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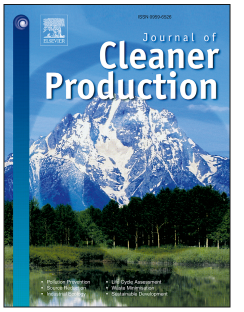


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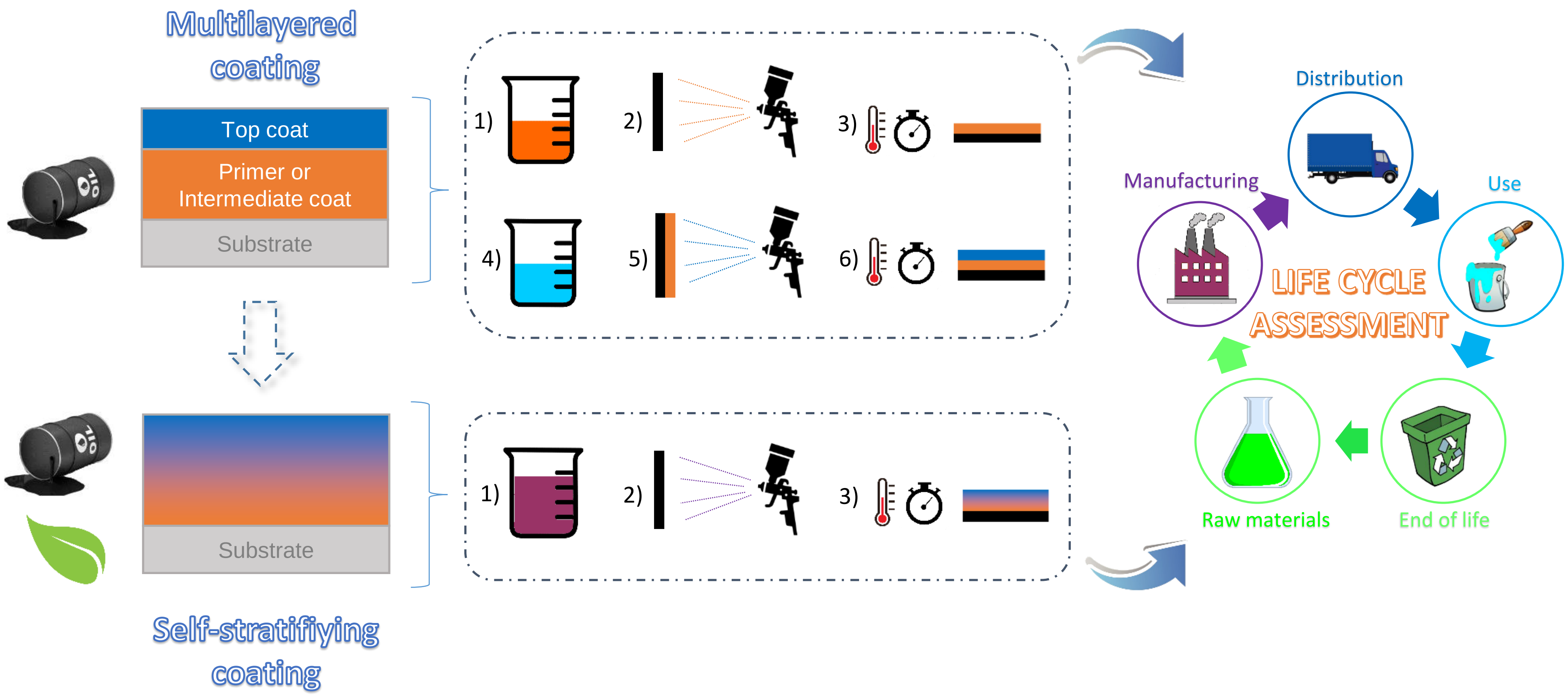
Multilayered coating
Top coat
Primer or Intermediate coat
Substrate
1)
2)
3)
4)
5)
6)
Substrate
1)
2)
3)
Self-stratifiying coating
Distribution
Manufacturing
Use
LIFE CYCLE ASSESSMENT
Raw materials
End of life

# Life Cycle Assessment of multi-step versus one-step coating processes using oil or bio-based resins

**Charlotte Lemesle[1,2], Jérôme Fremiot[1], Agnès Beaugendre[1], Mathilde. Casetta[1], Séverine Bellayer[1], Sophie Duquesne[1], Anne-Sophie Schuller[2], Maude Jimenez[1*]**

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**Abstract**

Nowadays, before setting up an industrial process, special attention has to be paid to its environmental footprint. This new way of thinking allows evaluating the hotspots so as to propose solutions to conceive more eco-friendly processes. Thus, the coating industry is increasingly preoccupied by the environmental impacts of newly designed paints. That is why innovation tries to take into consideration both the formulation composition and the application and drying methods. Following a Life Cycle Assessment (LCA) approach, the present study aims to compare the environmental impacts of self-stratifying coatings (with three process steps), either oil-based or bio-based, to those of an oil-based multilayered coating (with six process steps). The concept of the self-stratifying coating is to bring the primer, the intermediate and the top coat properties together in a one-pot formulation to produce a multi-functional coating through a less laborious process.

The total environmental impact of each process can be characterized by four main categories of impacts: Climate Change Human Health (CCHH), Human Toxicity (HT), Climate Change Ecosystem (CCE) and Fossil Depletion (FD), which together represent approximately 86% of the global impact for each system. The results obtained show that the self-stratifying oil-based coating process is more energy efficient compared to the currently used multilayered oil-based coating process, with a 15% decrease of CCHH, HT and CCE indicators and a 14% decrease of FD indicator, corresponding to a 13.6% decrease of the total environmental impact. However, as this decrease was lower than expected, particular attention was then paid to the chemicals used (resins, solvents) and to the process conditions. The substitution of the oil-based epoxy resin by a bio-based one and the use of less harmful solvents allow reducing the total environmental impact by 32.4% compared to the multilayered oil-based system (with 30% decrease of CCHH indicator, 50% of HT

indicator, 31% of CCE indicator and 34% of FD indicator). In a global way, electricity consumption control and the nature of chemicals used were reported to contribute in a significant way to the improvement of the process environmental impact.

**Keywords:** Life Cycle Assessment, self-stratification, coating, fire retardant, bio-based, environmental impact.

## 1. Introduction

The design of innovative coatings with multifunctional properties is a real challenge. Generally, a functional coating requires several layers with particular properties: a first layer to enhance the adhesion between the paint and the substrate, a middle layer with specific property (e.g. with flame retardant properties) and a protective topcoat to prevent the ageing and to protect the coating against external aggressions (scratches, humidity, UV rays...) (Benjamin et al., 1996). From an industrial point of view, these multilayered systems require complex formulation, application, and curing steps, implying many constraints (Verkholantsev, 1985). In order to comply with increasingly demanding environmental constraints, the new concept of self-stratification has been developed (Beaugendre et al., 2017a). A self-stratifying coating is based on a blend of polymers dissolved in a common solvent to produce a coating requiring only one step of: formulation, application, and curing (Verkholantsev and Flavian, 1996). This efficient and economical concept allows reducing the number of steps to coat a substrate, while providing a coating with equivalent or better performances than the common multilayer process (Figure 1). Reduction of energy, pollution and waste generation is therefore induced by this process (Baghdachi et al., 2015).

Furthermore, an increase in stringent regulations on toxicological and environmental aspects now favors the use of bio-based products. Fossil fuel reserve is expected to deplete around the end of the 21st century if the present consumption rate continues. For this reason, replacement of petroleum-based polymeric materials in paint by chemicals of natural and renewable origin has gained growing interest from industries. New perspectives for bio-based chemicals and polymers have been driven by the recent rise in oil prices and consumer expectations for environmentally friendly products (De Jong et al., 2012). In 2017, the total production volume of bio-based polymers reached 7.5 million tons in the world (Nova Institute, 2019). The coating industry, one of the main polymer consumers, has been able to integrate these bio-based polymers into paint formulations as a binder (Derksen et al., 1996). These bio-polymers are made from different renewable resources as vegetable oils: sunflower, soybean and linseed oil (Sharmin et al., 2015) (Athawale and Nimbalkar, 2011), cardanol and sugar (Kumar et al., 2018) among others (Meier et al., 2007).

