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Abstract

The environmental strategies of European Union on resource and waste management are based on lifecycle thinking. In surface finishing, they encourage the substitution of conventional wet deposition processes that involve chemicals and effluents by "dry" processes, such as thermal spraying. However, all metal finishing processes have certain impacts on human health, ecosystems, and resources. The life cycle assessment (LCA) methodology is used internationally for identifying, comparing and reducing the environmental impacts of processes, products and services. 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.

This paper presents a LCA comparison of electroplating and various thermal spray processes (plasma, twin-wire arc, HVOF, and cold spraying) for the formation of nickel coatings. It was carried out using a peer-reviewed database of upstream materials and energy (EcoInvent v2.0), and the LCA program SimaPro 7.1.7. The uses of materials and energy, and the corresponding emissions of each coating process, were converted to potential impact scores on human health, ecosystems, and resource conservation (fossil and mineral resources) by mean of the Eco-Indicator-99 method. The Impact 2002+ method that links the life cycle inventory results to four damage categories (human health, ecosystem quality, climate change, and resources) was also used to see how the results given by different methodologies differ in the cases of the study and, also, get more confidence for the interpretation of the results.

Introduction

Nickel electroplating is extensively used for decorative, engineering, and electroforming applications because coatings properties can be varied over a wide range by controlling the composition and other operating parameters of the plating process. The property generally sought for functional industrial uses is corrosion resistance, but wear resistance, solderability, magnetic and other properties may be relevant in specific applications (Ref 1-2).

Engineering and electroforming purposes represent about 20% of the nickel consumed in plating with the rest used for decorative applications. The annual worldwide consumption of nickel for electroplating, including nickel consumed as plating salts, was about 140,000 metric tons in 2006 and accounted for 11 to 12 percent of the world's nickel consumption (Ref 3).The most common nickel plating solution used for functional industrial applications is nickel sulfamate. It produces pure and ductile deposits with very low intrinsic stress (about 27.8 MPa) and hardness ranging from 150 HV to 500 HV, by varying the operating conditions.

The current wide application of nickel plating indicates that this process will continue to exist for at least another generation. However, price competition and, especially, new environmental regulations on processes, materials and products may force significant changes. For example, a recent survey of "The future of metal finishing" by Chalmer (Ref 4) identifies nine potential technology changes (Ref 4) that could affect this industry. A potential change is from conventional "wet" surface finishing to alternative "dry" technologies such as thermal spray processes. However, this will only occur if the novel alternative processes improve deposit quality, reduce costs, are more energy-efficient, and generally are deemed to represent more sustainable manufacturing. Indeed, regulatory restrictions on a growing number of finishing materials for whole categories of products, informed primarily by E.U. efforts to minimizing environmental and health risks, will continue to reshape market demand for various metals and coatings (Ref 4).

A Life Cycle Assessment (LCA) of electroplating and alternative processes can help to compare the environmental impacts associated with these processes. It is an internationally recognized methodology that provides a framework for identifying, comparing and improving "cradle-to-grave" environmental impacts associated with a product, process and service. This methodology is defined broadly by the ISO-standards 14040-44 (Ref 5).

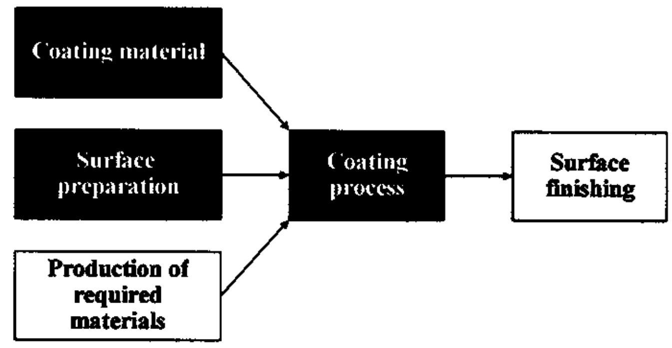

Figure 1: Generic surface coating model for the LCA study

LCA Methodology

LCA assesses the environmental impacts associated with a product, process, or service, by means of:

- compiling an inventory of relevant energy and material inputs and, environmental releases;
- evaluating the potential environmental impacts associated with identified inputs and releases.

It involves four phases: goal definition and scoping, inventory analysis, impact assessment, and interpretation.

Goal Definition and Scoping

This study compares nickel electroplating with four thermal spray processes (wire-arc, plasma, HVOF and cold spray) for the manufacturing of nickel coatings. The basis for comparison (functional unit) is a 1-m^2 surface that is coated with a 150 μm layer of nickel. The objective of the study is to establish baseline information for each process and, then, examine the relative environmental burdens. The baseline consists of the energy and resource requirements and the environmental loadings of each process.

