



TECHNICAL ARTICLE

A Preliminary Study on the Sustainability of Metallization of Polymer Matrix Composites through Cold Spray

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The cold spray (CS) process is the latest solid-state deposition method that has gained significant awareness for the metallization of polymer matrix composites (PMCs) materials to modify their surface properties, including electrical or thermal conductivity and electromagnetic shielding. In comparison with other coating processes, CS allows for the production of metallic coatings without the necessity to reach the melting temperature of the sprayed particles and provides a way to deposit resistant materials with improved properties onto various components to minimize wear, erosion and corrosion. For the first time in the literature, this work has the innovative goal of applying the life cycle assessment methodology to the case study of the CS production process in order to ascertain whether CS could be framed in the realm of green technologies offering interesting opportunities to improve manufacturing sustainability. In particular, the environmental impact of CS associated with the metallization process of PMCs was considered in terms of energy consumption and CO₂, NO_x and SO₂ emissions, which are used as indicators in the life cycle assessment. When compared to different coating processes, the results suggest that CS has a large potential to reduce the environmental effects connected with the products in terms of the amount of CO₂ and hazardous emissions created throughout the process.

Keywords cold spray, green industry, LCA, metallization, PMCs, sustainability

1. Introduction

The use of composite materials, in particular PMCs, is spreading in automotive and aerospace industries due to their low weight and high stiffness compared to metals (Ref 1). The applications of composite materials in engineering fields could be widely extended with the enhancement of some superficial properties of PMCs. For instance, the manufacturing of a metallic layer on the surface of PMCs can modify their surface properties such as electrical conductivity, thermal conductivity, electromagnetic shielding, erosion, radiation and lightning protection. In this way, a novel and more functional composite structure can be obtained. Hence, the metallization process of PMCs would seem to be the viable solution. To date, there exist

different techniques for the metallization of polymer-based materials, such as electroplating (Ref 2), chemical vapor deposition (Ref 3) or physical vapor deposition (Ref 4). Unfortunately, these techniques have disadvantages that strongly limit their uses and applications (Ref 5, 6). Among the thermal spray techniques, the cold gas dynamic technology (simply referred to as cold spray) is gaining interest due to its multiple advantages (Ref 7). The process can be schematized as follows: micro-sized particles (metal, polymer or ceramics) ranging normally from 10 to 100 μm (Ref 8) are delivered from a powder feeder into a de Laval nozzle by a stream of powder carrier gas and then accelerated by the propellant gas, usually helium, nitrogen or air. The propellant gas is heated by an electric heater and injected at high pressure into the nozzle where the mixing with the powders occurs. In the nozzle, the gas-powders mixture is accelerated to supersonic conditions. As the particles exit from the nozzle at high velocities between 300 and 1200 m s^{-1} and impinge on a substrate, they undergo significant plastic deformations and bond to the substrate (Ref 9). The particles are heated up below the melting point of the material and remain in a solid state for the entire process (Ref 10). The main characteristic of the CS technique is that it relies more on kinetic energy than on thermal energy and that makes this technology suitable for the metallization of temperature-sensitive materials like polymers and composite materials.

CS offers benefits in terms of efficiency in the use of resources which, added to some peculiarities in the design and production of the objects with the best performance, generate economic and environmental advantages (Ref 11). Although it has been shown that cold spraying is a viable metal deposition coating technology with numerous advantages, in the last years great attention is devoted to CS's contribution to sustainability (Ref 12). In particular, CS is a technology having the potential to be framed in the realm of green manufacturing processes, as

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propellant gases are inert (helium, nitrogen, or air), and the heating source is electric and controlled at temperatures below the melting temperature of the material being sprayed (Ref 13). In this way, the melting of metal powder and consequent harmful emissions are avoided. The CS process can also be used to replace less environmentally friendly processes such as chrome plating, which produces hexavalent chromium emissions or to overcome “spot” imperfections on the coating of the components obtained with other plating and thermal spraying processes, which normally should be completely stripped and repainted to heal the defect present in the area of interest (Ref 14). The advantages of the CS for sustainable production can be divided into four main areas: (i) low environmental impact of the process, (ii) repair rather than replacement, (iii) advanced performances, which improves the useful life of the components, and (iv) alternative to more polluting and/or less sustainable processes.

