). The global growth rates of carbon nanotubes and graphene markets are projected to reach 10.7% (Carbon Nanotube Market 2021, Allied Market Research) and 38.7% (Graphene Market Size, Share & Trends Analysis Report By Application (Electronics, Composites, Energy), By Product (Nanoplatelets, Oxide), By Region, And Segment Forecasts, 2020 - 2027), respectively, up to 2027. Based on these market growth rates, and following the trend of nanomaterial incorporation for introduction of unique properties to the 3D printing filament materials, it can be projected that production volumes of nano-enabled filament will increase and the prospect of these products ending up in landfills and incineration plants should be avoided.

In this regard, LCA plays a critical role in the evaluation of the potential impacts of these new technologies and the different EoL treatment management options; in this manner LCA assists in the direction of research and innovation activities aimed at achieving environmentally compatible, sustainable products that could comply to the circular economy framework (Arvidsson R, Tillman AM, Sandén BA, Janssen M, Nordelöf A, Kushnir D, Molander S (2018)).

The current study examines the environmental performance via LCA of multiple nanocomposite filament through melt-compounding (extrusion) suitable for FFF investigating three different thermoplastic matrices (PA, PP and PLA), three different carbon-based NMs (CNT, GNP and rGO) and alternative EoL treatment options (current and future projections EoL treatments). Moreover, the environmental impact of fillers' loadings on different matrices has been coupled with the nanocomposite performance, taken as electrical conductivity, to assist the decision-making process by choosing the compounding with the required performance and the least environmental impact. As a result, the purpose of this study is to highlight key open difficulties that must be solved with a solid holistic approach in order to support
sustainable nano-enabled products in accordance with the European Green Deal and the circular economy era that it promotes.

**Methodology**

The current attributional LCA study employs the general framework provided in ISO 14040:2006 (ISO 14040:2006) and ISO 14044:2006 (ISO 14044:2006) including the four iterative phases: (a) Goal and scope definition; (b) Inventory analysis; (c) Impact assessment; and (d) Interpretation. Finally, LCA analysis was performed using SimaPro v9.1.

1.1. Goal and scope

The goal of the study is to evaluate a variety of nano-composite filaments in terms of their environmental impact and use the respective LCA results to strengthen decision-making in order to achieve an efficient and sustainable filament production. Three thermoplastics PA, PP and PLA in combination with different concentration of nano-fillers (0.5-10wt%): multiwall carbon nanotubes (MWCNTs) synthesized by chemical vapour deposition (CVD), graphene oxide (GO) via Hummer's method and graphene nanoplatelets (GNPs) synthesized by graphite exfoliation method have been investigated. Since PLA, PA and PP are the most common used polymers in the AM sector and the most widely used polymers in 3D printing fabrication, they have been selected to be examined as the matrices, of the nano-composites filaments, in combination with the most-used NM fillers for the respective applications.

Table 1 presents the different combinations-scenarios, among the polymeric matrixes, the filler types and their respective loadings in wt% of each filler as well as the EoL options that has been considered in the LCA study. For this study a ‘cradle-to-gate plus EoL’ approach has been considered. Thus, the system boundaries of this study include Materials: the production of raw materials (NMs and thermoplastics), Manufacturing: the melt-compounding (extrusion) process for the filament die manufacturing and the EoL management where different scenarios were examined; the use stage has been excluding (Figure 1).