The boundaries of each analyzed system include (i) the process and materials used to manufacture the coating material, (ii) the substrate surface preparation process and (iii), the deposition process. They exclude the manufacturing of the infrastructure materials (spray gun, powder feeder, electroplating tanks, etc.), ventilation installation (considered equivalent for all processes) and any surface finishing process. Also, the degreasing process before the surface preparation process was not included.

Figure 1 shows a generic diagram of a deposition system and its boundaries in this study while Figures 2a and 2b present the respective electroplating and thermal spray sub-processes.

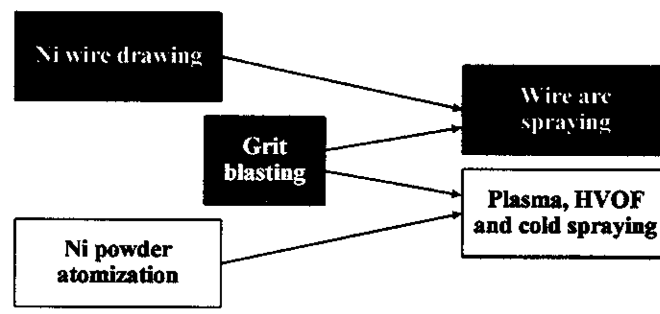

Figure 2 a: Nickel thermal spray model

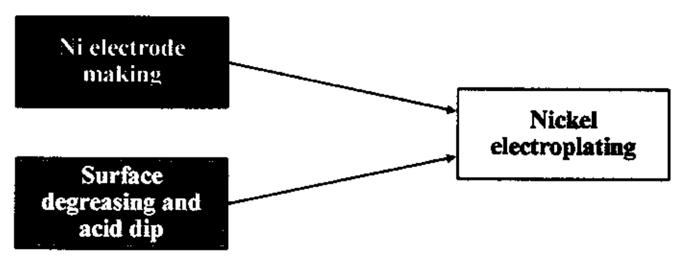

Figure 2 b: Nickel electroplating model

Inventory

The data for nickel electroplating were obtained from the literature while the thermal spraying models describing material and energy inputs and emissions were based on the recommendations of spray systems manufacturers and the authors' experience.

Nickel Electroplating

As noted earlier, the sulfamate nickel (nickel salt of sulfamic acid) solution is the most commonly used solution for electroplating, with the sulfamate ion contributing to the low stress characteristics of the deposit. Boric acid is used as pH buffer and chloride is used to aid in dissolving the nickel anodes more uniformly and prevent anode polarization. The process involves a "clean" or anodic etch, followed by the main plating step. Air emissions from the plating process are scrubbed through a wet scrubber. There are also "drag-out" losses and related aqueous nickel emissions, when the coated parts are removed from the plating bath. Aqueous emissions are sent to a wastewater treatment plant. The plating process parameters are summarized in Table 1.

Table 1: Nickel sulfamate electroplating parameters (Ref 1, 2, 6)

	Best case	Worst case	Unit
Deposition time	2.567	3.484	h
Nickel bath efficiency	95	70	%
Ni sulfamate	300 (180g/l Ni)		g/L
Ni chloride	10		g/L
Boric acid	30		g/L
Anti-pitting agent	0.3		g/L
Sodium saccharinate	5		g/L
Anode	Sulfur depolarized nickel		
Anode bag	polypropylene		
Part moving	cathodic or air		
Filter	1 vol/h		
Current density	5		A/dm²
Voltage	5		V
Nickel density	8900		kg/m³
Deposited mass	1.335		kg

Two scenarios, based on a set of best and worst case assumptions for nickel plating, were considered. They differed in deposition process efficiency (95 and 70%) and water use per square meter for the rinsing step (8 and 13 l/m²). The French regulation requires 8 l/m² treated per rinsing function (Ref 6). The rinsing baths after each cleaning or coating steps are responsible for drag-out losses and then emissions to water. A drag-out loss of 0.1 l/m² was assumed after each step (Ref 7).

Table 2: Alkaline degreasing and acid dip parameters (Ref 6, 7)

Alkaline cleaning		
Solution	Sodium carbonate (Na_2CO_3)	10 g.L^{-1}
	Trisodium Phosphate ($Na_5P_3O_{10}$)	20 g.L^{-1}
	Sodium gluconate ($NaC_6H_{11}O_7$)	24 g.L^{-1}
	Caustic soda (NaOH)	50 g.L^{-1}
Bath temperature	60°C	
Treatment time	10 min	
Acid dip		
Solution	Sulphuric acid (100 ml/l)	184 g.L^{-1}
Anode	Lead	
Current density	25 Adm^{-2}	
Bath temperature	50°C	
Treatment time	1 min	

In this study, alkaline degreasing was considered, followed by an electrolytic acid dip to remove oxides before plating. The parameters of these processes are shown in Table 2.

