



ESTELLA

# DELIVERABLE 1.2

## Material requirements for composite manufacturing

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## Abbreviations

**CAN:** Covalent Adaptive Network

**DA:** Diels-Alder

**DGEBA:** Bisphenol A diglycidyl ether

**DGEBF:** Bisphenol F diglycidyl ether

**OOA:** Out-of- Autoclave

**RTM:** Resin Transfer Moulding

**SCRIMP:** Seeman Composite Resin Infusion Moulding Process

**TEMPO:** 2,2,6,6-Tetramethylpiperidinyloxy or 2,2,6,6-Tetramethylpiperidine 1-oxyl

**VARTM:** Vacuum Assisted Resin Transfer Moulding

**VBO:** Vacuum bag-only curing

## Executive Summary

The sustainable and recyclable composites to be developed in the ESTELLA project are made up of matrices and fibres that must meet certain requirements. On the one hand the mechanical and thermal requirements that will confer the desired properties to future applications. And on the other hand the requirements to be processed by out-of-autoclave (OOA) processes.

This deliverable compiles the study carried out by NIC, ICSO, WR, CEL, WK+ and CID on the analysis of commercial materials currently used in the manufacture of thermoset composites. Based on this study, and their previous experience, this deliverable presents the specific requirements to be met by the materials developed in ESTELLA. The result of this deliverable will feed the work packages 2 and 3 dedicated to the design of the materials and the manufacture of the composites respectively.

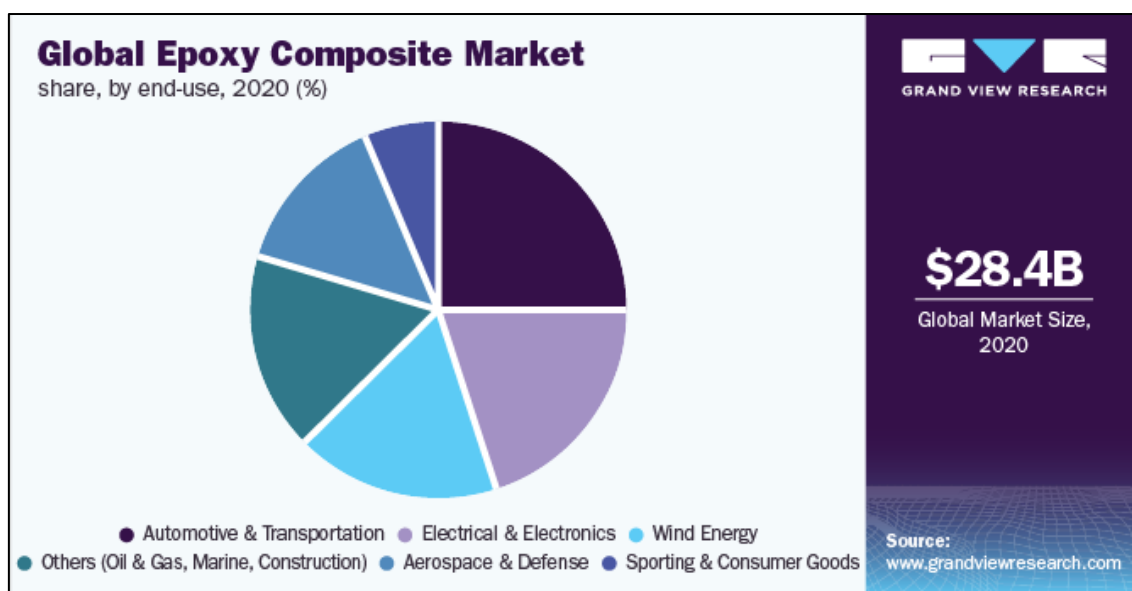
This deliverable D1.2 has fully met its objectives as set out in the Grant Agreement. It has suffered a slight delay in time due to vocational season but this has not had any impact in the progress of the project in terms of content or impacts.

## 1. INTRODUCTION

Polymer composite materials or composites are widely used in sectors of high interest for the international economy. So much so that the European market for polymer composites reached a value of 16.6 billion dollars in 2018 and is estimated to increase by 7.5% by 2025(1) due to the growing demand for lightweight materials in strategic sectors such as wind energy, automotive, aeronautics and defense. In addition, their use is expected to expand to other sectors with lower profit margins such as leisure.

The importance of these materials lies in the excellent properties that can be achieved, allowing them to replace metals reducing weight and emissions. For example, they can be found in structural elements of an aircraft or an automobile or in construction.