The life cycle assessment (LCA) methodology is used internationally for identifying, comparing and reducing the environmental impacts of processes, products and services (Ref 15). When applied to processes, it requires quantifying the environmental impacts of resources used (materials and energy) and emissions (solid, liquid and gaseous) at each stage of a process (Ref 16). Most environmental impact assessment models or methods are developed based on the general framework of LCA and quantified environmental impact data can be obtained based on new unit process models of various processes with different LCA boundaries and cutoffs (Ref 17, 18). Currently, at the base of every multi and interdisciplinary approach, the concept of environmental sustainability is central, whose boundary, due to the breadth of its potential application context, can range from economic growth to respect for nature, affecting the most diverse disciplines. In the same way, also in the manufacturing field, much of the attention is increasingly paid to the identification and understanding of the main environmental impact factors in order to grasp the ability of industries to contribute to making production processes sustainable and in order to reduce carbon dioxide emissions and reduce problems related to ecotoxicity (Ref 19-22).

To the authors’ best knowledge, there are only a few papers in the literature dealing with the sustainability aspects of CS (Ref 23, 24) and the questions that arise from the above considerations are: (i) How sustainable is the CS process? (ii) How much does the CS impact the reduction of carbon dioxide emissions? In this scenario, the LCA approach can represent the possible solution to answer these questions. Among the few papers in the literature dealing with the sustainability aspects of CS, none of these deals with LCA applied to the case study of metallization of PMCs through CS. Therefore, this research aims to assess the metallization of the polymer matrix composites materials through the CS technology from a global environmental standpoint. In particular, the environmental impact of CS associated with the metallization process of PMCs was considered in terms of energy consumption and CO₂, NO_x and SO₂ emissions, which are used as indicators in the life cycle assessment.

For this purpose, Onyx material has been chosen for the substrate to be coated. This material is made of nylon thermoplastic polymer with chopped carbon fibers randomly dispersed in it (Ref 25, 26). A DYMET low-pressure cold spray (LPCS) machine was used for the metallization process; air was used as the carrier gas while the inlet gas pressure was kept constant at 0.6 MPa (Ref 27, 28).

The influence of the CS process parameters on the deposition was analyzed in terms of gas temperature effects; for this scope, four different inlet gas temperatures were considered: $T_1 = 200\text{ }^{\circ}\text{C}$, $T_2 = 300\text{ }^{\circ}\text{C}$, $T_3 = 400\text{ }^{\circ}\text{C}$ and $T_4 = 500\text{ }^{\circ}\text{C}$.

The main goal of this research was to evaluate the ecological impacts of the CS manufacturing processes with respect to process parameters in order to give relevant information about environmental impacts.

2. Materials and Methods

2.1 Schematization of the Process

The LCA methodology was used in this study to calculate the environmental impacts of the metallization of PMCs by means of CS process. LCA is a cluster of methods and procedures used to examine the ecological effects and physical flows related to the manufacturing of products and the supply of services over the entire life cycle, from raw material acquisition and processing to manufacturing, implementation and end-of-life disposal. The LCA methodology evaluates all phases of the product’s life in considering all the inputs and outputs (such as polluting emissions, energy and material utilization and wastes), in contrast to conventional methods, which focus on just one stage of the product life. The LCA framework is divided into four primary instances: (i) *defining the research’s aims and scope*: which products are being investigated, the functional units, system boundaries, assumptions and limits, the intended application and rationale; (ii) *inventory analysis (LCI)*: comprises of data gathering and processing techniques aimed at quantifying the relevant input and output flows of a product system based on the purpose and field of application; (iii) *impact assessment (LCIA)*: life cycle impact assessment aims to estimate the magnitude of possible environmental consequences using the results of life cycle inventory analysis; (iv) *the interpretation phase* that is a systematic process that identifies, qualifies, verifies and evaluates the results of the inventory and impact assessment phases in order to present them in a form that fulfills the requirements described in the objective and scope of the application definition phase, as well as to draw conclusions and indications (Ref 29). In this work, a simplified LCA was applied in order to obtain a more suitable study. The LCA was applied according to the guidelines provided by ISO 14040 and ISO 14044 (Ref 30). The LCA software application Gabi was used to tackle the development of the study.