As EoL scenarios incineration, landfill and recycling alternatives have been considered. The functional unit (FU) for all the assessed scenarios is mass-based at 1 kg of nanocomposite filament produced by melt-compounding. Concerning NMs they were modelled considering lab scale production and the following process routes per case: GNP: Expanded, exfoliated graphite pulverized, GO: Hummer’s method, mild bath sonication and CNTs: Fluidized bed CVD, N$_2$/20 bead cycles, lab scale production.
| Polymer matrix | Filler type (NM) | Filler load wt% | End-of-life (shares%) |
|----------------|-----------------|-----------------|-----------------------|
| PLA            | GNP             | 0.5, 1, 2, 3, 4, 5, 7.5 & 10 wt% | Incineration (100%)   |
|                | GO              |                 |                       |
|                | CNT             |                 |                       |
| PA             | GNP             | 0.5, 1, 2, 3, 4, 5, 7.5 & 10 wt% | Incineration (35%)    |
|                | GO              |                 | Landfill (10%)         |
|                | CNT             |                 | Recycling (55%)        |
| PP             | GNP             | 0.5, 1, 2, 3, 4, 5, 7.5 & 10 wt% | Incineration (35%)    |
|                | GO              |                 | Landfill (10%)         |
|                | CNT             |                 | Recycling (55%)        |

### 1.2. Life Cycle Inventory (LCI)

Background life cycle inventory data were sourced from ecoinvent v3.6 (Moreno-Ruiz E, Valsasina L, FitzGerald D, Brunner F, Symeonidis A, Bourgault G, Wernet G (2019)), where available, and supplemented by data from open access literature as detailed in sections below. In terms of electricity mix, the background data was updated for EU requirements. The present study did not take distribution and transportation into account. For the type of PA used in composites, nylon 6-6 was selected.

#### 1.2.1. Production of Nanomaterials

CNT production process routes can vary in terms of their environmental impact in a product life cycle perspective; for the present LCA study a ‘high quality’ CNT synthesis methodology has been selected for the data inventory’s development. Teah et. Al. (Teah HY, Sato T, Namiki K, Asaka M, Feng K, Noda S (2020) ) gave the high environmental impact of on-substrate CVD method, indicating its higher compatibility towards a smaller scale, in contrast with the fluidized-bed methods of CVD, which offer increased suitability for larger scale applications or bulk uses. In this study, the electricity generation was based on Japan data, and the background system data were acquired from the Ecoinvent 3.4 database. Therefore, data were regenerated in ecoinvent 3.6.

Based on the step-based process that has been presented above, additional LCI information is provided. Taking into consideration that CVD-based methods have been defined as commercially viable technologies due to cost, uncomplicated operation, as well as ease of scalability, CVD has been selected as the most representative technology (Upadhyayula VKK, Meyer DE, Curran MA, Gonzalez MA (2012)).
Considering the limitations of the present literature that has evaluated the environmental aspects of CNTs, within the context of this study, the CVD method is defined as the most promising technology for carbon nanotube synthesis, since substantial variations are seen in environmental impacts based on CNT synthesis method. The definition of CVD as a synthesis method is coupled with the best-case scenario adapted from (Teah HY, Sato T, Namiki K, Asaka M, Feng K, Noda S (2020)) for the laboratory scale manufacturing of CNTs, as well as the application of N₂ as a carrier gas and 20 times of substrates cycle.

The inventories of the Graphene oxide (GO) and the Graphene Nanoplatelet (GNP) production process have been developed based on data received by (Serrano-Luján L, Víctor-Román S, Toledo C et al (2019)) and (Pizza A, Renaud M, Mehrdad H, Jean-Louis B (2014)), respectively.

1.2.2. Manufacture of the composite filament

Plastic film extrusion as in ecoinvent 3.6, modified to include only the process (extrusion) resources was used as proxy to model the melt-compounding process. The process's productivity maintained at 98%.

1.2.3. End-of-life management

The study considers landfill disposal, incineration or recycling processes as EoL scenarios, since most products are not biodegradable. Considering that there are displayed different rates of EoL management across different time periods and sectors in Europe, the EoL pathways for 2030 based on the EC principles outlined in the “European strategy for plastics (EC (2018)) in a circular economy in 2030” are defined based on Maga et al. (Maga D, Hiebel M, Aryan V (2019)). Specifically, a target for establishing a recycling rate for plastics in packaging to 55% by 2030 has been set, within the EU strategy for Packaging and Packaging Waste (Directive (EU) 2018/852 of the European Parliament and of the Council of 30 May 2018 amending Directive 94/62/EC on packaging and packaging waste.), while there has been an aim to reduce landfilling to a 10% maximum, within the context of the Circular Economy Package. As a result, incineration treatment will cover the remaining 35% max. In Table 2 all the relevant details concerning the materials examined and their different EoL options modelling in Ecoinvent 3.6 are presented.