The main composition of these composites is thermosetting resin (mainly epoxy) and carbon or glass fibre. Figure 1 shows the use of epoxy composites in different sectors.



**Figure 1. Epoxy composite on the market**

Therefore, the advantages of using composites are clear, especially from an environmental point of view. However, despite the advantages of using composites, they have not been fully implemented in most sectors and this is due to its **low recyclability rate**. As a result, the ESTELLA project was created with the main objective of improving the recyclability of these composites; by acting on the whole value chain of the materials for that purpose. Inherently recyclable polymer resins will be designed and developed through the introduction of reversible covalent links between the polymer chains, which provide the material with the ability to return from the processed state to the original monomers. Furthermore, the manufacturing processes of epoxy resins will be conveniently adapted, in order to maximize the economic revenue and the eco-efficiency along the value chain.


The **design of the new material requires some important aspects** to be taken into account and their due fulfilment will allow the ESTELLA project objective to be achieved. These aspects are:


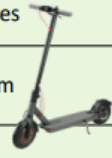
- 1) The **final properties of the raw materials** (matrices and fibres).
- 2) The **ability to be processed by OOA techniques**.
- 3) **Recyclability** (ESTELLA project objective).

The final properties of the raw materials and their processability are objectives of this report (D1.2) and the recyclability properties are covered in D1.4.

## 2. REQUIRED PROPERTIES OF THE RAW MATERIALS

The composite materials to be developed in the ESTELLA project consist of the elements shown in the Figure 2:



COMPOSITE		
	C-1	C-2
Type	Discontinuous fibre	Continuous fibre
Components	Two selected resins + (Ligno)cellulose	Two selected resins + Hemp
Manufacturing technology	Out-Of-Autoclave (OOA) processes	
Prototypes	Window frame 	Scooter platform 
Partners	CIDAUT	

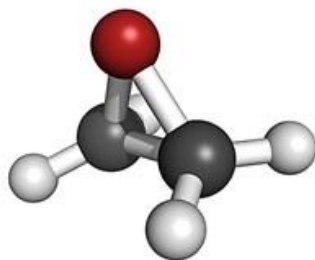
**Figure 2. ESTELLA composites manufacturing**

The nature of the matrices will be epoxy resins, one of which is a modified commercial epoxy and the other two will be synthesised using biomaterials. The nature of both reinforcements is biological and the major difference between them is the morphology: cellulose is a discontinuous fibre and hemp is continuous.

The required properties of each material are described below:

### 2.1. MATRICES - EPOXY

The epoxy resin consists of a broad group of reactive compounds that are characterized by the presence of an oxirane or epoxy ring. This is represented by a three-member ring containing an oxygen atom that is bonded with two carbon atoms already united in some other way (Figure 3).



**Figure 3. Epoxy molecular scheme**

Hence, the presence of this functional group defines a molecule as an epoxide – where the molecular base can vary widely resulting in **various types of epoxy resins**. And they are successful because they offer the diversity in molecular structure that can be produced using the same chemical method.

Further, epoxy resins can be combined with varied **curing agents, modifiers to achieve the properties required for a specific application**.

The types more common in composite applications are: phenolic glycidyl ethers, aromatic glycidyl amines, and cycloaliphatics (2).

The most relevant properties of Epoxy resins are:

- High strength
- Low Shrinkage
- Excellent adhesion to various substrates
- Effective electrical insulation
- Chemical and solvent resistance

The ranges of mechanical properties are shown in Table 1.

**Table 1. Commercial Epoxy mechanical properties range**

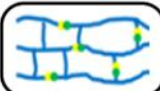
Property	Range
<b>Tensile Strength (MPa)</b>	90 - 120
<b>Tensile Modulus (MPa)</b>	3100 - 3800
<b>Glass transition temperatures, Tg (°C)</b>	150 - 220

Aside from the properties mentioned above, epoxy resins have two main drawbacks which are their brittleness and moisture sensitivity.

### 2.1.1. ESTELLA epoxies

The ESTELLA project, in order to complete the objective, will develop three epoxy resins with specific properties. Figure 4 shows the nature (origin), the type of covalent adaptive network (CAN) and its recyclability and the ESTELLA partner involved.