In this activity, the whole process, from the manufacturing of the substrate to the metallization of the substrate, has been schematized by identifying three macroblocks, as shown in Fig. 1: (i) the production of the powders (usually obtained with gas atomization); (ii) the manufacturing of the PMCs (generally indicated with fibers and matrix); and (iii) the metallization process through CS. For each block, it should be necessary to determine the resources and the necessary electrical energy consumption, as well as the emissions and waste produced.

In this preliminary work, the LCA analysis was focused only on the CS deposition process and a gate-to-gate analysis was applied. In particular, the energy consumption of the process itself with respect to the CS process parameters is reported. In this perspective, two different input subsystems

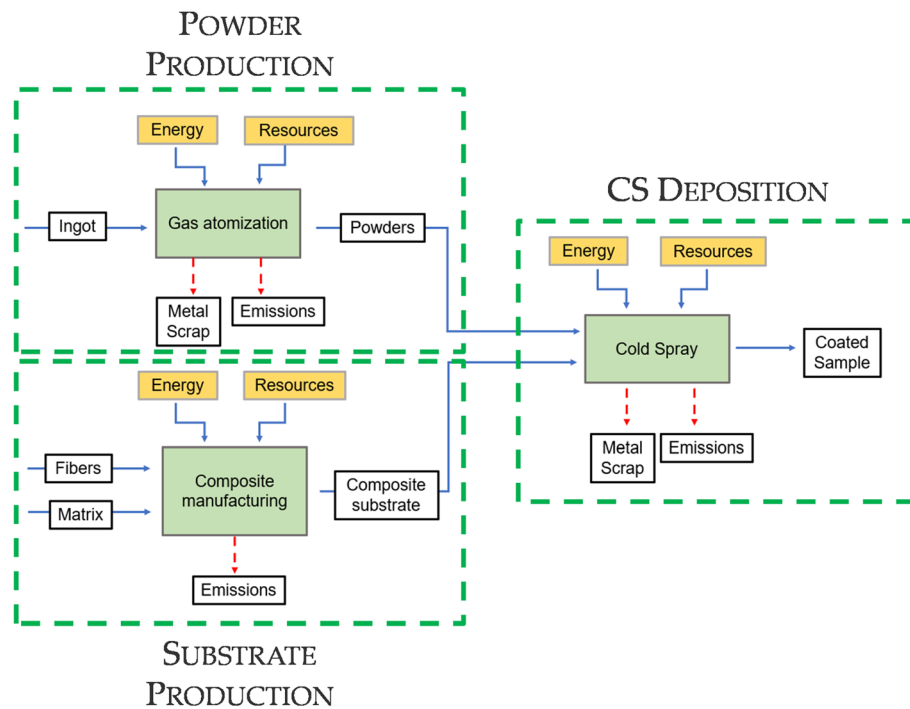


Fig. 1 Scheme of the metallization process through CS of PMCs

were considered: (i) the CS equipment (comprehensive of the inlet gas heater, the powder feeder and the nozzle that can move with a given scan velocity) and (ii) the gas supply system. Field data for the study were obtained from software databases and from the electrical energy consumption of the equipment, as described as follows.

2.2 System Description and Boundaries

The PMCs panels (80 × 80 mm) were manufactured through the Markforged facility. In this process, the plastic filament (Onyx filament made of nylon mixed with short carbon fibers) is molten by means of a heated extrusion head and applied in a single layer to the platform. Each additional layer is applied with the previous links (since it is melted) and, after cooling, the material hardens for the manufacturing of the final products (Ref 31).