In the electroplating process, all the aqueous emissions are sent to a wastewater treatment plant. The water treatment process was not included in the LCA model of the present study.

Materials: Only the chemical products used during the coating process (chemicals swept with the "drag-out" losses) were taken into account in the inventory. The anode material was considered to be pure nickel and its consumption was equivalent to the mass of nickel deposited, plus a small amount of nickel loss in the bath, due to process inefficiency.

Emissions to air and water: The French regulation on water emissions of the electroplating process limits the nickel emissions to 2 mg/l. Therefore, when considering a water use of 8 l/m², the corresponding nickel emission to water was estimated to be 16 mg after wastewater treatment and 26 mg for water use of 13 l/m². However, in the actual plating process, 18 g of nickel were lost in drag-out and will require a special treatment for recovering. Other chemicals swept with drag-out losses were considered as water emissions.

Nickel emissions to air depend on process efficiency and deposition time and were calculated according to the EPA AP-42 document (Ref 6, 9). Calculations made for a wet scrubber system yielded 0.056g of air nickel emission for a process efficiency of 95% and 0.076g for a process efficiency of 70%. The full inventory of chemicals used in nickel electroplating is summarized in Table 3 (Ref 6).

Table 3: Nickel electroplating inventory (per square meter)

Alkaline cleaning	best case	worst case	Unit
Sodium Carbonate use	1		g
Trisodium Phosphate use	2		g
Sodium gluconate	2.4		g
Caustic soda use	5		g
Rinse with wastewater discharge	8	13	L
Acid dip			
Electricity use	0.42		kWh
Sulfuric Acid use	18.4		g
Rinse with wastewater discharge	8	13	L
Electroplating			
Nickel use	1.405	1.735	kg
Electricity use	6.418	8.711	kWh
Emission to water			
Nickel to water (from nickel sulfamate)	16	26	mg
Nickel chloride	1		g
Boric acid	3		g
Sodium saccharinate	0.5		g
Rinse with wastewater discharge	8	13	L
Emission to air			
Nickel (using wet scrubber system)	0.056	0.076	g

Thermal Spraying

Four thermal spraying systems are studied: plasma spraying using a PTF4 gun, wire-arc spraying with a JP 9000 gun, cold spraying using a GT3000 gun and HVOF using a JP 5000 spray gun that uses kerosene and oxygen. The use of energy for ventilation was not included in the study. Non-deposited nickel is assumed to be present as airborne particulate and is removed through a HEPA filter whose spent filter cartridges are disposed of in a hazardous waste landfill or preferably returned to a nickel smelter. The parameters of the thermal spray processes are summarized in Table 4.

Table 4: Parameters of the thermal spray processes

	Wire arc	Cold spray	Plasma	HVOF	Unit
Coating porosity	15	0	5	1	%
Spraying efficiency	70	80	80	90	%
Deposition rate	3.15	2.88	1.92	3.78	kg/h
Power input	27 V, 100 A	/	55 V, 600 A	/	/
Gas	Air	N_2	Ar / H_2	Kerosene K1/ O_2	/
Gas flow rate	76.44 (60 PSI)	61 (28 bar)	3.3 / 0.6	0.0193 (liq) / 51 (gas)	m^3/h
HEPA filter efficiency	99%				

Before the coating process, the surface was prepared by grit-blasting using white corundum (α-Al_2O_3). Fifty kg of corundum was used to prepare a surface of 1 m^2. The parameters of the preparation process are described in Table 5. Two scenarios were considered of the number of times that the blasting grit was recycled: 2 times and 50 times.

Table 5: Grit blasting parameters

Grit blasting	
Blasting media (250 µm average diameter)	a-Al_2O_3
Blasting time [min]	25
Air flow [$m^3.min^{-1}$]	0.373
Media flow rate [$kg.min^{-1}$]	2
Electric energy used to the blast system [kWh]	3
Alumina consumption (2x recycled) [kg]	25
Alumina consumption (50x recycled) [kg]	1

Materials: The four thermal spraying processes use nickel, either in the form of powder or wire. The powder used in the cold spray, plasma and HVOF processes was assumed to be produced by the EIGA powder atomization process (ALD Company (Ref 8)) that includes vacuum melting of nickel metal and inert gas atomization with argon. In this process, argon was assumed to be 70% recycled and 30% of energy was lost during the melting process.

Nickel wires used in the wire-arc process are made by a wire drawing process that consists in the successive reduction of the diameter of a drawn wire, produced by vacuum melting of nickel and forging. Forging and annealing of the drawn wire were not considered, due to a lack of information, and the melting energy was assumed to be the same as the one used for powder making. The energy and gas consumption for producing 1 kg of powder and wire are shown in Table 6.