MATRIX			
	M-1	M-2	M-3
Origin	Fossil	Renewable	Renewable
CAN type	Associative (ADCAN)		Dissociative (DDCAN)
CAN mechanism	Transesterification		Diels-Alder
CAN stimulus:	Heat:	Acid hydrolysis:	Heat:
Recycling mechanism	Reprocessing and depolymerisation	Depolymerisation	Reprocessing and depolymerisation
Recycled products	Reprocessing: Composite thermostable		Depolymerization: Monomers (or other molecules) and fibres separately
Partners	ICSO	NIC	WR

**Figure 4. ESTELLA matrices summary**

**WR** will develop a resin that is strictly speaking not an epoxy resin. Although the final mechanical properties of typical cured epoxy resins are pursued, the curing of the reactive components is not based on epoxy groups, but on unsaturated groups susceptible to Diels-Alder (DA) cycloaddition. The reprocessability of Diels-Alder resins is based on the fact that the DA reaction is reversible; at high temperatures, chemical bonds formed by the DA reaction break ('retro-DA') to give back the original reactive groups. Similar to most epoxy resins, the DA resin is a two-component resin, which means that both reactive components need to be mixed prior to application. One of the aims of WR is to design a 'one-pot' version of the resin. This reduces processing times and handling difficulties.

In the Estella project, **NIC** will design a lignin bio-based epoxy resin. Lignin products will be prepared with appropriate reactive sites (hydroxyl and carbonyl groups). In order to form a resin having a covalent adaptive network, the lignin products can be functionalised, for example, by the synthesis of glycidylated lignin products or formation of imine bonds. The use of lignin itself as curing agent with or without the presence of a co-curing agent will be evaluated in order to reduce the complexity of the system.

The **ICSO** concept consists of imparting of CAN into fossil epoxy resin cured with selected carboxylic acids and/or anhydrides, to obtain polymers with inherent recyclability. Transesterification among hydroxyester bonds present in the obtained resins will allow for reaction mechanism comprising diffusion of solvent molecules into the network, breakage of polymer chain, diffusion of chain segments into the solvent, and further repolymerization. Viscosity of resins compositions will be adjusted by epoxidized plant oils serving as a reactive diluents to provide optimal processability in the composite production.

In order to establish the properties that can be achieved with the designed resins, a thorough review of the state of the art has been carried out.

#### 2.1.1.1. ESTELLA Lignin-based epoxy resins

Table 2 shows the most relevant information obtained from the review related to resins.

**Table 2: Properties of lignin-based epoxy resins**

Lignin type	Lignin-modifying agent	Curing agent	Glass transition T <sup>a</sup> (°C)	Tensile strength (MPa)	Young's modulus (GPa)	Flexural strength (MPa)	Ref.
<b>Lignoboost Kraft lignin from Spruce or Eucalyptus, extracted by organic solvents</b>	epichlorohydrin	Jeffamine D2000 and Jeffamine D400	79	66	1.7	---	(3)
<b>Organosolv lignin isolated from poplar wood</b>	succinic anhydride	Bisphenol A diglycidyl ether (DER 332) (cat.: Zn(acac) <sub>2</sub> )	71-111	60-69	---	101	(4)
<b>De-polymerized Kraft lignin and de-polymerized organosolv lignin</b>	epichlorohydrin	4,4'-diaminodiphenylmethane	---	150-250	---	---	(5)
<b>Alkali lignin</b>	epichlorohydrin	methyl nadic anhydride + 2-ethylimidazole	131-143	---	---	120-127	(6)
<b>Hydrolysis lignin, residue from Aspen wood</b>	epichlorohydrin	4,4'-diaminodiphenylmethane	102-153	138-187	12.3 - 23.2	47 -258	(7)
<b><u>Enzymatic hydrolysis</u> lignin</b>	epichlorohydrin	Bisphenol A diglycidyl ether + dodecanedioic acid (+zinc catalyst)	---	39.5-46.8	2.00	---	(8)
<b>Kraft lignin</b>	succinic anhydride	Bisphenol A/F diglycidyl ether (DER 353) + nadic methyl anhydride	---	---	3.62	48.8	(9)

Based on the reviewed papers, the formation of lignin-based epoxy resins, glass temperatures range from 70 to 150 °C, tensile strength from 40 to 150 MPa, density from 2 to 7 mmol/cm<sup>3</sup> and Young's modulus from 2 to 12 MPa. However, it should be highlighted that these properties are dependent on various factors, namely chosen biomass, lignin composition and content, curing agent/process, reaction conditions, etc.