| EoL pathway | Modeled in Ecoinvent 3.6 | PP (%) | PA (%) | PLA (%) |
|-------------|--------------------------|--------|--------|---------|
| Incineration | Municipal solid waste {RoW} | 35     | 35     | 100     |
|             | treatment of incineration |        |        |         |
|             | APOS, U                  |        |        |         |
| Landfill    | Municipal solid waste {CH} | 10     | 10     | 0       |
|             | treatment of sanitary landfill |        |        |         |
|             | APOS, U                  |        |        |         |
| Recycling   | PP (waste treatment) {GLO} | 55     | 55     | 0       |
|             | recycling of PP | APOS, U |        |        |         |

Table 2
EoL alternative management routes (shares) considered in this study per polymer
Additional studies performed by Boldrin et al., (Boldrin A, Hansen, S F, Baun A et al. (2014) ) and Hansen et al., (Hansen SF, Heggelund RS, Besora PR, Mackevica A, Boldrin A, Baun A (2016) ), which studied the predicted disposal paths patterns for nanomaterial-including waste present in municipal waste, have demonstrated that >50% is projected to end up in recycling. Incineration and landfill EoL pathways display a tendency between 13-38% and 8-29%, respectively. Previous studies that examined the EoL pathways for nanomaterials, had evaluated that most of the waste including or contaminated with nanomaterials would be landfilled (60-91%) or found in sludge (19-52%) (Keller AA, Wang H, Zhou D, Lenihan HS, Cherr G et al. (2010) ), (Keller A A, Lazareva A). For the PA recycling process, the PP waste treatment recycling from ecoinvent has been considered as proxy and Nylon 6.6 as proxy for the avoided material. Three different alternative options have been examined for the worst-case filament, on an aim to define climate change mitigation, since the EoL treatment pathways display inherent variations. These EoL alteratives, include a) 100% recycling, b) 100% landfill and c) 100% incineration.

1.3. Life Cycle Impact Assessment

On the basis of recommendations by the Joint Research Centre (JRC) of the European Commission on Life cycle assessment methods, the International Life Cycle Data (ILCD) method (Sala S, Wolf M, Pant R (2012) ) was used in the LCA study, for the calculation of the environmental impacts, and assessment of all 16 midpoint impact categories was performed. The global warming potential (GWP) is presented in the following sections, since it is considered the most robust impact category, on the basis of the Intergovernmental Panel on Climate Change's (IPCC) 2007 report for a 100-year period. The data for the rest of the impact categories is displayed on the Supplementary Information.

Results

Both Life Cycle Impact Assessment and interpretation of the LCA results are presented in this section including: i) the environmental impact of the assessed materials, ii) a sensitivity analysis illustrating the effect of filler % on the Climate Change (CC) of the nanocomposites, (iii) the effect of different EoL scenarios in LCA results and (iv) the coupling of the nanocomposites’ conductivity performance into the LCA findings.

2.1. Environmental impacts of thermoplastics and NMs

When considering only the polymer matrix ‘from cradle-to-gate’, PLA has the lowest value 0.93 kg CO₂ eq/kg PLA, followed by PP with 1.84 kg CO₂ eq/kg and PA with 8.03 kg CO₂ eq/kg. PLA's low CC value is attributable to biogenic carbon absorbed from the atmosphere, as opposed to the other two petroleum-based thermoplastics PA and PP, which are made from fossil fuels.

However, PP presents the lowest CC value 1.57 kg CO₂ eq/kg, followed by PLA 2.20 kg CO₂ eq/kg and PA 4.36 kg CO₂ eq/kg, when the EoL (on a ‘cradle-to-grave’ approach for 1 kg of polymer) was considered. Once a cradle-to-grave approach is applied, the biogenic carbon balances out since the amount of CO₂ absorbed in PLA is released to the atmosphere by thermal treatment during the EoL stage. The difference
in CC values for the examined polymers indicate the importance of considering the EoL stage into a LCA, especially when accounting for biobased products.