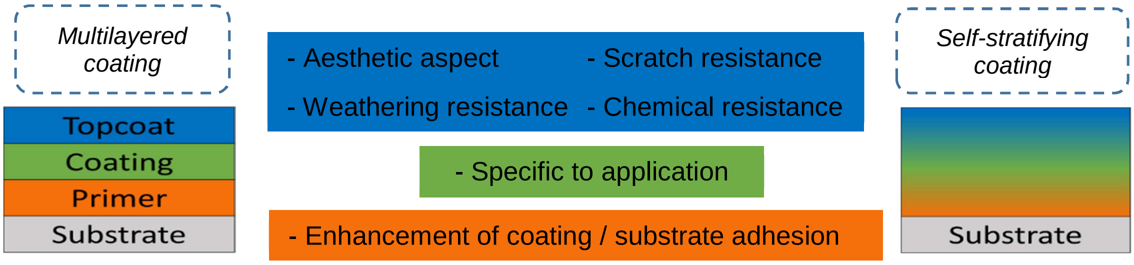



Figure 1: Self-stratifying concept

In order to quantify the so-called benefits on the environment of the self-stratifying process and the use of bio-based polymers, a life cycle assessment was performed on three kinds of coatings showing equivalent fire retardant (FR) properties. It was in the 1960's that the Life Cycle Assessment (LCA) was born. Scientists affected by the fast depletion of fossil fuel devised a method to assess the impact of energy consumption on the environment. At the beginning of the 1970's, energy and raw materials were factors mainly considerate for LCA calculation. Later other parameters were also considered, such as air and water emission and solid wastes. Even later, in the 1980's, hazardous waste management became the primary environmental concern (Svoboda, 1995). Nowadays, LCA is a commonly used and successful method to investigate and quantify environmental impacts of a product or service throughout its whole life cycle (Heijungs et al., 1992).

Indeed, several LCAs in the coating field have been conducted, however dealing with other applications than fire protection. For example, the environmental impact of antifouling coatings in marine (Rossini et al., 2019) and food engineering (Zouaghi et al., 2019) was investigated. Both reports demonstrate that the production step of antifouling coating significantly affects the environmental impact of the system due to the chemicals used. The positive environmental impact resulting from the use of antimicrobial coatings for packaged fresh milk was also reported (Manfredi et al., 2015). The results show how the reduction of milk waste achievable through the use of a coating generates environmental benefits and demonstrates the importance of including food waste in the LCA of packaging systems. Another paper (Babaizadeh and Hassan, 2013) reports that the application of a nano-sized titanium dioxide coating on residential window glass has a positive effect on several indicators (acidification potential, eutrophication potential, criteria air pollutants and smog formation potential) but also damages some of them (global warming, fossil fuel depletion, water intake, human health, and ecological toxicity).

One last example is the study of the environmental impact of the production and construction of reflective coatings for cool pavements to reduce the contribution of the formation of urban heat islands in urban areas (Li et al., 2016). It reports that the environmental impact could be reduced by applying these coatings instead of traditional urban pavement, but it also depends on their durability: solvent-based coatings have longer lifetime but can emit Volatile Organic Compounds into the atmosphere, with a negative impact on both the environment and human health and water-based coatings have relatively shorter lifetime, but with less impact on the environment.

The objective of the present study is to enhance the environmental impact of innovative coating processes. In this paper, different processes to coat a polymeric substrate with a flame retardant coating were compared using LCA: (i) the classical multilayered approach (REF), (ii) a "one pot" self-stratifying approach using a silicone resin and an oil-based epoxy resin (SSO) (Beaugendre et al., 2017b) and (iii) a "one pot" self-stratifying approach using a silicone resin, a bio-based epoxy resin and less toxic solvents (SSB). As far as we know, no paper compares a multilayer versus a self-stratifying coating process in term of environmental impact. However, Montazeri et al. and Gustafsson et al. already reported, through a "from cradle to grave" LCA the positive environmental impact of bio-based ingredients in coating formulations (Montazeri and Eckelman, 2018) (Gustafsson and Börjesson, 2007).

## 2. Experimental methods

Life Cycle Assessment is a tool used for eco-design standardized by ISO 14040 (2006) (ISO (International Organization for Standardization), 2006) and ISO 14044 (2006) (ISO (International Organization for Standardization, 2006). In this paper, a comparative life cycle assessment is performed on three flame retarded coatings applied on a polycarbonate (PC) substrate.

All processes are carried out in the North of France, in the city of Villeneuve d'Ascq. Moreover, this assessment takes into account the impact created by inputs and outputs from extraction step to the waste treatment step.

### 2.1. Functional unit

A functional unit (FU) aims at providing a basis to compare different systems in a fair and valid manner (Wolf et al., 2010). LCA was performed on the three lab scale coating processes (REF, SSO and SSB). The FU chosen was: "Deposit on a 100 cm² Polycarbonate plate of a FR coating allowing reaching a V0 rating at UL94".

### 2.2. Description of the studied systems

The first two systems (REF and SSO) are oil-based, so the chemicals used to formulate coatings are the same. The third system (SSB) is bio-based, explaining why the epoxy resin and the solvents are different from those used in the two other systems.

In both coatings red iron oxide (Grolman, Cathaycoat Red RA11A, Brno, Czech Republic, diameter 0.3 µm) was incorporating as flame retardant additive at 10% PVC (Pigment Volume Concentration, i.e. the volume percentage of solid particles in the system after film formation). It was mixed with the epoxy resin at 1200 rpm. The substrate considered for the study is polycarbonate plate (Lexan, Polydis, Ligny Le Chatel, France, 3 mm thickness).

#### 2.2.1. Oil-based systems

To design these systems, an epoxy resin (Bisphenol A epoxide from Sigma-Aldrich, Midland, EEW 172-176) and a silicone resin (Dowsil RSN-0217, Dow Corning, Seneffe, Belgium) were selected. A polyamine DETA (Diethylene triamine (99%), Sigma-Aldrich, St. Louis, MO) was selected as curing agent. The resins were dissolved in a blend of m-xylene (99%) and butylacetate (BuAc, ≥ 99.5%), from Sigma-Aldrich (Midland) (Beaugendre et al., 2017b).

##### 2.2.1.1. Reference multilayered oil-based coating system (REF)

Epoxy resin was dissolved at 30% wt./wt. in BuAc: xylene (1:1). Once iron oxide was mixed with the epoxy resin, the cross-linking agent was slowly incorporated. This system was applied on the substrate with a Devilbiss spray gun to reach a 100 µm wet thickness. It was then cured during 1 day at 22°C and 2 hours at 110°C, using two different ovens.
Afterwards, the silicone resin was prepared in the same conditions: at 30% wt./wt. in a blend of BuAc: xylene (1:1). This system was sprayed (100 µm wet thickness) on the previously applied epoxy layer, using the same spray gun. It was then cured for one day at 22°C and two hours at 110°C, using two different ovens. The pre-curing step has been set up to avoid a too brutal evaporation of the solvents from the paint film. The curing step is carried out at 110°C because at this temperature the epoxy resin reacts with the hardener to form a 3D network in order to form a durable film. These drying parameters were defined in a previous study but they could be optimized (Beaugendre et al., 2017b). Therefore, the REF system process is divided into six steps. All inputs, outputs, equipment and process conditions are detailed in Figure 2.