A DYMET 423 low-pressure CS machine was used for the metallization process. It consists of a composite nozzle with a converging-diverging fixed nozzle and an interchangeable divergent part; all the geometry features are summarized in Della Gatta et al. work (Ref 32). Air was used as propellant gas for the acceleration of the particles. Gas-atomized aluminum powder, with a size ranging from 15 to 45 μm , was chosen as feedstock material (Ref 8). The most suitable CS process parameters in terms of gas pressure, standoff distance (SoD) and scan velocity were set through preliminary experimental tests as well as the literature results (Ref 27) and were kept constant during the deposition. Four different inlet gas temperatures were considered for the analyses and the gas flow rate was calculated through the isentropic flow models, with respect to de Laval nozzle dimensions, as shown in this work (Ref 33). In detail, a single-track coating was developed on the surface of each laminate by spraying only one layer of aluminum particles. Three tracks on each specimen were manufactured under the same process conditions for the

repeatability of the results. An experimental example of the metallized specimen through CS under given working conditions is shown in Fig. 2. Moreover, the coating properties in terms of adhesion to the substrate were evaluated. The adhesive strength was measured following the ASTM D4541 standard and by using a PosiTest ATM. Aluminum dollies with a 10 mm diameter were bonded to the top surface of the cold-sprayed deposits using cyanoacrylate glue. Any excess adhesive or coating surrounding the dolly was removed with a drill bit. All sets of the CS process parameters along with the resulting adhesion test values are reported in Table 1.

For simplicity of calculation and progressive evaluations, an interval of 280 s has been considered as the required time to successfully metallize the Onyx substrate at a fixed scan velocity. Qualistar Plus Power and Energy Quality Analyzer CA8331 (Chauvin Arnoux) was the device adopted to record electrical power and energy consumption and the Power Analyzer Transfer PAT2 software was used to analyze these parameters over time. The study was conducted with respect to the time variable, with a measurement period of 1 s.

ReCiPe2016 midpoint level method has been adopted for the analysis of LCA results (Ref 34). It provides a state-of-the-art method to convert life cycle inventories to a limited number of life cycle impact scores on midpoint (problem oriented) and endpoint (damage oriented) level. In the case of midpoint level, 17 impact categories are taken into account (climate change, photochemical ozone formation, terrestrial acidification, etc.). At endpoint level, most of the midpoint impact categories are multiplied by certain damage factors and then they are aggregated into 3 endpoint categories. (These impact categories include: Human health, Ecosystems and Resources scarcity.)

The midpoint characterization factor selected for climate change is the widely used global warming potential (GWP), which quantifies the integrated infrared radiative forcing increase of a greenhouse gas (GHG), expressed in kg CO₂ Eq.

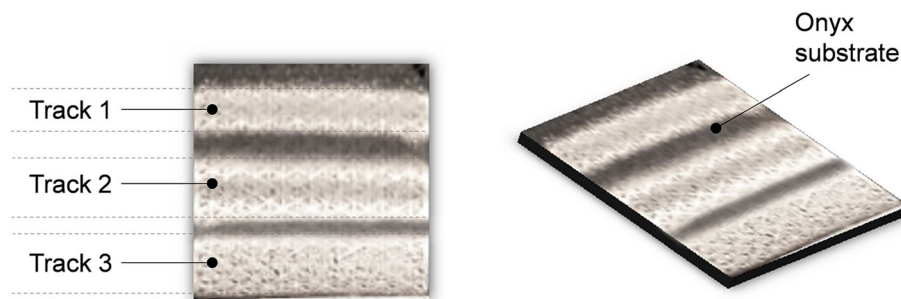


Fig. 2 Different views of onyx-based PMCs metallized using the CS process parameters under Set 2

Table 1 Set of CS process parameters and resulting coating properties

Set	Gas temperature, °C	Gas flow-rate, g/s	Gas pressure, MPa	SoD, mm	Scan velocity, mm/s	Adhesion strength, MPa
1	200	5.39	0.6	45	3.5	5.5 ± 0.3
2	300	5.06	0.6	45	3.5	4.2 ± 0.4
3	400	4.5	0.6	45	3.5	3.8 ± 0.2
4	500	4.2	0.6	45	3.5	4 ± 0.1