Table 6: Energy and gas consumption for producing 1 kg of nickel wire and powder (Ref 8)

Powder atomization (EIGA from ALD)		
Vaccum melting		
Electrical energy use (induction, 30% loss)	0.38	kWh
Gas atomization		
Argon (55 bar, 70% recycled)	3.73	kg
Wire drawing process (SAIKAWA DS60)		
Vacuum melting		
Electrical energy used (induction, 30% loss)	0.38	kWh
[illegible]		
Electrical energy used by motors	0.34	kWh

The cold spray system was assumed to use decompressed liquid nitrogen. The plasma and HVOF processes used argon, hydrogen, and oxygen bottled gas, respectively. For wire arc spraying, only the energy used to compress the gas was considered. The inventory for the thermal spray processes is shown in Table 7.

Table 7: Thermal spraying inventory

	Wire arc	Cold spray	Plasma	HVOF	Unit
Electricity (heating)	0.97	4.91	21.8	/	kWh
Electricity (cooling system)	/	/			kWh
Electricity (used to compress gas)	1.56 (30% loss)	0.93	/	/	kWh
Nickel	1.62	1.67	1.59	1.47	kg
Air	27.54	/	/	/	m^3
Argon	/	/	4.13	/	kg
H_2	/	/	0.035	/	kg
N_2	/	35.4	/	/	kg
O_2	/	/	/	23.92	kg
Kerosene grade 1	/	/	/	5.33	kg
Spraying duration	21.61	27.8	39.6	21	min
Emission to air					
Ni particulates	0.0049	0	0.0016	8.10^{-04}	kg
Disposal					
Ni in filter	0.4851	0	0.1584	0.074	kg
Ni solid waste	0	0.33	0.16	0.075	kg

Emissions to air: Thermal spray processes result in air emissions of particulates. Nickel fumes can be hazardous for human health and may cause cancer and other diseases. The emission ratio varies among thermal spray process. The cold spray nickel emissions were assumed to be negligible because of the low spraying temperature. For wire arc spraying, 30 % of the sprayed material was assumed to evaporate during the deposition process while for HVOF and plasma spraying the non-deposited material was considered to be 50% emission to air and 50% dust collected on the floor. The HEPA filter is 99% efficient. So the nickel final emission to air was assumed to be only 1% of the thermal spray emissions. The nickel emissions are summarized in Table 7.

Results and Interpretation

The SimaPro 7.1.7 LCA program (Ref 10) and a peer-reviewed database of upstream materials and energy (EcoInvent v2.0 (Ref 11)) were used to conduct the upstream analysis of materials and processes. A standard impact assessment methodology, called Eco-Indicator 99 (Ref 12), was used to convert the results into impact scores to the categories of human health, ecosystems and resource depletion, as discussed below.

Damage to Human Health was expressed in terms of Disability Adjusted Life Years (DALYs). Eco-Indicator 99 (EI99) includes models that link emissions to fate and transport, increase in concentrations and human exposure. Damage models link exposures to DALYs based on expected death and disability.

Damage to Ecosystem Quality was expressed as the percentage of species that may disappeared in a certain area due to the environmental load (potentially disappeared fraction (pdf) x m^2 x yr).

Damage to resources was measured in terms of the excess energy (MJ surplus) that will be required to extract future fossil and mineral resources based on the consumption quantifies in the analysis (Ref 12).

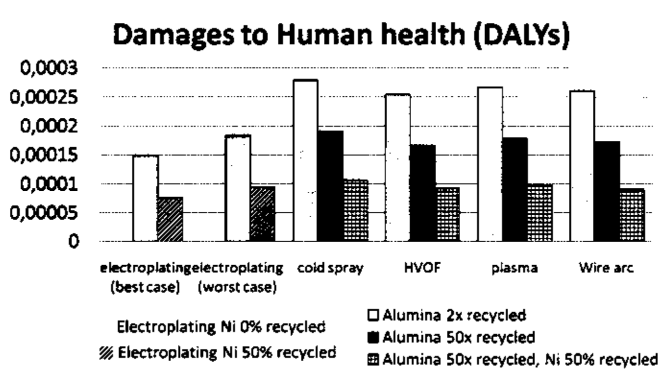

Figure 3a : Comparing damages to human health with different parameters

Comparison Between Processes

The first thing that can be observed, while comparing damages to human health and resources is that thermal spraying processes have a similar impact, while electroplating exhibits a lower impact when alumina for grit blasting is recycled two times (Figs. 3a and c). These results show that the nickel production process and also the alumina production for grit blasting have the largest impact, in comparison to energy production and other processes used during the coating step itself. Indeed, by increasing the recycling rate of grit blasting alumina (black bars of Figs. 3a and c) and then using 50% recycled nickel (cross ruled bars of Figs. 3a and 3c), one notices an impact reduction of more than 60%.