When assessing only the fillers in a ‘cradle-to-gate’ approach, CC shows the least impactful NM is GNP releasing 0.074 g CO$_2$ eq/g followed by CNT 0.246 kg CO$_2$ eq/g and GO 0.293 kg CO$_2$ eq/g. The selected nanomaterials’ production pathways are energy-intensive procedures; hence electricity is the main contributor to the CC values. Results from all sixteen impact categories assessed are presented in Tables S1 (CNTs, GNPs and GO) and S2 (PA, PP and PLA) of the supplementary information.

2.2. Environmental impacts of nanocomposite filaments

The Climate Change impacts of the examined nanocomposite filaments at filler concentrations from 0.5% – 10% are illustrated in Figure 2. In the Table S3 of the supplementary information, each scenario's specific values are presented. Increase filler concentration - independent of the nano-filler type, results in higher environmental impacts. However, the CC impacts of thermoplastics reinforced with GNPs, as expected from the results in section 2.1, performs better in terms of environmental impact in CC category than the respective nanocomposites with CNTs and GO inclusions.

For the polymer-GO scenarios, CC values higher than 35 kg of CO$_2$ eq per kg of lament occur at 10 wt% of filler, releasing approximately 5.99 (PA-GO system), 9.95 (PP-GO system) and 8.64 (PLA-GO system) times more CO$_2$ emissions than the respective 0.5 wt% scenarios. In terms of Global Warming Potential (GWP), all GO-based filaments perform worse than CNT-based alternatives, whereas GNP-based filaments are the most sustainable in terms of CC impact. For the polymer-CNT scenarios, CC values between 25-30 kg of CO$_2$ eq per kg of filament are obtained when 10 wt% of NMs are used releasing approximately 4.91 (PA-CNT system), 8.51 (PP-CNT system) and 7.28 (PLA-CNT system) times more CO$_2$ emissions than the respective 0.5 wt% scenarios. Therefore, it becomes obvious when CNT or GO are used as fillers, they dominate the CC even at low amounts (2-3%). The same does not count when GNP is used as it has the least CC compared to CNT and GO. Hence, the polymer-GNP scenarios release significant lower CO$_2$ eq amounts even at high filler percentages (i.e., less than 11.3 kgCO$_2$eq at 10wt% NMs).

For the nine different polymer-nanomaterial scenarios it is also observed a similar trend in the rest of the impact categories. Tables S4, S5 and S6 (in the supplementary) are presented results for the PLA, PP and PA, respectively. The sixteen impact categories assessed were normalized and results are shown in Figure 3 for the system 1 kg PA-GO 3 wt%. As illustrated in Figure 3, the categories with the most significant impact are the freshwater ecotoxicity, the human toxicity both cancer and non-cancer, and the ionizing radiation. The hot spot analysis indicates that for the examined 3 wt% filler, the GO production process dominates almost all impact categories.

2.3. Environmental impact of alternative EoL treatment

Regarding the EoL stage, different EoL management routes have been considered for the composite filaments, as shown in Table 2. The environmental impacts of the nine products are shown normalized by
the worst-case scenario (PA-GO) in Figure 4, while the absolute values in kgCO$_2$eq. are presented in Table 3 for the greenhouse gas (GHG) emissions excluding biogenic carbon impact category. The FU was 1kg of nano-enabled polymer filament in all cases. The fillers seem to increase the total impacts of the composite filament, even though the polymers play a significantly smaller role. This applies for the nanocomposite materials incorporating CNT and GO. On the other hand, in the event of GNP additivation, the contribution of the polymer has a considerably higher influence in the total impacts.