For the midpoint characterization factors of photochemical ozone formation related to human exposure, the human population intake of ozone was considered. Human health ozone formation potential (HOFP) is expressed in kg NO_x Eq. The change in ambient concentration of ozone after the emission of a precursor (nitrogen oxides (NO_x) or non-methane volatile organic compounds (NMVOC)) was predicted with the emission concentration sensitivities matrices for emitted precursors from the global source–receptor model TM5-FASST (Ref 35). For the midpoint characterization factors of acidifying emissions, the fate of a pollutant in the atmosphere and the soil were taken. Acidification potentials (AP) are expressed in kg SO₂ equivalents. Changes in acid deposition, following changes in air emission of NO_x, NH₃ and SO₂, were calculated with the GEOS-Chem model (Ref 36). Subsequently, the change in acidity in the soil due to a change in acid deposition was derived with the geochemical steady-state model PRO-FILE.

3. Results and Discussion

3.1 Electrical Power and Energy Consumption

The electrical power that was absorbed by the CS equipment during the deposition of the single-track aluminum coating on the PMCs is reported in Fig. 3. Note that the electrical power measurements obtained by the power and energy consumption instrument are reported over time.

Different curves corresponding to the different inlet gas operating temperatures are compared in Fig. 3. The first peak of absorbed power can be observed when turning on the equipment, then the power drops to a fixed value and remains constant throughout the entire process. For all the operating temperatures, the first peak is about 2.8 kW. Considering the mean value of the absorbed power, the mean energy consumption expressed in watt-hour of the CS equipment is also reported on the right side of the figure. It can be seen that the

energy consumption increases with the increase of the inlet gas temperature, reaching the highest value when the temperature is set to 500 °C, due to the greater electrical energy required by the CS heating system (Ref 37). However, under these process conditions, the electrical power consumption of CS can be considered relatively low (ranging from 1.4 to 2.8 kW) if compared to traditional thermal spray processes (Ref 38, 39). In particular, the electric power consumption in typical plasma torches for atmospheric plasma spraying (APS) techniques is up to 80 kW and can reach 200 kW in a water-stabilized torch or even 250 kW in high-power ones. Moreover, the electric power consumption is typically in the range 5–10 kW in arc spraying and more than 80 kW in vacuum plasma spraying. Finally, the electric power consumption of the relatively new pulsed plasma spraying ranges from 100 kW to 1 GW (Ref 40). The benefits on energy consumption of using the cold spray technology for deposition are clear; moreover, based on the results found in the literature dealing with the CS metallization of Onyx-based composites, when the inlet gas temperature is relatively low and set to 300 °C, degradation phenomena of the polymer are avoided, and the manufacturing of the coating is possible efficiently (Ref 27, 28).

3.2 CS Emissions

To assess the sustainability of the CS process applied to PMCs, as described above, only the results from the metallization of the substrate are shown. In particular, the two different subsystems including the CS equipment and the compressed gas system are considered for the analysis. The characterization assigns all inputs and outputs of the production systems to the impact categories. The individual results of the impact categories will be discussed below. Especially, the factors that have a great influence on the impact category results will be analyzed. From the LCA analysis, three impact categories are shown: (i) the climate change quantified in terms of equivalent kilograms of CO₂ produced (kg CO₂ Eq.) (Ref 41), (ii) photochemical ozone formation quantified in terms of equivalent kilograms of NO_x produced (kg NO_x Eq.) (Ref 42)

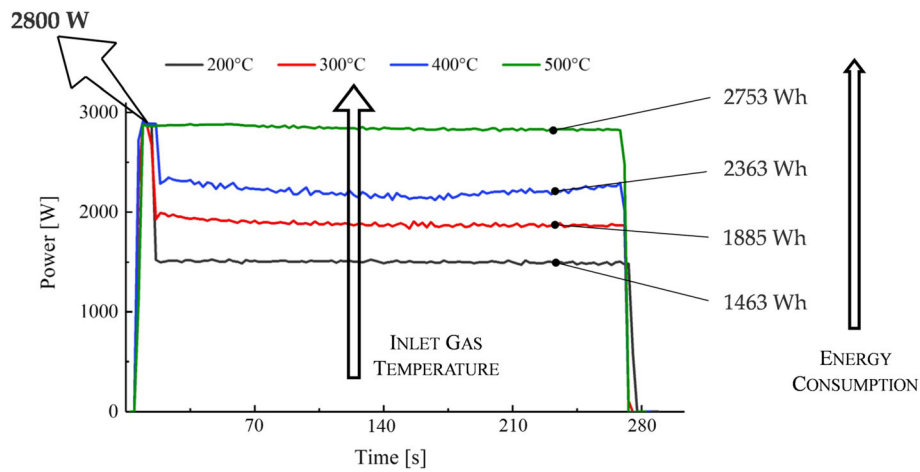


Fig. 3 Results of electrical power consumption of the cold spray equipment during the deposition of the single-track aluminum powder for the surface metallization of PMCs