According to the expert opinion of NIC and WR, supported by the state of the art, the development of a resin with a bio-based content of more than 60% is foreseen, with properties as shown in the Table 3.

**Table 3. Targeted mechanical and thermal properties of the ESTELLA biobased resins**

Mechanical properties	Thermal properties
<b>Tensile Strength (MPa)</b>	>40
<b>Young Modulus (GPa)</b>	≥1
<b>Glass transition temperatures, Tg (°C)</b>	50-100
<b>Strain at break (%)</b>	10
<b>Compressive strength (MPa)</b>	20-60
<b>Flexural strength (MPa)</b>	20-60

### 2.1.1.2. ESTELLA fossil resin

Table 5 shows the most relevant information obtained from the review related to resins.

**Table 4: Properties of fossil-based epoxy resins**

Resin	Curing agent	Glass trans. T <sup>a</sup> (°C)	Tensile strength (MPa)	Young's modulus (GPa)	Ref.
<b>Bisphenol F diglycidyl ether (DGEBF)</b>	Aminobenzylamine/ Terephthalaldehyde	100,2	72,77	2,7	(10)
<b>Bisphenol A diglycidyl ether (DGEBA)</b>	amino-capped aniline trimer/ DGE- DHBP	58	-	1,4	(11)
<b>Bisphenol A diglycidyl ether (DGEBA)</b>	Fatty acid (Pripol 1040) + Graphene Nanoplate (GnP)	5,8	3,7	-	(12)
<b>Bisphenol A diglycidyl ether (DGEBA)</b>	Vanilin/IPDA polymer	121	60,1	2,6	(13)
<b>Bisphenol A diglycidyl ether (DGEBA)</b>	Sebacic acid and Acrylonitrile/ zinc methacrylate copolymer	42,9	33	0,6	(14)
<b>Bisphenol A diglycidyl ether (DGEBA)</b>	Nadic methyl anhydride/ triethanolamine 1,4-	119,9	94	2,3	(15)
<b>Bisphenol A diglycidyl ether (DGEBA)</b>	cyclohexanedicarbo xylic acid/ sebacic acid	46,6	35	1,3	(16)

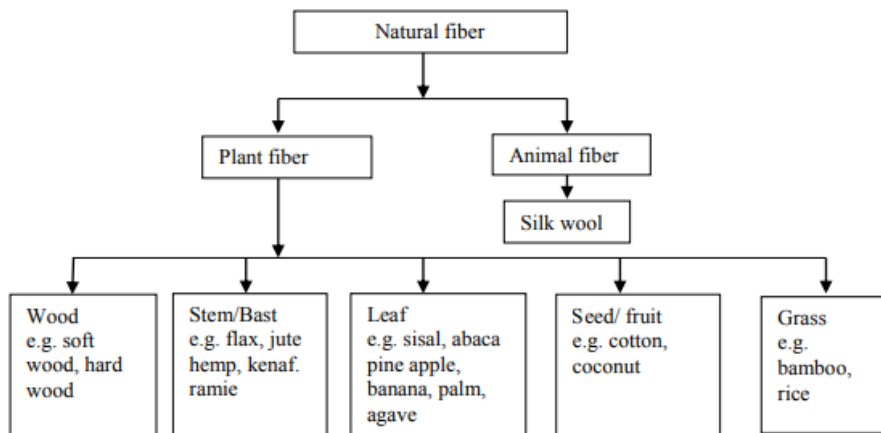
Based on the reviewed papers, epoxy vitrimers based on fossil epoxy resins, are characterized with these values: glass temperatures range from 6 to 121 °C, tensile strength from 4 to 94 MPa, and Young's modulus from 0,6 to 2,7 GPa. According to the expert opinion of ICSO, supported by the state of the art, targeted properties of the resin to be developed are shown in the Table 5.

**Table 5. Mechanical and thermal properties of the ESTELLA fossil resins**

Mechanical properties	Thermal properties
Tensile Strength (MPa)	>40
Young Modulus (GPa)	>1,5
Glass transition temperatures, T <sub>g</sub> (°C)	40-90
Strain at break (%)	5-15
Compressive strength (MPa)	> 50
Flexural strength (MPa)	> 50

## 2.2. Reinforcements

From natural reinforcement, plant fibres are one of the most popular and most used for the fabrication of composites. Animal reinforcement is used to a lesser extent. Figure 5 shows some commonly used natural fibres in composite fabrication.