A positive environmental impact appears for PA and PP due to the recycling share that was considered at the EoL stage. This alternative EoL treatment gives credits to the overall GWP by avoiding the production and use of virgin raw material.

| Table 3 | Contribution Climate change impacts (kgCO$_2$eq.) of the filaments with different matrices and constant (3wt%) filler concentration |
|---------|------------------------------------------------------------------------------------------------------------------|
| Product | Polymer | Filler | End-of-life management | Total |
| PA-CNT  | 8.1086115 | 7.3868693 | -3.6768119 | 11.818669 |
| PA-GNP  | 8.1086115 | 2.2351327 | -3.6768119 | 6.6669323 |
| PA-GO   | 8.1086115 | 8.7784867 | -3.6768119 | 13.210286 |
| PLA-CNT | 1.0398487 | 7.3868693 | 1.264576 | 9.6912941 |
| PLA-GNP | 1.0398487 | 2.2351327 | 1.264576 | 4.5395574 |
| PLA-GO  | 1.0398488 | 8.7784867 | 1.264576 | 11.082911 |
| PP-CNT  | 2.0489122 | 7.3868693 | -0.24089991 | 9.1948816 |
| PP-GNP  | 2.0489122 | 2.2351327 | -0.24089991 | 4.04314499 |
| PP-GO   | 2.0489123 | 8.7784867 | -0.24089991 | 10.586499 |

PA with GO inclusions is presented as the filament with the highest contribution in comparison with the alternatives with an impact of 13.21 kg CO$_2$ eq. in climate change impact category. The PA-GO baseline scenario was modeled accordingly: 55% Recycling, 35% Incineration and 10% Landfill. Due to the fact that the EoL management alternatives, that were assessed in this work, are based on future estimations, different EoL scenarios were also examined and compared with the worst case (worst performing filament) scenario which was the PA-GO as shown in Figure 5 (normalized results) while the respective absolute values in kgCO$_2$eq. are presented in Table 4. Table S7 of the supplementary information illustrates the impact on climate change of the composite filaments produced by PA and GO as a filler in various loadings (wt%) and EoL management pathways.
Table 4
EoL scenarios for the worst performing filament (PA-GO) in terms of kgCO$_2$eq. at 3 wt% filler

| EoL scenarios | PA-GO baseline | PA-GO 100% recycling | PA-GO 100% landfilled | PA-GO 100% incinerated |
|---------------|----------------|----------------------|-----------------------|------------------------|
| kgCO$_2$eq.   | 13.21          | 9.27                 | 17.57                 | 18.15                  |

In 100% recycling scenario, the PA-GO filament shows a reduction of 30% in CC value due to the credits gained from the increased avoided materials than the baseline. A negative effect is observed in CC values for the 100% Incineration and 100% landfill scenarios. A respective increase of 37% and 33% was calculated.

An efficient, standardized and sustainable waste management approach for nanomaterials is hindered due to nano-enabled products not possessing specific-labelling as of yet, leading to challenges in sorting and separation of the nanomaterial-containing waste streams. Hence, according to a study performed by Musee et. al. (Musee N (2011) ), the current waste management principles are projected to be significantly challenged as inadequate, due to the rise of nanotechnologies, requiring novel management pathways.

Based on the findings of several studies investigating the potential for incineration of waste containing CNTs, it has been reported that CNTs can decompose through the process, provided that temperatures are retained constantly high at the incinerator. However, if temperature control is not achieved in these high levels, CNTs will end up in bottom ash (Andersen L, Christensen F M, Nielsen J M, COWI A/S (2014)).

Bouillard et al. (Bouillard J, Bullet B, Bullet M, Moranviller D (2013) ) examined the incineration of nanocomposite materials containing CNTs (ABS + 3 wt.% MWCNTs). The authors demonstrated that release of CNTs is observed, on case of low temperature (450 ºC) upstart phase of the incineration process, thus also highlighting the significance of continuous function of solid waste incinerators to achieve high temperatures that can cause CNT decomposition.

2.4. LCA results coupled with performance to support decision-making

Ranking environmental impacts of composite filaments at ascending order, as shown in Figure 6, allows to compare impacts and offers a range of appropriate options to the end-user towards low-emission materials.