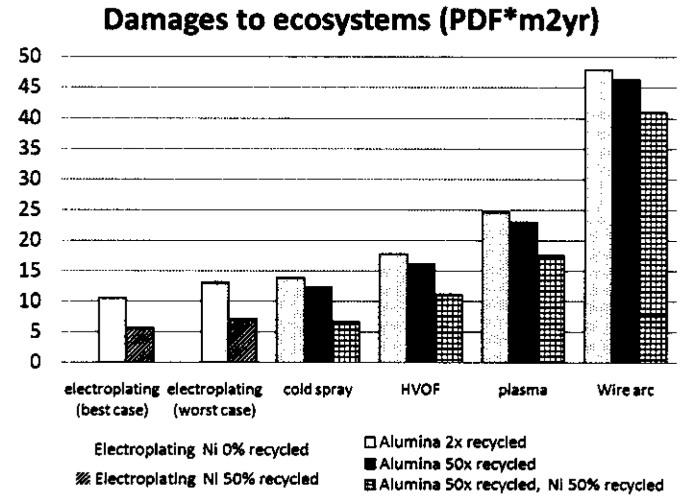

Figure 3b : Comparing damages to ecosystems with different parameters

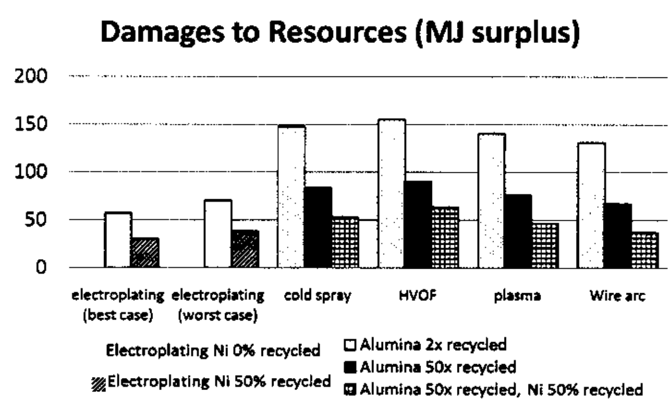

Figure 3c : Comparing damages to resources with different parameters

It is noted that when using 50% recycled nickel for all deposition processes and 50 x recycled alumina for grit blasting, the impacts on the human health and resource categories are approximately the same for nickel electroplating as for thermal spraying.

The impact to resources is partially due to the nickel and alumina use and partially to the use of fuels and chemicals, depending on the process. For HVOF spraying, the use of kerosene and liquid oxygen is responsible for 1/3 of the impact, for plasma spraying, electricity consumption is 1/7 of the total impact and for cold spraying, nitrogen and electricity are 1/5 of the impact.

Regarding the electroplating process, it can be seen (Fig. 4) that the use of nickel contributes to over 95% of the total environmental impacts. Only 4% of the impact on ecosystem is due to the electroplating process itself (more precisely the emissions of nickel to air and water) and only 5% of the impact on resources is due to electricity consumption.

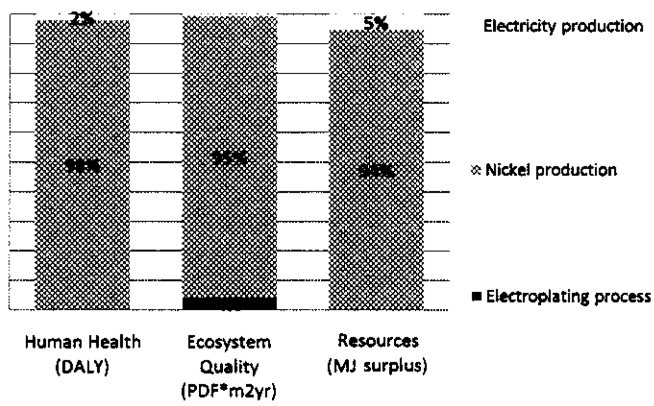

Figure 4 : Electroplating, contribution of processes to environmental impacts (worst case, Ni 0% recycled)

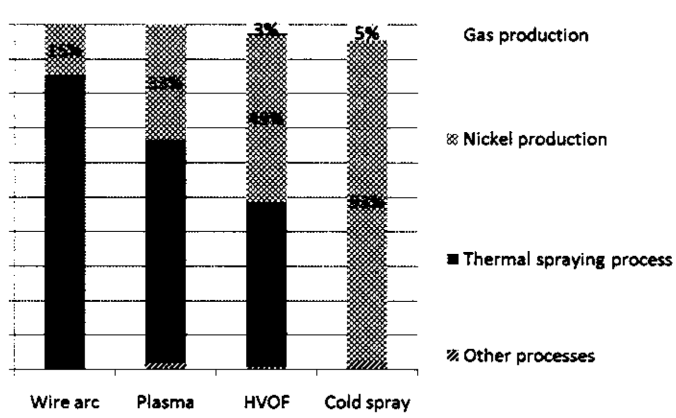

Figure 5 : Impact on Ecosystem : contribution of processes for thermal spraying (Nickel 50% recycled, alumina 50x recycled)