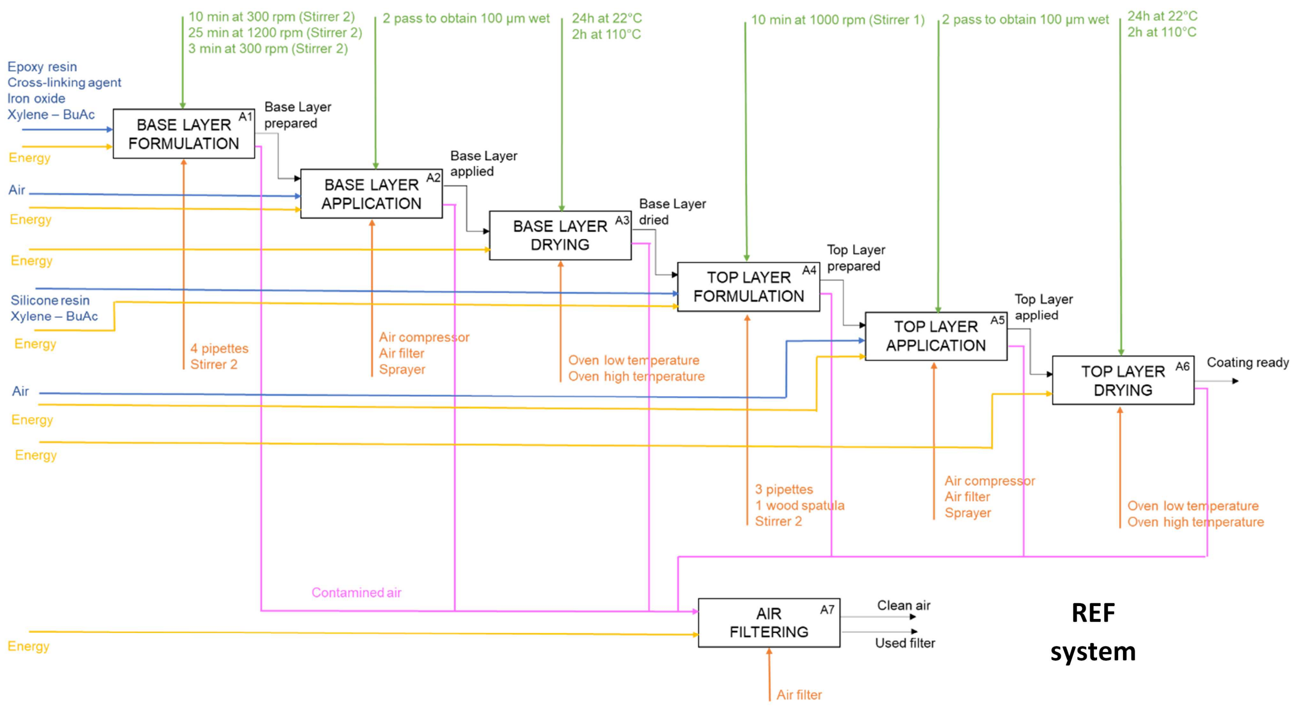



Figure 2: Structured Analysis and Design Technics (SADT) diagram for the multilayered (REF) coating process

#### 2.2.1.2. Self-stratifying oil-based coating system (SSO)

Each resin was dissolved at 30% wt./wt. in BuAc: xylene (1:1). Iron oxide was mixed in the epoxy resin. Then, both resins were mixed together (1:1 wt./wt.) for 10 minutes at 400 rpm. The cross-linker was slowly incorporated into the mixture and stirred during 3 minutes. The formulation was applied (200 µm wet thickness) on polycarbonate with the same spray gun. The curing process was the same as previously described. SSO system only requires three processing steps summarized in Figure 3.

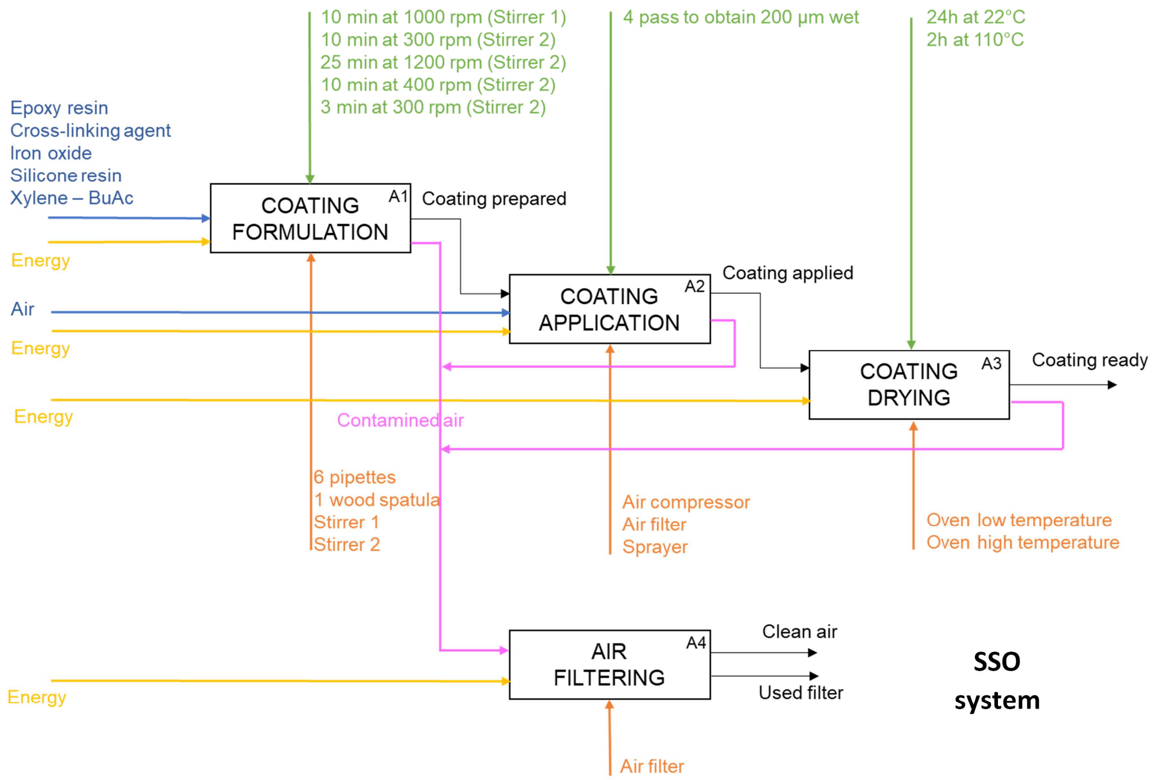



Figure 3: SADT diagram for the self-stratifying oil-based coating process

### 2.2.2. Self-stratifying bio-based coating system (SSB)

In that case, a lab scale epoxy resin (Epoxy A, reaction product of isosorbide and epichlorohydrin with 70% bio-content, equivalent weight: 176), a silicone resin (Dowsil S217, Dow Corning, Seneffe, Belgium) and a diamine (Diethylene triamine (99%), Sigma-Aldrich, Midland) as curing agent were selected. Ethyl lactate (99%) and butyl acetate (99.5%) purchased from Sigma-Aldrich (Midland) were chosen as solvents.

The selected resins were separately dissolved at 30% wt./wt. in a blend of BuAc: Ethyl lactate (10:90). Iron oxide was dispersed in the epoxy medium. Pigmented epoxy resin and silicone resin were then mixed together and stirred at a 1:1 ratio by weight (epoxy: silicone). The hardener was added dropwise in the polymer blend and mixed for 5 minutes before coating application in the same conditions as SSO system. The coating was dried for 15 minutes at room temperature (~20°C), and cured for 1 hour at 80°C in an oven. The pre-curing stage of 15 minutes mimics industrial conditions. Indeed, in the coating industry, there is often a small delay between the application of the coating and its curing. Moreover, the drying step is carried out at 80°C for the same reasons as with the epoxy resin used for REF and SSO systems. 80°C is the temperature at which the bio-based epoxy resin and the amine reaction takes place and where the conversion rate is high (>80%).

The three main processing steps of SSB system are visible on Figure 4.