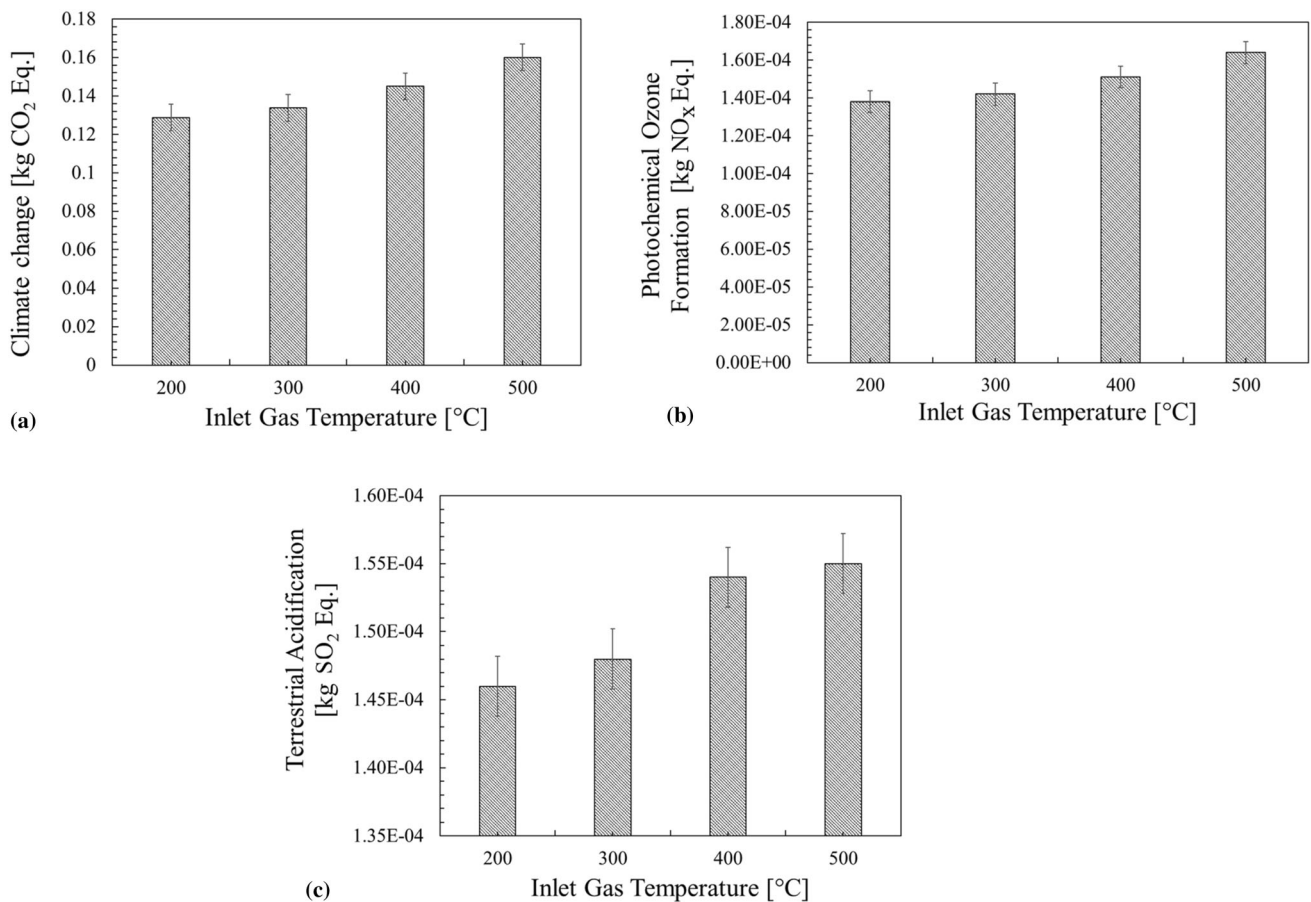


Fig. 4 LCA results of cold spray process applied to PMCs: (a) the climate change quantified in terms of equivalent kilograms of CO₂ produced per sample; (b) the photochemical ozone formation quantified in terms of equivalent kilograms of NO_x produced per sample; (c) the terrestrial acidification quantified in terms of equivalent kilograms of SO₂ produced per sample

and (iii) terrestrial acidification quantified in terms of equivalent kilograms of SO₂ produced (kg SO₂ Eq.) (Ref 43).

The total average results for each impact category from the manufacturing processes are shown in Fig. 4 and reported with respect to each sample. It is evident that when increasing the inlet gas temperature, the amount of emissions increases due to

the greater electrical energy required, as shown above. However, it can be assumed that, in general conditions, the CS system is characterized by a relatively low number of emissions when used to metallize PMCs (Ref 44). Also, it is worth noting that the emissions do not depend on the material powder used for the CS deposition but only on the equipment

and the operating process parameters. In addition, the benefits of CS, in terms of CO_2 , NO_x and SO_2 emissions were proved by comparing the results obtained with those reported in the literature. There were found a reduction of CO_2 , NO_x and SO_2 emissions equal around to 80%, 75% and 82%, respectively, if compared to those of laser engineered net shaping (LENS) technologies used for the same production time (Ref 45). Note that only the electrical energy consumption of the LENS equipment and the gas supply system data, in terms of emissions, were considered for comparison, in agreement with what was analyzed in this paper.

Aiming to analyze the dependence of the process parameters on the CS environmental impact, the contribution of the CS equipment (consisting of the inlet gas heater, the powder feeder and the nozzle that can move with a given scan velocity) and of the compressed air supply system in terms of emissions were analyzed separately and reported in Fig. 5.

At low operating temperatures, the compressed air supply system was the main contributor to all impact categories during the manufacturing phase. When increasing the inlet gas temperature, the contribution of the compressed air supply

system in terms of emissions is decreased while the contribution of the CS system is increased. This is due to the higher electrical power consumption of the CS equipment, as shown in Fig. 3, which is required for increasing the inlet gas temperature. On the other hand, when increasing the gas temperature, the air flow rate is reduced (see Table 1) and the air mass for the entire deposition is reduced (Ref 46, 47). As a consequence, the amount of emission decreased as decreasing the air mass. Anyway, it is worth noting that the influence of the CS system on the total amount of emission is more prominent compared to the compressed air supply system.

4. Conclusions

The increased awareness of the environmental impact of products and production processes has led to the development of many methods and tools for assessing the environmental impact of different manufacturing processes. In this work, a first approach for the analysis of the CS process in terms of