For thermal spraying, the damage to ecosystems linked to the amount of alumina used is relatively low in comparison to nickel emissions to air (Fig. 3b). Indeed, the impact on ecosystems grows significantly with the vaporization rate experienced during spraying (Fig. 5). Cold spraying seems to have a lower impact on environment, because no nickel emission is related to this process. In contrast, wire arc spraying is much more polluting, because of the high vaporization rate, while plasma and HVOF spraying are somewhat less polluting.

Comparing impact assessment methods

Establishing a linkage between the product or process and its potential environmental impacts is the most delicate stage of LCA since it requires interpretation of the data and value judgments to be made. Several standard impact assessment methods make it possible to evaluate the potential environmental impacts associated with the inputs and releases identified in the inventory. They are based on a general approach that involves at least four steps: (i) identification of the relevant environmental impact categories, (ii) assignation of all the inventory data to the impact categories, (iii) modelling of inventory data impacts within impact categories using science-based conversion factors, and (iv) expression of potential impacts in ways that can be compared.

Theoretically, the choice of an impact assessment method should be done considering its compatibility with the database used (Ref 13), the impact categories that are relevant for the study (for example climate change should be used to assess CO_2 air emissions) and also the addressed audience (Ref 10).

In the first part of this study, impact assessment was made by using a well established method: Eco-Indicator 99 that sorts the impacts in three categories (Human health, Ecosystems and Resources) (Fig. 6). In this section, the Eco-indicator 99 results are compared to the results obtained with the IMPACT 2002+ methodology, a recent method adapted from the Eco-indicator 99 and CML 2002 methodologies. According to the definition study of the SETAC/UNEP Life Cycle Initiative, this methodology uses the advantages of two different approaches: the CML 2002 methodology, a classical impact assessment method that restricts quantitative modelling to the early stages in the cause-effect chain in order to limit uncertainties, and the Eco-indicator 99 methodology, a damage-oriented method that tries to model the cause-effect chain up to damage, with high uncertainties (Ref 14). Consequently, the IMPACT 2002+ damage categories differ from Eco-indicator 99 categories (Figs. 6 and 7).

The potential damages to human health, ecosystems and resources, obtained by these two methods will now be compared for the four thermal spraying processes and electroplating. It was assumed that the plating process used "virgin" (non-recycled) nickel and that grit blasting used 50% reused alumina. The IMPACT 2002+ results for damages to human health are 7 to 10% lower than those estimated by means of Eco-indicator 99. However, both methods show the same trend for this impact category and, thus, lead to the same conclusions (Fig. 8). The difference of a few percent points may be explained by the fact that climate change is not considered to cause direct damage to human health in the IMPACT 2002+ methodology which considers that the impact of climate change on ecosystem quality and human health is not accurate enough to be taken it into account in any category (Fig. 7), (Ref 14).