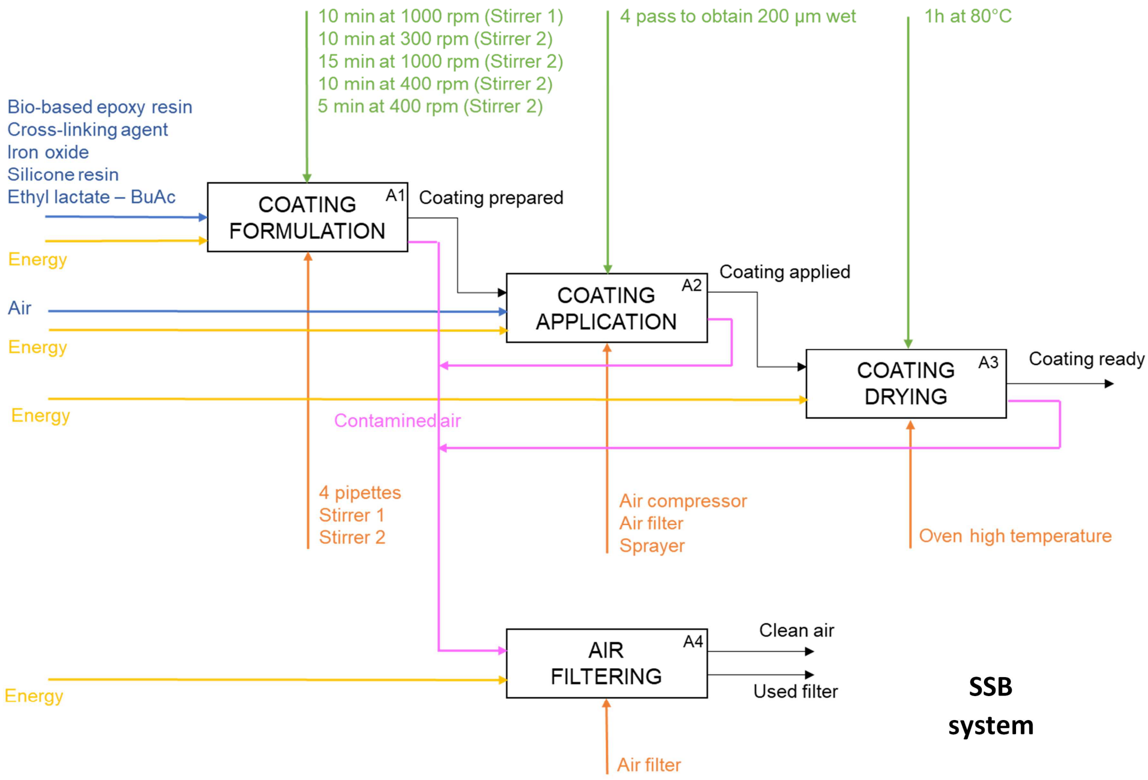



Figure 4: SADT diagram for the self-stratifying bio-based coating process

### 2.2.3. Coatings properties

The cross-section of each dry coating films were then analysed by Scanning Electron Microscopy with X-Ray analysis (SEM-EDX). In the reference system (not shown here), two distinct layers are obtained, with epoxy as bottom layer and silicone as upper layer. In the SSO coating, the silicone resin migrates toward the top to form the upper layer, similarly as iron oxide and the base layer is composed by the epoxy resin (Figure 5). Observations are the same for the SSB coating (Figure 6).

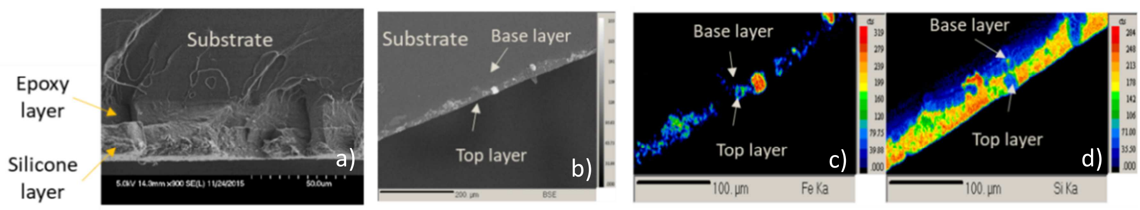


Figure 5: SEM micro micrograph of a cross-section of the SSO system (a, b) with EDX mapping of Iron (c) and Silicon (d).

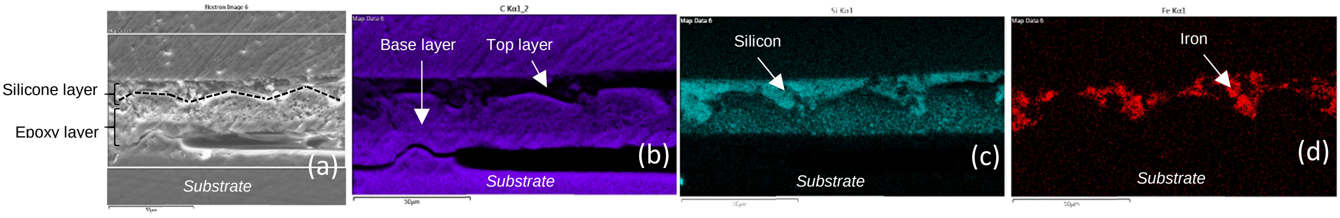
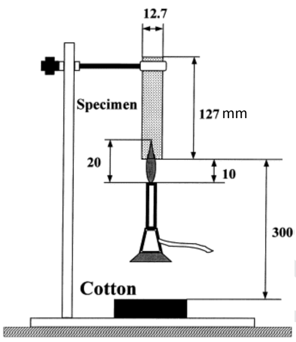



Figure 6: (a) SEM picture of SSB cross section coating with X-ray mappings of (b) Carbon, (c) Silicon and (d) Iron showing iron oxide in the silicone top layer.

The samples (100*10*3 $mm^3$) were then submitted to the fire test UL-94 (Figure 7) combustion rate and time and production of flaming drop (with ignition of a cotton sample) are evaluated to qualify the sample resistance to the action of a flame according to a ranking from: non-classified (NC) to V0 (the best rating which corresponds to short burning times without ignition of cotton).




| **Criteria** | **V-0** | **V-1** | **V-2** |
|---|---|---|---|
| Total flaming combustion for each specimen | ≤ 10s | ≤ 30s | ≤ 30s |
| Flaming and glowing combustion for each specimen after second burner flame application | ≤ 30s | ≤ 60s | ≤ 60s |
| Total flaming combustion for all 5 specimen | ≤ 50s | ≤ 250s | ≤ 250s |
| Cotton ignited by flaming drops | NO | NO | YES |
| Glowing or flaming combustion of any specimen to holding clamp | NO | NO | NO |

Figure 7 : UL-94 device with UL-94 classification (according to IEC 60695-11-10)

All processes lead to similar fire properties (V0 at UL94 vertical fire test). Further details on the fire retardant properties of the SSO system are described in another paper from our team (Beaugendre et al., 2019). Other characterizations were conducted: Figure 8 summarizes the main characteristics of the two types of systems, and compares them in terms of adhesion at cross hatch test (standardized method ISO 2409 (International Organisation for Standardisation, 2013)) , visual aesthetic aspect, fire properties (UL-94 testing) and weathering resistance (humidity and UV accelerated ageing).

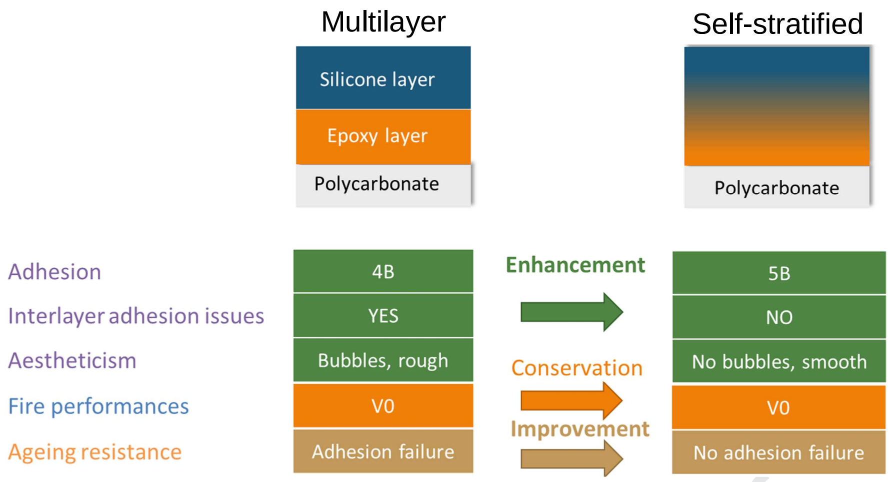



Figure 8: Comparison of the properties of multi-layered and self-stratified systems.