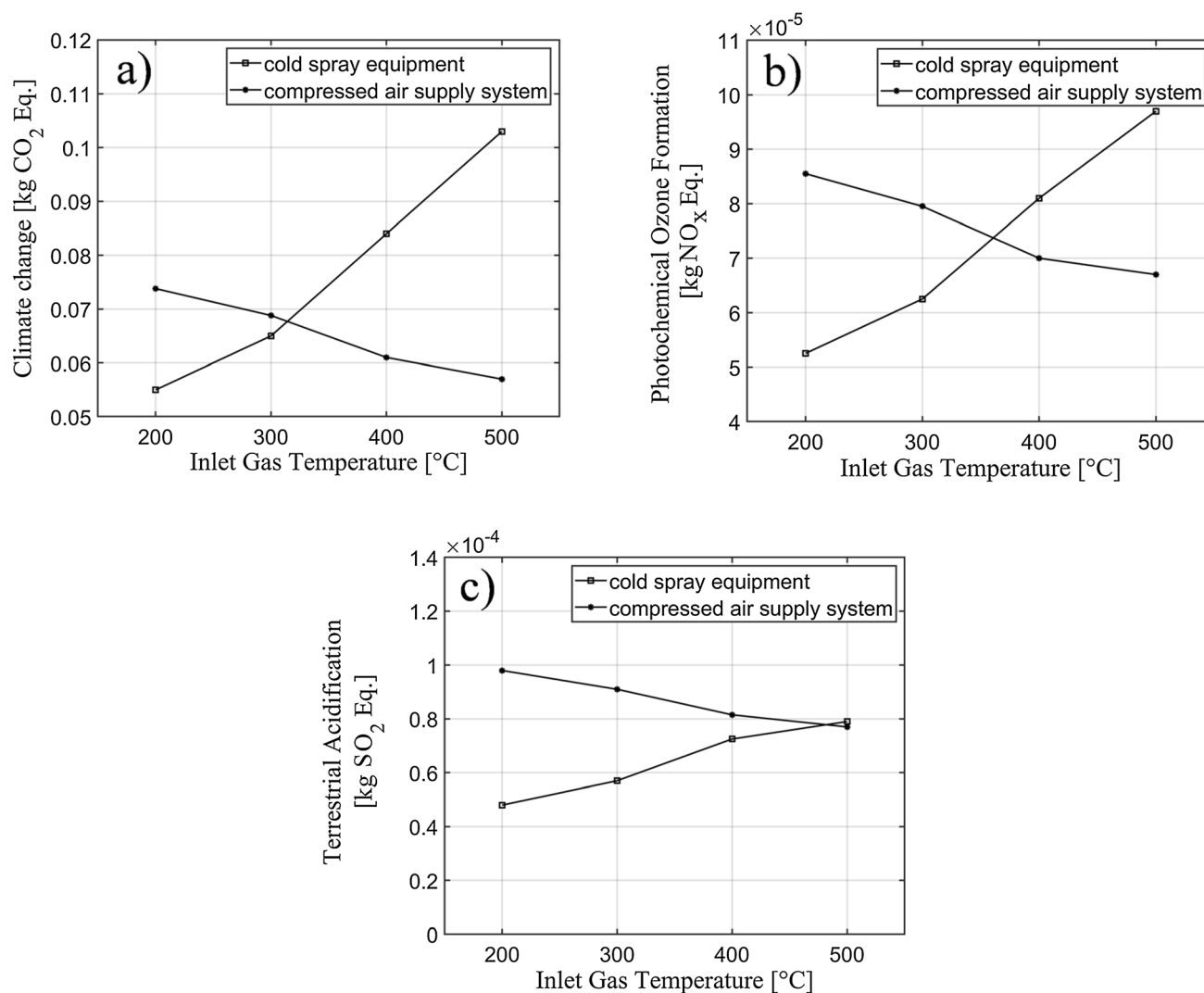


Fig. 5 Comparison of emissions from the cold spray equipment and the compressed air supply system: (a) the climate change quantified in terms of equivalent kilograms of CO_2 produced per sample; (b) the photochemical ozone formation quantified in terms of equivalent kilograms of NO_x produced per sample; (c) the terrestrial acidification quantified in terms of equivalent kilograms of SO_2 produced per sample

sustainability has been proposed. The results show that the relatively new CS technology in the field of additive manufacturing has great potential to reduce the environmental impacts, relating to the products. In particular, the amount of emissions produced during the process is relatively low compared to other coating techniques. In particular, there were found a reduction of CO₂, NO_x and SO₂ emissions equal around to 80%, 75% and 82%, respectively, if compared to those of laser engineered net shaping (LENS) technologies used for the same production time. The main impacts of the CS process are attributed to the electrical energy associated with the CS equipment. Energy consumptions differ according to the process parameters. In fact, according to the operating parameters, power consumption of the CS equipment and emissions increase when increasing the temperature. It can be said that the new CS technology shows the potential to lead to an increase in efficiency and a reduction of environmental impacts in the metallization of PMCs. In fact, there are no toxic fumes or harmful emissions from the process compared to other coating techniques. A deeper analysis of the relationship between the process setup and emissions is needed and further scientific research and technological development based on this study are required.

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