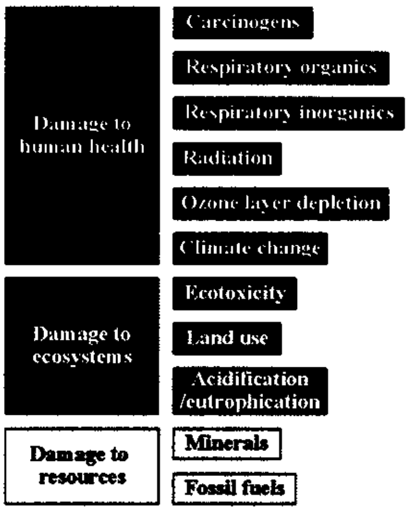

Figure 6 : Detailed damage categoriesin the Eco-indicator 99 methodology

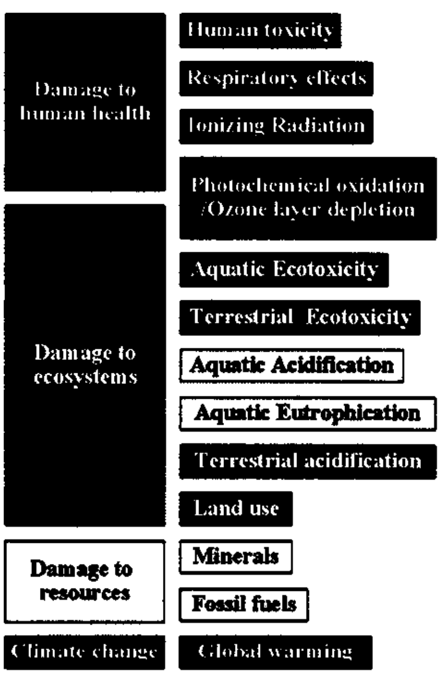

Figure 7 : Detailed damage categoriesin the IMPACT2002+ methodology

Interpreting the comparison between these two methods for the damage to ecosystems category is by far more difficult. Indeed, the differences cannot be clearly established by comparing the grouping schemes at Figures 6 and 7. The results obtained with IMPACT 2002+ are up to five times higher than those obtained with Eco-indicator 99 (Fig. 9). So, understanding the significant differences in the interpretation of damages to ecosystems requires having a closer look at the substances taken into account in impact assessment by both methods. For example, terrestrial acidification includes the same substances for both methods but this is not the case for land use and ecotoxicity. For land use, about 70 substances are taken into account with Eco-indicator 99 and only around ten with Impact 2002+. The main difference is observed with ecotoxicity that includes around 150 substances with IMPACT 2002+ and only 50 with Eco-indicator 99. These remarks are summarized in Table 8.

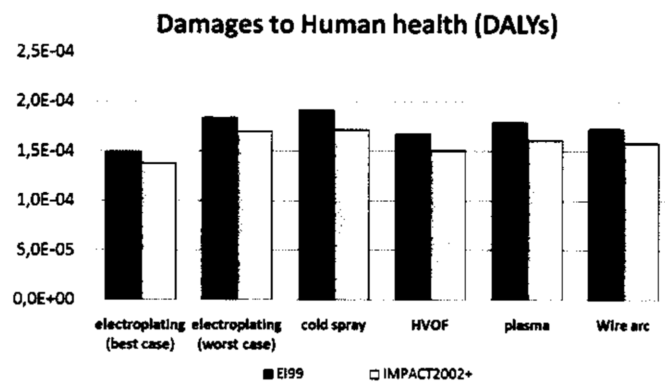

Figure 8 : Comparison of damages to human health according to Eco-Indicator 99 and IMPACT 2002+

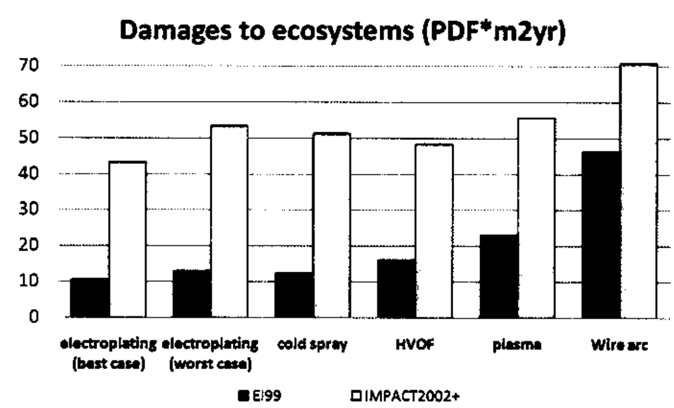

Figure 9 : Comparison of damages to ecosystemsaccording to E-I99 and IMPACT 2002+

It was found that in the E-I99 method the thermal spraying processes have a higher potential impact on ecosystems than electroplating, whereas IMPACT 2002+ results in less obvious results (Fig. 9). Indeed, while wire-arc spraying seems to be still the most potential impacting process, it is only 1.27 times more impacting than plasma, with IMPACT 2002+, and 2 times more, with E-I99. Also, the impacts of cold spraying and HVOF spraying are reversed with IMPACT 2002+ and E-I99 methods. Furthermore, it becomes difficult to draw clear conclusions about the comparison of electroplating and thermal spray processes, as plasma spraying exhibits a potential impact similar to the worse case of electroplating.

Table 8: Number of substances included in damage to ecosystems subcategories in the assessment methods used in this study

	Number of substances	
	Eco-indicator 99(E) V2.05	Impact 2002+ V2.04
Terrestrial acidification	3	3
Land use	69	13
Ecotoxicity (aquatic)	45	151
Ecotoxicity (terrestrial)		149