Although both systems are similar in terms of fire performance, the self-stratified system allows improving adhesion: a 5B adhesion (best rating) is obtained for the SS systems, compared to 4B for the REF system. The self-stratified system also provided a better aesthetic aspect and weathering resistance. The global performances of the SS systems thus appear more interesting than those of the REF system.

As REF, SSO and SSB coatings show equivalent flame retardant properties, this work aims at quantifying to what extent the SSO and SSB systems improve the environmental impact compared to REF system.

### 2.3. Life Cycle Inventories

To achieve a whole LCA, this study compiled Life Cycle Inventories (LCI) to quantify every input and output regarding their amount, origin and end of life scenarios.

#### 2.3.1. Hypotheses

In order to get closer to reality, many experimental data were collected. Furthermore, some literature data as well as hypotheses are inevitable to execute a complete LCA study. These hypotheses are detailed below.

##### 2.3.1.1. Chemicals

As some of the chemicals used are not listed in the Ecoinvent 3 database, some hypotheses were made on how the components were synthesized. For more than three compounds, one synthesis way among others was chosen, aiming at obtaining new intermediate products available in the database. Moreover, process parameters like heating time, temperature and stirring time have been taken into account when the information is released. They provide important guidance to be near the real environmental impact of the final product. The choice is certainly not the most relevant in terms of industrial constraint, but these syntheses are those with the most available background data. Nevertheless, simulations concerning transport and energy consumption for each compound are close to reality but remain uncertain.

#### 2.3.1.2. Electrical consumption

According to the device specifications from manufacturer's sheets, electrical consumptions were calculated with the average data of the French energy mix from the latest version of the Ecoinvent 3 database.

Carbon mix as German mix would increase the importance of electricity consumption, by contrast to the Norway mix based on hydroelectric power which would minimize it. Therefore the French energy mix is a good compromise.

#### 2.3.1.3. End of life

The end of life scenario was the following one: all the samples were considered to be used for energy recovery.

#### 2.3.1.4. Allocation and process hypothesis

In order to achieve an LCA as fair as possible, assumptions and allocations on the process have to be made: (i) for the coating preparation : the power required to stir the mixtures is not correlated with the agitated mass due to the very low viscosity of the coating formulation, (ii) for the coating application : the paint spray booth is used for 64 seconds (2 seconds per pass x 4 passes per plate x 8 plates), the energy taken into account corresponds to the consumption of the compressor and to the electricity required for the fan extraction during this working time, (iii) for the coating curing : the oven used can contain up to 24 plates, the heating is considered for the entire enclosure reduced to 8 plates and (iv) for all processing steps : retreatment of gaseous and/or liquid effluents was taken into account.

Nevertheless, the most important hypothesis formulated in this study concerns the wear of the equipment and devices used. Given the short operating time of the equipment and the lack of information on their lifetime, the wear of the devices will be overlooked. This approximation makes sense as only the process constraints change, but not the equipment used.

### 2.3.2. LCI example for SSO system

Life Cycle Inventory (LCI) is a methodology used to assess resource consumption and the quantities of waste flows and emissions caused by the product life cycle (Rebitzer et al., 2004). For each of the three systems, exhaustive lists of chemical components, consumables, energy consumed and transportation are established. The LCI of the SSB system is shown in Table 1 for 8 coating plates, allocations are made to refer to the functional unit for the quantification of environmental impacts.

Table 1: LCI of self-stratified bio-based coating system

| PROCESS | | | |
|---|---|---|---|
| **Materials** | **Quantity** | **Unit** | **Origin** |
| Silicone resin | 15 | g | Belgium |
| Butyl Acetate | 3.5 | g | USA |
| Ethyl Lactate | 31.5 | g | USA |
| Energy stirrer 2 | 1.5 | Wh | French mix |
| | | | |
| Epoxy resin | 15 | g | France |
| Butyl Acetate | 3.5 | g | USA |
| Ethyl Lactate | 15 | g | USA |
| Energy stirrer 1 | 99.294854 | Wh | French mix |
| Iron Oxide | 3.12 | g | Czech Republic |
| Energy stirrer1 | 54.604518 | Wh | French mix |
| | | | |
| Energy stirrer 1 | 57.374369 | Wh | French mix |
| | | | |
| DETA | 0.88 | g | USA |
| Energy stirrer 1 | 28.687185 | Wh | French mix |
| | | | |
| Acetone | 13.84 | g | France |
| | | | |
| Recycled paper sheet | 10.88 | g | France |
| Pipette | 4.8 | g | France |
| Energy pass paint gun | 291.936 | Wh | French mix |
| | | | |
| Energy drying high temperature | 7.71608 | Wh | French mix |
| | | | |
| Air treatment | 103.20257 | Wh | French mix |
| | | | |
| Coal filter for air treatment | 0.04076927 | g | France |

| END OF LIFE | | | |
|---|---|---|---|
| **Designation** | **Quantity** | **Unit** | **Origin** |
| Transport to end of life center<br>*one part burying other part burning for energy valorization | 0.03167 | ton/km | France |

| TRANSPORT | | | |
|---|---|---|---|
| **Material transported / Means of transport** | **Quantity** | **Unit** | **Origin** |
| Silicone | | | Belgium |
| *Truck* | 0.00444 | ton/km | |
| Epoxy bio-based | | | France |
| *Truck* | 0.00066 | ton/km | |
| Butyl Acetate | | | US |
| *Truck* | 0.002296 | ton/km | |
| *Train* | 0.00756 | ton/km | |
| *Sea ship* | 0.037051 | ton/km | |
| *Ethyl Lactate* | | | US |
| *Truck* | 0.020664 | ton/km | |
| *Train* | 0.06804 | ton/km | |
| *Sea Ship* | 0.037051 | ton/km | |
| DETA | | | US |
| *Truck* | 0.00028864 | ton/km | |
| *Train* | 0.06804 | ton/km | |
| *Sea ship* | 0.00465784 | ton/km | |
| Iron oxyde | | | Czech Republic |
| *Truck* | 0.000156 | ton/km | |
| *Train* | 0.0029016 | ton/km | |
| Acetone | | | France |
| *Truck* | 0.00025 | ton/km | |
| *Train* | 0.0022 | ton/km | |
| Consumable (Paper, Pipette) | | | France |
| *Truck* | 0.000844 | ton/km | |
| *Train* | 0.0074272 | ton/km | |

### 2.4. Method for LCA calculations

SimaPro 8.2.3 software allows the LCA calculations in combination with the Ecoinvent 3.4. database where the nominal environmental impacts were recovered. Recipe E 1.12 (Europe) was chosen as calculation method due to the relevance of its indicators (Goedkoop et al., 2008) and by the fact that this method calculates also the impacts for the European territory (except for categories with global impacts (climate change, ozone depletion and consumption of natural resources)). Moreover, this method is very efficient when electricity is an important impacting factor on the studied processes.

The results will be presented in midpoint and then analyzed in the discussion part.

## 3. Results and discussions

### 3.1. Impact categories selection

In order to focus on relevant data, generally accepted pollution indicators have been selected to estimate the consequences on the environment. Impact categories (IC) representing less than 0.5% of the total "whole process" score were excluded, leaving eight ICs to be discussed: Climate Change Human Health (CCHH), Human Toxicity (HT), Particulate Matter Formation (PMF), Climate Change Ecosystem (CCE), Agricultural Land Occupation (ALO), Natural Land Transformation (NLT), Metal Depletion (MD) and Fossil Depletion (FD). Each indicator can be associated with one or more damages as shown on Figure 9.