The LCA results presented in this figure are useful information considering the performance in terms of composite filament’s environmental impact. PP-GNPs 4 wt% appears to have lower CC impact in comparison to PLA-GNPs 0.5 wt%. This is not an expected result considering that PLA has an impact of 0.93 kgCO$_2$eq/kg while the PP presents a higher impact around 1.84 kgCO$_2$eq/kg. However, including the EoL stage into the assessment we observe that it plays an important role to the overall filament’s impact
indicating the PP based composite filament as the least environmentally friendly option. These values in combination with the respective conductivities (or any relevant performance indicators), could assist the decision makers towards a more sustainable option based on their application needs.

The critical aspect for the transition from an insulating polymer to conductor is that amount of the filler above which no major differences in electrical conductivity of the composite exist, known as electrical percolation threshold (EPT) (Rahaman M, Aldalbahi A, Govindasami P, Khanam NP, Bhandari S, Feng P, Altalhi T (2017)). The goal of producing conductive nanocomposite filaments is to break through the EPT and the same time retaining 3D printing rheological and processing parameters. Various factors such as the filler type, quality, dispersion, aspect ratio have proved to affect the percolation threshold of nanocomposites (Gnanasekaran K, Heijmans T, Van Bennekom S, Woldhuis H, Wijnia S, De With G, Friedrich H (2017)). Fillers with high aspect ratio ratio show a significant electrical conductivity evident even at low concentrations in thermoplastic polymers (Gao Y, Picot O T, Zhang H, Bilotti E, Peijs T (2017)) (Gomez J, Villaro E, Perez J, Haidar B A (2020)). Single wall CNTs and MWCNTs have an inherent electrical conductivity that ranges between 102-106 (S/cm) and 103-105 (S/cm) respectively, while for graphene this range is reported to be between $10^4$-10$^5$ (S/cm) (Mittal G, Dhand V, Rhee K Y, Park S J, Lee W (2014)) (Marsden AJ, Vallés C, Liscio A et al (2018)).

In Figure 7a decision-making map is presented that could support a greener AM process concerning the production of PLA nanocomposite filaments. Conductivity values and percolation threshold for the PLA composite filaments have been provided by literature, as presented in Table 5 and Table S8 of the supporting information. As shown, for PLA the options that surpass the percolation criterion per ascending CC values are: PLA-GNP 7.5wt% < PLA-rGO 2wt% < PLA-CNT 5wt% < PLA-CNT 7.5wt%. The filler concentration and the type of filler are strongly influence filament’s performance and this is illustrated in the decision-making map. PLA-GNP 7.5wt% would be the best option in terms of performance and environmental impact despite the fact that is under the almost red area. However, even at low concentration of rGO the electrical percolation threshold is achieved while for CNT and GNP higher concentrations are required. Therefore, PLA-rGO 2 wt% would be the second most preferable option. Relevant stakeholders such as manufacturers could set a threshold regarding performance (e.g., Conductivity) above which could select among different sustainable options (green area). Thresholds can be specified based on the specific requirements of nanocomposites' applications.
While the literature examined and utilized in this work provides data that enable the study of the NMs and nanocomposite materials, substantial variations in data are observed in terms of the environmental impacts of the materials/processes. For example, for the GO materials, inventories are limited, and significant uncertainty is present on the data. Based on this shortcoming, it is highlighted that the development of LCI databases for nanomaterials, supported by the optimal alternative nanomaterial synthesis routes would be an important enabler for future LCA studies. Additionally, the potential nanoparticle emissions during the various processes of nanomaterial synthesis or nanocomposite manufacturing, as well as the recycling process routes are not taken into consideration. Seeing that these life cycle phases could present environmental impact hotspots, future studies could contribute to addressing this information gap, as well as consider broader environmental and societal aspects.