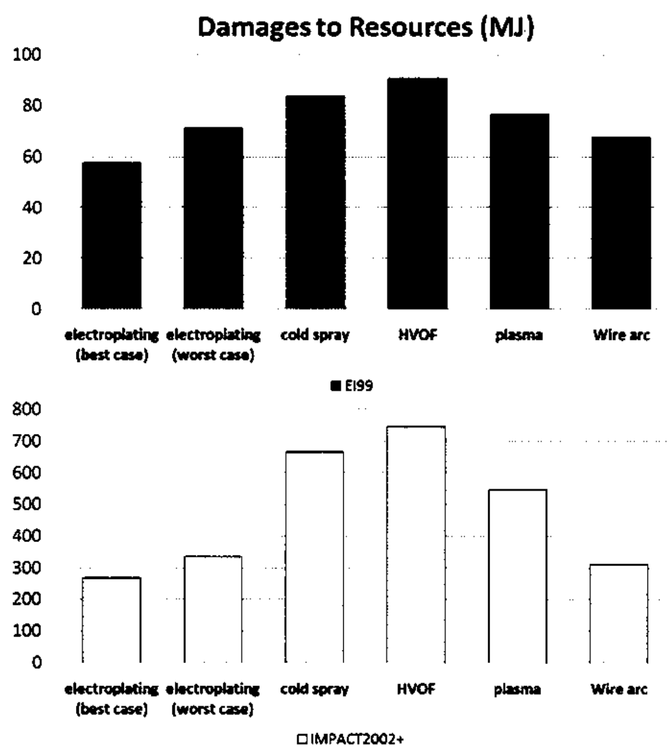

Figure 10 : Comparison of damages to resources according to E-I99 and IMPACT 2002+

The "damages to resources" category are considered in different ways in IMPACT 2002+ and EI99 methods. Both use the "fossil fuel use" subcategory but it is counted in "MJ surplus", in E-I99,and "MJ primary", in IMPACT 2002+. "MJ primary" represents the energy content of fossil fuels whereas "MJ surplus" is the surplus energy for the future mining of resources. This explains why IMPACT 2002+ yields much higher potential impacts (Fig. 10). On the basis of this method, cold spraying, HVOF and plasma spraying of nickel have about twice the impact on resources than electroplating and wire arc spraying. These results can be explained by the huge gas consumption in HVOF and cold spraying (Fig. 11). In the case of plasma spraying, damages to resources are increased by the extraction of uranium caused by electricity consumption as a European mix for energy (39% nuclear) has been assumed in this study.

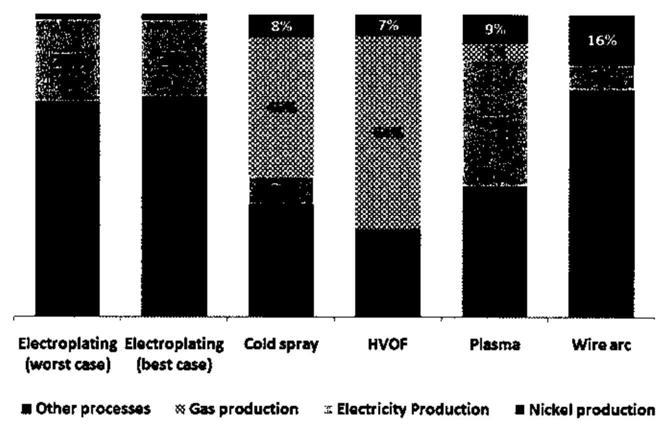

Figure 11 : Damages to resource:. Comparison of the various processes of the study with the IMPACT 2002+method

Conclusion

Life Cycle Analysis is a powerful tool for determining the potential environmental impacts of surface coating processes but it requires accurate data of process parameters and releases that are often difficult to obtain; also, it relies on many assumptions as to manufacturing of the materials (powder, wire, chemicals, etc.) used in the operation of the process. The operation of the processes and In addition, the impact assessment stage that seeks to establish a linkage between the coating process and potential impacts requires the interpretation of the data and value judgments to be made.

In this study, the LCA framework has been used to compare the potential environmental footprints of various thermal spray processes (cold spray, HVOF, arc wire and plasma) and electroplating for nickel coating deposition. Two authoritative assessment methods, Eco-Indicators99 and Impact 2002+ were used to estimate the potential environmental impacts of the various processes.

The analysis showed that comparing the environmental impacts of the surface coating processes is not an easy task, because of the number of parameters that can affect the final result. For example, the deposition efficiency for all the processes and the recycling rate of blasting media, for the thermal spray deposition processes, can vary considerably and have a large effect on the environmental impacts of nickel plating processes. The electroplating process can also be highly pollutant in countries that do not apply stringent environmental regulations on air and water emissions. Moreover, to be more realistic, the environmental impact modelling of nickel electroplating processes should incorporate the environmental impacts of the production of the chemicals used in this process.

The study also showed that the translation of inventory data into potential impacts on the environment is a delicate issue. The two impact assessment methods used in this work reached different and sometimes inconsistent conclusions. However, the results indicate that:
- thermal spraying processes and electroplating have a similar potential impact on human health for nickel deposition,
- potential damage to ecosystems is about the same for electroplating , HVOF and cold spraying while it is higher for plasma and wire-arc spraying.
- wire-arc and electroplating have a much lower impact on resources in comparison with HVOF, plasma and cold spraying.

With time, as more companies start to publish the information derived in internal LCA studies, it will become easier to make more detailed comparative analyses of the environmental impacts of different processes.

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