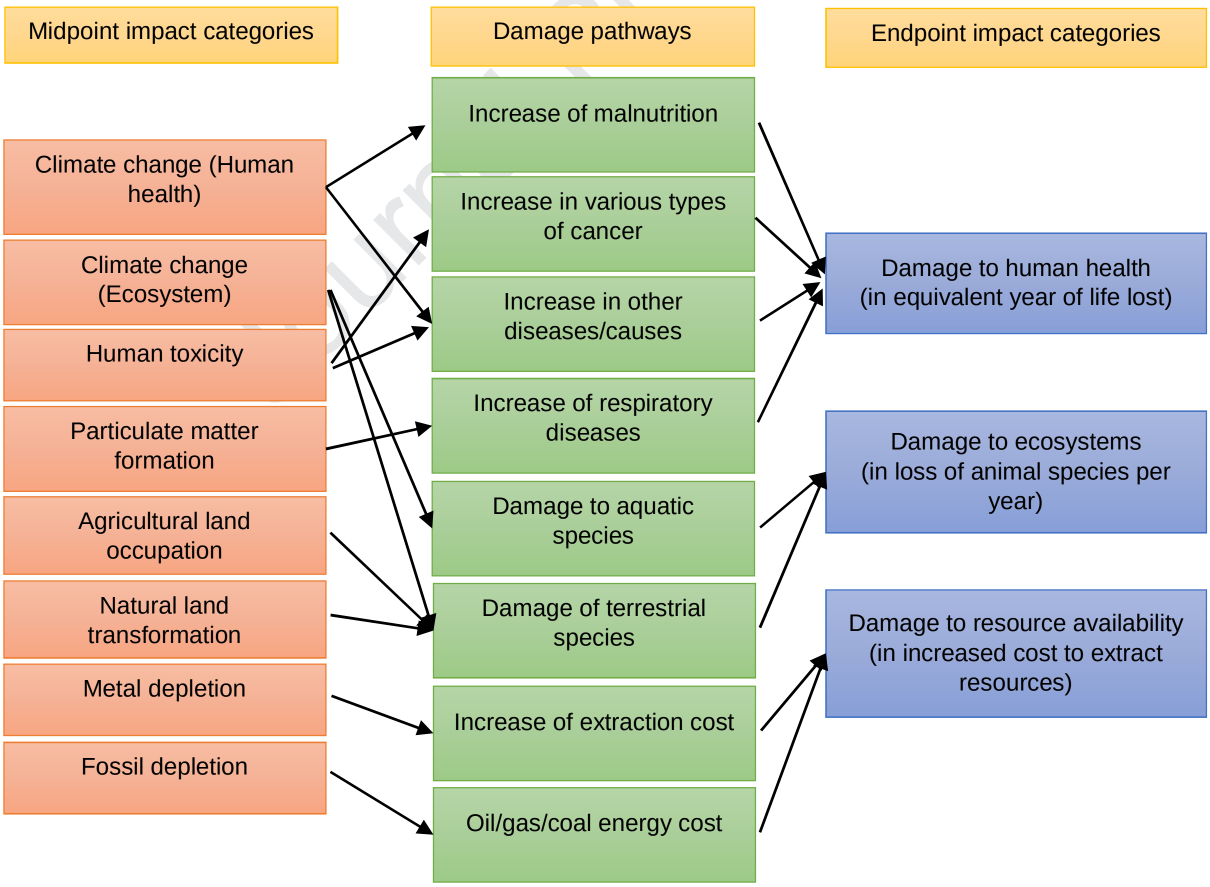



Figure 9: Midpoint impact categories and their consequences (Thériault and Reyes, 2011)

### 3.2. Life cycle impact assessment

LCA carried out on the REF system first determined its total environmental score, i.e. 18.2 mPts for one deposit on a 100 cm² Polycarbonate plate of a FR coating allowing reaching a V0 rating at UL94. Total environmental score of 15.7 mPts was obtained for the SSO system, corresponding to a 13.6% decrease compared to the REF system. For the SSB system, a total environmental score of 12.3 mPts was obtained, corresponding to a 32.4% decrease compared to the REF system.

Figure 10 compares the environmental impacts for major indicators of each system, extrapolated for 10 years use. First of all, it can be noticed that Metal Depletion (MD) is the only impact not significantly affected by the processes. Then, four impact categories out of the eight present a contribution higher than 0.5%, namely CCHH, HT, CCE and FD, which together represent approximately 86% of the global impact for each system. It should be noted that a reduction between 31% and 50% of the environmental score is visible for each of these four impacts between REF and SSB processes.

Each contributor to these main environmental impacts is graphically summarized in Figure 11. In the following part, each impact will be detailed precisely.

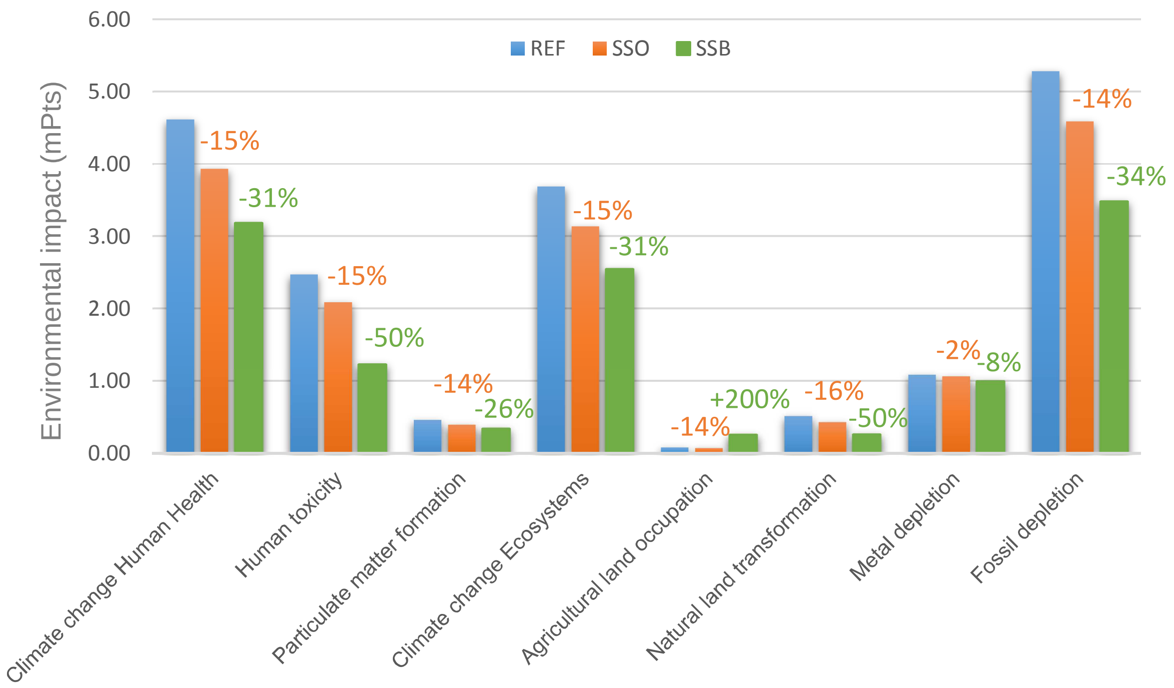



Figure 10: Environmental impacts (for 10 years) for major indicators of REF, SSO and SSB systems and Comparison of SSO and SSB systems with the REF one.