Seeing that the carbon-based nanomaterials may also entail occupational Health & Safety issues (Cossutta M, McKechnie J (2021)), that could be displayed throughout the nanocomposite life cycle (ISO/TR 22293:2021), an important complemental element in the approach presented in this work would be the introduction of occupational safety aspects within the decision support criteria. 

| Thermoplastic + Filler (wt. %) | Conductivity (S/m) | Reference |
|-------------------------------|-------------------|-----------|
| PLA/1.5% GNP                  | 2.51E-08          | (Ivanov E, Kotsilkova R, Xia H, Chen Y, Donato RK, Donato K, Godoy AP, Di Maio R, Silvestre C, Cimmino S, Angelov V (2019)) |
| PLA/3% GNP                    | 9.90E-08          | (Ivanov E, Kotsilkova R, Xia H, Chen Y, Donato RK, Donato K, Godoy AP, Di Maio R, Silvestre C, Cimmino S, Angelov V (2019)) |
| PLA/6% GNP                    | 8.35E-03          | (Ivanov E, Kotsilkova R, Xia H, Chen Y, Donato RK, Donato K, Godoy AP, Di Maio R, Silvestre C, Cimmino S, Angelov V (2019)) |
| PLA/1.5% MWCNT                | 1.40E-08          | (Ivanov E, Kotsilkova R, Xia H, Chen Y, Donato RK, Donato K, Godoy AP, Di Maio R, Silvestre C, Cimmino S, Angelov V (2019)) |
| PLA/3% MWCNT                  | 7.86E-04          | (Ivanov E, Kotsilkova R, Xia H, Chen Y, Donato RK, Donato K, Godoy AP, Di Maio R, Silvestre C, Cimmino S, Angelov V (2019)) |
| PLA/6% MWCNT                  | 2.10E-02          | (Ivanov E, Kotsilkova R, Xia H, Chen Y, Donato RK, Donato K, Godoy AP, Di Maio R, Silvestre C, Cimmino S, Angelov V (2019)) |
| PLA/0.5% rGO                  | 8.00E-10          | (Gomez J, Villaro E, Perez J, Haidar B A (2020)) |
| PLA/1% rGO                    | 4.00E-07          | (Gomez J, Villaro E, Perez J, Haidar B A (2020)) |
| PLA/2% rGO                    | 1.00E-03          | (Gomez J, Villaro E, Perez J, Haidar B A (2020)) |

\( ^{a} \) approx. values from graph
To achieve this, nano-specific risk assessment/management techniques such as control banding (e.g., the Stoffenmanager Nano module (Van Duuren-Stuurman B, Vink SB, Verbit KJV, Heussen HGA, Brouwer DH, Kroese DED, Van Niftrik MFJ, Tielemans E, Fransman W et al (2012)) could be applied, since they display compatibility towards connection with the LCA outcomes. Materials and processes involved throughout the life cycle of each scenario could be assessed and allocated in control bands, indicating the risk levels displayed in each process/material, and thus the level of occupational safety controls required. Therefore, each life cycle scenario would be accompanied by an occupational risk classification, characterizing the processes applied to reach the final product. In this way, scenarios involving inherently safer nanoprocesses (e.g., nanomaterial synthesis through wet chemistry, as opposed to synthesis in powder phase) could be highlighted.

Additionally, another dimension through which the concept of Health & Safety may be introduced within the analysis is the emission potential of the studied filaments during the print process. It has been previous reported that FFF 3D printing could display emission hazards, particularly concerning Ultrafine Particles (UFP) and Volatile Organic Compounds (VOCs) (Karayannis P, Petrakli F, Gkika A, Koumoulos EP (2019) ). This could cause to operators in charge of the printing process possible health hazards. Imperatively, nano-enabled filaments may release airborne particles containing nanomaterials into the 3D printing workplace air, as has been confirmed in the literature (Stefaniak AB, Bowers LN, Knepp AK, et al (2018) ), rendering the importance of introducing this aspect all the more crucial for the nanocomposite filament materials. Recent advances in the emission assessment research for FFF 3D printing have led to the development of standardized methodologies to perform emission measurements (UL Standards, ANSI/CAN/UL) during the 3D printing processes. Through application of such methodologies, a series of emission measurement campaigns for select highly promising materials (e.g., the cases that present favorable results in terms of conductivity/environmental impact) could reveal which of the material alternatives display higher or more hazardous emissions during their ultimate use as 3D printing filaments. In this way, the concept of consumer safety (filament purchaser/user in this case) is also introduced in the approach.