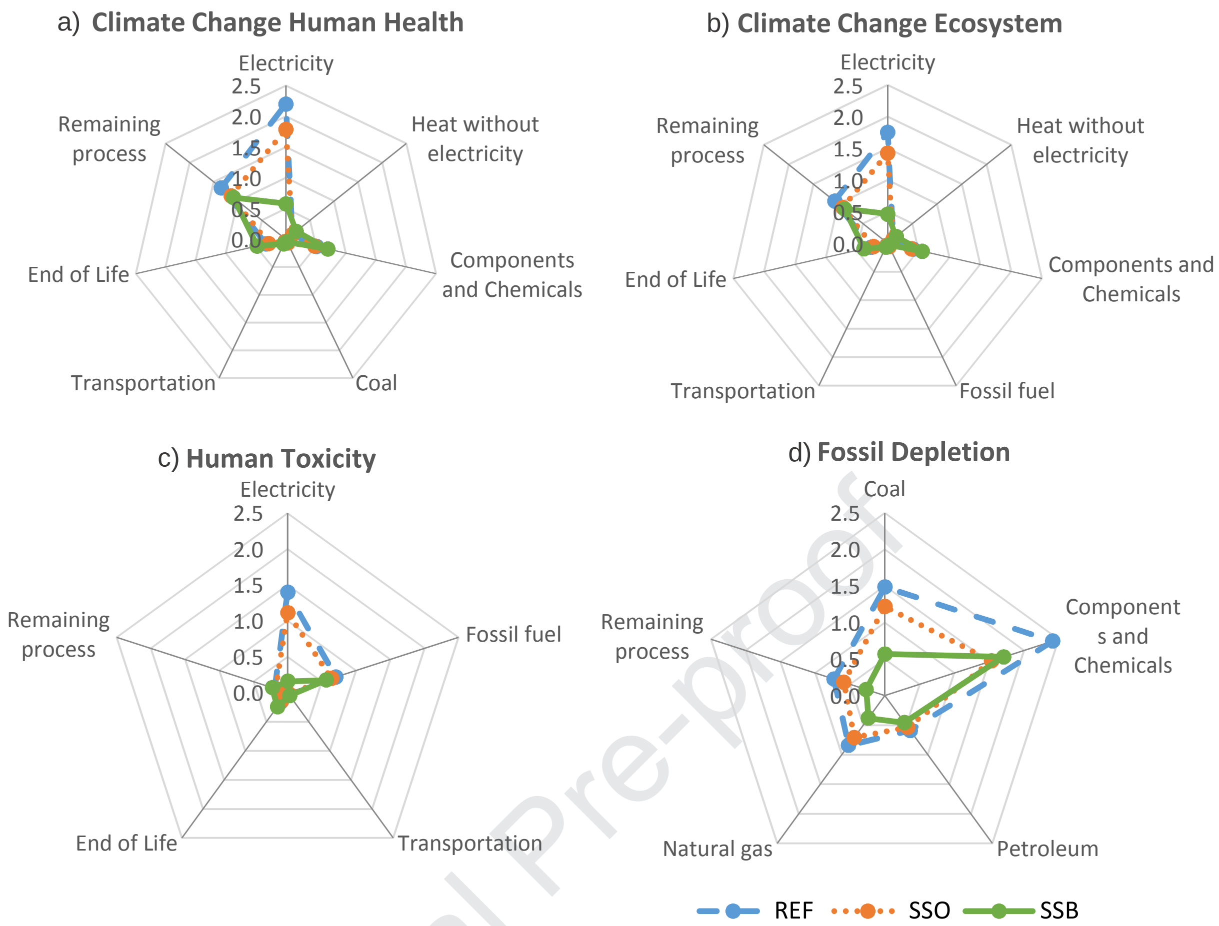



Figure 11: Contributors of environmental impact for mains indicators (mPts): (a) Climate Change - Human Health, (b) Climate Change - Ecosystem, (c) Human Toxicity and (d) Fossil Depletion.

## 3.3. Discussion

For greater clarity, the comments are gathered under three main categories based on the endpoint impacts: effect on human health, effect on ecosystems and effects on resource availability.

### 3.3.1. Influence on human health

Influence on human health is mainly explained by the CCHH, HT and PMF indicators (Figure 9). In this part, these indicators and the main contributors associated will be discussed.

The **CCHH indicator** (Figure 11, a) reflects the impact on global warming and its consequences on human health (Barnes et al., 2008). Nevertheless, it is difficult to consider all the potential effects of global warming on human health (McMichal et al., 2003). The preponderance of climate change related scores is not surprising, due to the chemicals involved in the coating formulation (resins and solvents) and to the curing procedures. In fact, to synthesize raw materials, to formulate, apply and cure coatings, electricity and heat are needed. Upstream, this energy requires the production of high voltage electricity from power plant. A 74% decrease of **electricity contribution** in the SSB process compared to the REF process and a 68% decrease compared to the SSO process is highlighted in Figure 11, a. These improvements are mainly due to the reduction of drying times and temperatures between the processes: (i) REF process: 24h in the oven at 22°C and 2h at 110°C two times due to two steps of curing, (ii) SSO process: 24h in the oven at 22°C and 2h at 110°C and (iii) for the SSB process: 1h at 80°C in the oven. The decrease in the energy consumption is particularly noticeable for the SSB system because of the less drastic curing conditions. The reduction of preparation time, temperature and curing steps thus results in a decrease in energy consumption between multilayer and self-stratified coatings.

Concerning the **Component and Chemicals contributor**, the difference is less significant between the three processes (Figure 11, a). In fact, many chemicals are used to produce coating raw materials for all processes. However, chemicals used in the SSB process are less harmful than those used in oil-based coatings. It is noteworthy that xylene represents 45% of this contributor for the REF and SSO systems. Xylene is an aromatic hydrocarbon easily absorbed by lungs. The main effect on human health of inhaling xylene vapors is depression of the central nervous system. For these toxicity reasons, the use of xylene is increasingly contested in some fields such as rail transportation. The influence of the substitution of this chemical by less toxic solvents will be discussed in the following part dealing with the toxicity for human health.

**Human toxicity (HT) indicator** (Figure 11, c) refers to the potential health damage caused by chemicals emitted into the atmosphere and the environment. Many human activities require the use of substances that may be released into the atmosphere and the environment (Krewitt et al., 2002). This is the case for many industrial and energy activities (steel mills, nuclear power plants, coal-fired power plants) and activities based on chemistry. All these activities can be potentially dangerous for human health if inhaled or swallowed.

For this indicator, the **substitution of some components** and **the decrease in electricity consumption** result in a 50% reduction in human toxicity impact for SSB system compared to REF system (Figure 10). More precisely, bisphenol A, major component of the oil-based epoxy resin, has a high environmental impact in the REF coating formulation. Bisphenol A is indeed known to be an endocrine disrupting chemical with an impact on development, metabolism and reproductive system (Flint et al., 2012). In order to lower the effect on human toxicity factor, bisphenol A epoxy resin was substituted by a bio-based epoxy resin in the SSB system. The Epoxy A resin used was obtained from the reaction between isosorbide and epichlorohydrin. Isosorbide is a molecule of interest for the replacement of fossil based products (Chrysanthos et al., 2011) (Rose and Palkovits, 2012). It can be produced by different routes including the enzymatic hydrolysis of starch or the catalytic dehydration of sorbitol, a derivative of glucose. On the other hand, xylene was replaced by less toxic solvents. Butyl acetate and ethyl lactate were selected as they exhibit few toxicological consequences (INRS, 2009) (INRS, 2011) (INRS, 2008). Thus, the replacement of toxic chemicals by less harmful components in the SSB system has a positive impact on LCA results in terms of toxicity for human health.

As well as for the CCHH indicator, electricity consumption has a major impact on the HT indicator. Indeed, electricity factor is divided by 8 for SSB process compared to the REF one (Figure 11, c). This decrease explains the fact that the environmental impact of the HT indicator of the SSB system is lower than the REF process.

In the case of the REF system, more solvents are used compared to SSO and SSB systems, and thus **more electricity and transportation** are required. Moreover, for oil-based systems (REF and SSO), chemical products are mainly imported from the United States whereas for the SSB system, the epoxy resin is produced only few kilometers by truck from the coating formulation site. It results in a higher impact on the environment with an increase in **Particulate Matter Formation (PMF)**: +14% and +26% for the REF system compared to the SSO and SSB systems respectively. The positive influence on the environmental impact of the change of the location, from China to Netherlands, of the glass manufacturing for the application of an anti-reflective coating for greenhouses has been also demonstrate recently by Natalya Tsoy (Tsoy et al., 2019).

#### 3.3.2. Influence on ecosystems

Influence on ecosystems is mainly explained by the ALO, NLT and CCE indicators. In this part, these indicators and the main contributors will be discussed.

**Agricultural Land Occupation (ALO)** and **Natural Land Transformation (NLT)** are indicators with minor environmental impact (around 4%) for each of the three processes. These indicators are linked to the loss of available habitat for living species caused by land use through human activities (agriculture, logging and deforestation, transport network control, urbanization, etc.). This phenomenon is considered to ultimately lead to a loss of biodiversity (Souza et al., 2015). A 200% increase in the agricultural land occupation indicator for SSB system was however observed in comparison with REF system. Indeed, more agricultural land is needed to cultivate plants (potatoes, beets, etc.) from which molecules used to synthesize the epoxy A resin are extracted. This growing occupation of agricultural land reduces the living space of animal species and serves the environmental impact badly for the SSB system. A recent article reports that three-quarters of all land have been transformed into agricultural fields or other structures on the Earth. In addition to that, human activities in two-thirds of the marine environment and three-quarters of lakes and rivers mean that more than 500 000 species do not have sufficient habitats for their long-term survival. Many of them are on the point of disappearing within a few decades (Watts, 2019). Nonetheless, natural land transformation impact was divided by half between REF and SSB processes. Indeed, the need for oil wells and uranium mining sites is less important for the SSB system.

**Climate Change Ecosystems (CCE)** indicator evolution (15% and 30% decrease for SSO and SSB, respectively in comparison with REF system) may be due to a **decrease in pollution, particulate matter and $CO_2$ levels**. These factors impact ecosystems (Mooney et al., 2009) by leading to a temperature rise and thus to environmental degradation with forced migration of some species and to aquatic environment degradation (Hoegh-Guldberg and Bruno, 2010). As mentioned before, the 26% reduction in electricity requirements and 72% reduction in petroleum products (Figure 11, b) for the SSB system compared to REF system mechanically reduces the impact on the climate change, and therefore consequences on the ecosystem.

### 3.3.3. Influence on resource availability

Influence on resource availability is mainly explained by metal depletion (MD) and fossil depletion (FD) indicators (Figure 9). As mentioned previously, MD is largely unaffected by the type of process used because the quantity of iron oxide used to provide fire retardant properties is the same for each formulation. Thus, only FD indicator will be considered in this part.

**Fossil Depletion (FD) indicator** is used to measure the potential depletion of fossil resources in terms of resource scarcity in relation to remaining stocks, or in relation to the additional price induced by this scarcity (Klinglmair et al., 2013).

It is clearly visible that, compared to the REF system, SSB process allows a reduction of the contributors impacting fossil depletion indicator: compounds and chemicals -70%, coal -38%, petroleum -77% and natural gas -45% (Figure 11, d). Indeed, Xylene, used as solvent in REF and SSO processes, is obtained primarily from crude petroleum, similarly to DGEBA resin, which is 100% oil-based from BPA and leads to an increase in the "compounds and chemicals" indicator into this Fossil Depletion category.

In addition to that, coal, petroleum and natural gas are natural resources. Primary energy is the amount of energy stored in these natural resources in their raw state. To make these primary energies available to users, they must be extracted, transformed, stored and distributed (Nelson, 2015). If significant amount of energy is required for a process, more natural resources will also be needed. The higher number of coating steps or hard drying conditions of oil-based processes thus requires more energy and influences this FD indicator.

#### 3.3.4. Total environmental impact of each process

When the scores of each indicator (Figure 10) of the same process are combined, the total environmental score is obtained (Figure 12) and allowed to make a general comparison.

A 13.6% gain on the total environmental impact is observed when the SSO process is used instead of the multilayered process (REF). This relatively low decrease, despite the reduction of the number of processing steps and of the power consumption, is explained by the use of the same oil-based chemicals, which have intrinsically high impact on climate change and on human toxicity, and the same curing conditions. A recent paper dealing with the influence of the surface treatment on the environmental impact of a pasteurization process also reaches the same conclusion. The coating system used to increase the anti-fouling properties of the process has been identified as the most important contributor in terms of environmental impact. This is due to many solvents and oil-based chemicals used to synthesize the coating (Zouaghi et al., 2019).

A 32.4% gain on the total environmental impact is however observed when the SSB process is used instead of the multilayered process (REF). The use of a bio-based epoxy resin and less toxic solvents is responsible for this improvement. Moreover, the less drastic (time and temperature) drying conditions applied to cure the self-stratifying bio-based coatings reduced the electricity consumption and therefore also the total environmental impact.

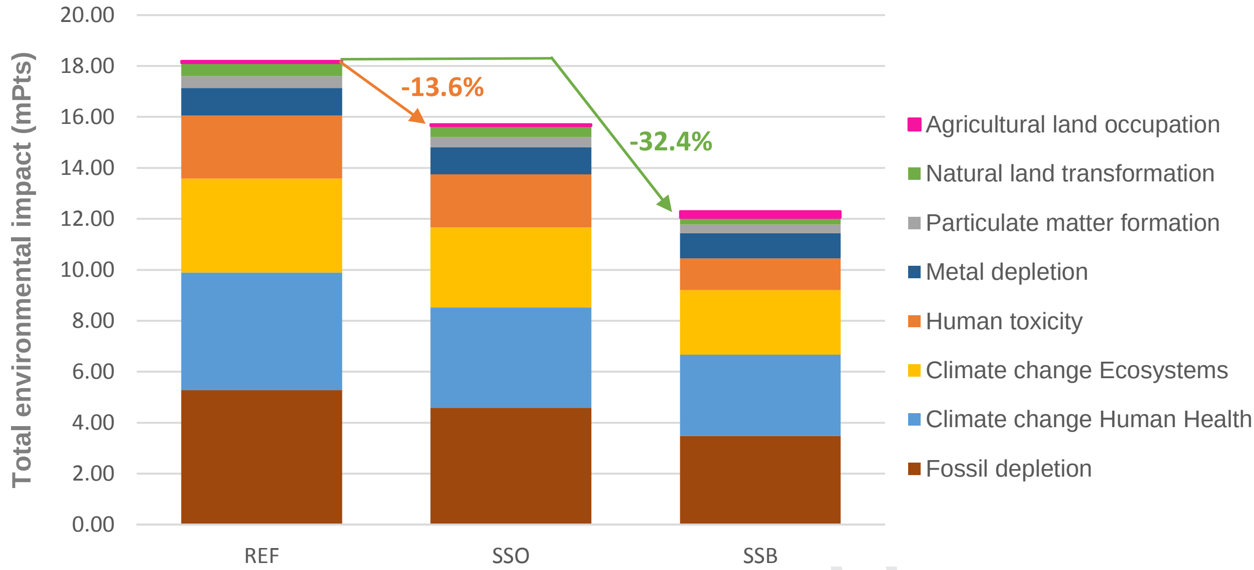



Figure 12: Comparison of total environmental score (in mPts) for REF, SSO and SSB systems for one coated 10cm² polycarbonate plate.

## 4. Conclusion

The present study aimed at quantifying the environmental impacts of multilayered and self-stratified processes through Life Cycle Analysis. Three different processes to coat a polycarbonate substrate with a flame retardant coating were compared. The one pot processes, i.e self-stratifying processes, lead to similar fire properties (UL-94 test) than those of the two-layer reference process. Furthermore, overall SS processes show better performances in terms of substrate adhesion, interlayer adhesion, visual aspect and weathering resistance than the multilayered system.

Based on the analysis conducted, it may be concluded that the use of bio-based products and less harmful solvent has a positive effect on the climate change, human health and human toxicity for the SSB process. The decrease in electricity consumption due to the reduction of the number of processing steps and the application of softer drying conditions also have a positive influence on the SSB environment impact. Indeed, the need in fossil energy and the particulate matter formation are reduced.

Finally, the global environmental impact of a coating process may be significantly improved through the use of bio-based or less harmful components and by the implementation of the innovative self-stratifying process. Life Cycle Assessment is thus an essential tool in trying to quantify and reduce the environmental footprint of processes. The difficulty lies in the accurate definition of each parameter influencing the study to carry out a fair analysis.

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# Life Cycle Assessment of multi-step versus one-step coating processes using oil or bio-based resins

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## Highlights

* Comparison of the environmental impact of multilayered vs self-stratifying coatings
* Major contribution of energy consumption for all the studied processes
* Importance of the use of a bio-based resin and of greener solvents is highlighted
* Influence of the number of processing steps and of the drying conditions