Crucially, a set of additional aspects referring to the processability of the various filaments represented in the life cycle scenarios could further embellish the presented decision support methodology. It has been reported in the literature that introduction of nanomaterials in the polymer filaments could significantly impact material printability (Dey A, Eagle INR, Yodo N (2021) ). Therefore, cross evaluation of the printability aspects with the effectiveness and sustainability criteria would enable identification of the all-round more viable and advantageous alternative.

The development and refinement of a complementary approach involving the above-described elements (LCA, safety, performance, processability), could facilitate alignment and coordination between the technical work towards process/material optimization and the study of sustainability aspects of the respective technologies.

**Conclusions**
In this work, production and disposal of nanocomposite 3D printing filaments are studied in terms of life cycle climate change impacts, on an aim to provide a decision-support tool towards sustainable development. End of life management of such materials was also discussed. Based on the results, the GWP of the nanocomposite filaments varies based on polymer and nanofiller type, ranging from 4.04 kg CO₂ eq. (PP-GNP) to 13.21 kg CO₂ eq. (PA-GO). GO and CNT fillers have been identified as key hotspots within the nanocomposite filament life cycle. For the polymers, specifically in the case of PA matrix, significant climate change impacts are displayed, due to comparatively more energy-intensive processes than the other thermoplastic alternatives. Considering the considerable climate change impact displayed for PA-based nanocomposites, the EoL treatment options present high significance towards reducing climate change impact potential across the material life cycle. The worst-performing filament (PA-GO) was examined in terms of alternative EoL treatment scenarios, and the results showed a ± 30% variation based on the applied treatment approach, while recycling presented the most favorable performance amongst the EoL alternatives in this case. Coupling LCA results with the respective conductivities (or any relevant performance indicators) offers an area of suitable development alternatives, which can support the selection of options balanced in terms of sustainability/performance, in accordance with application-specific requirements. Additive manufacturing and nanotechnology are expected to gain significant market share among emerging technologies. Still, nanotechnology introduces potential unknown risks to human health and ecosystems. Thus, waste community need to anticipate recycling and EoL management opportunities and in close collaboration with LCA practitioners to achieve the zero waste and circularity targets considering that both sectors (AM and nanotechnology) are rapidly developing.

Declarations

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Consent for publication
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The authors declare that they have no competing interests

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Foteini Petrakli (FP), Anastasia Gkika (AG), Anestis Vlysidis (AV), Panagiotis Karayannis (PK) and Elias Koumoulos (EK) were contributed to the development of this manuscript. AV and FP analyzed the data regarding LCA. FP and AG performed the literature review and were the major contributors in writing the manuscript. PK contributed to the nanosafety part. EK made the literature review concerning performance. All authors read and approved the final manuscript.

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Figures

System boundaries of the examined LCA study
Figure 2

Climate change impacts per FU for all nanocomposite filaments with NMs, at different filler concentration (0.5%-10%)

Figure 3

Normalization of the LCA results for the sixteen impact categories accessed and hot spot analysis
Figure 4

Normalized contribution to Global Warming Potential (GWP) impacts of the filaments with different matrices and constant (3wt%) filler concentration.
Figure 5

Normalized EoL scenarios for the worst performing filament (PA-GO) in terms of GWP impacts at 3 wt% filler

![Graph showing normalized EoL scenarios for PP, PA, and PLA filaments.]

Figure 6

Ranked Climate change values at ascending order for PP, PA & PLA composite filaments at different filler concentration (0.5%-10%)
Figure 7

Climate Change values coupled with measured conductivity and EPT, based on literature, to support

Supplementary Files

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