**Figure 5. Some Commonly used Natural Fibers in Composite Fabrication**

Bio-polymers or synthetic polymers reinforced with plant fibres give the best quality of composites with properties like light weight, bio-degradability and renewability. However, to be efficiently used in composite, two aspects should be considered:

- The reinforcements should **not present any thermal degradation** during the composite manufacturing and the recycling processes.
- The **compatibility reinforcements-matrix** should be suitable to ensure good mechanical properties.

In the ESTELLA project, two natural reinforcements will be used, one for applications with higher mechanical requirements (hemp) and the other for applications with lower requirements (cellulose). Both will undergo modifications (chemical and/or physical), as shown in Figure 6, to comply with the above mentioned aspect.



FIBRE			
	(LIGNO)CELLULOSE	HEMP	
Origin	Renewable		
Fibre type	Discontinuous	Continuous	
Challenge faced	Reduce moisture absorption		
Modification	Chemical: Funtionalization	Physical: Plasma	Chemical: Funtionalization
Partners	CEL and NIC	WK+	

Figure 6. ESTELLA reinforcements summary

### 2.2.1. (Ligno)cellulose (discontinuous fibre)

Essentially, cellulose fibres can be considered as natural substances consisting of cellulose, lignin and hemicelluloses, also known as lignocellulosic natural fibres. Cellulose is a linear polysaccharide capable of forming strong natural fibres due to the intramolecular and intermolecular bonds it is able to form. Because of this strength and its high abundance, cellulose has been widely studied as a potential reinforcement for polymers. Table 6 lists the main properties reported in the literature on cellulose fibres.

Table 6: Discontinuous (ligno)cellulose nanofibres properties

Type	Source	Diameter (nm)	Length (µm)	Thermal decomposition T <sup>a</sup> (°C)	Ref.
Cellulose nano fibre	Industrial	35	2–3	---	(17)
Cellulose nano fibre	TEMPO (2,2,6,6-Tetramethylpiperidinyloxy or 2,2,6,6-Tetramethylpiperidine 1-oxyl)-mediated oxidation	12.3	1.53	---	(18)
Cellulose nano fibre	TEMPO-mediated oxidation	7.7-37.7	0.087-1.343	230 -300	(19)
Lignocellulose nano fibre	Mechanical pre-treatment OR enzymatic pre-treatment OR TEMPO-mediated oxidation	---	---	300	(20)

The introduction of cellulose nanofibre in the epoxy will have a positive effect on the mechanical properties and it has been corroborated with the state of the art study shown in Table 6. The degree of improvement could vary depend mainly on the nature of the matrix, the aspect ratio of the fibre and the quality of dispersion. For example, the tensile strength and the tensile modulus have a range of improvement between 50% and 300%.

As it is mentioned, the morphology of the fibre has a huge impact on the quality of the properties achieved in the final composite. Higher the aspect ratio (length/diameter) is, higher the potential mechanical improvement is but higher the price is and higher the difficulty to achieve an appropriate dispersion is. The morphology, mainly the aspect ratio, will then be varied (diameter: 20 nm-1µm length: 1µm-10µm) to optimize the mechanical reinforcement. The lignin/cellulose ratio will also be a parameter that will be varied to improve the reinforcing properties.

In this project a 100% reinforcement of the tensile modulus will be targeted by playing on the morphology of the fibre and on the nature of the functionality to achieve adequate dispersion of the fibre and interactions with the matrix.

According the most relevant aspects, especially for thermal stability, the (lingo) cellulose to be developed in the ESTELLA project will resist high temperature of composites manufacturing (80-120°C) and recycling processes (reversibility of bonds at temperature below 160°C).

A thermal stability of 200°C for all the different kind of cellulose fibres should then ensure the absence of any thermal degradation during the whole life of the product. As it is reported in literature, the degradation of cellulose fibres occurs only at temperature above 300°C. However, the needed defibrillation of the cellulose fibre induces a reduction of the thermal stability with starting point of thermal degradation going from 333°C to 318°C (5°C/min rate)(21). When specific functionalization is introduced on the surface of the cellulose nanofiber, degradation steps like decarboxylation of sodium anhydroglucuronate for TEMPO oxidized nanocellulose fibres can occur at lower temperature but the starting degradation temperature (218°C) (ref 3) remains above the targeted value (200°C) confirming that no specific issue should originate from the degradation of the fibres.

As a resume, Table 7 shows a summary of ESTELLA (lingo) cellulose properties:

**Table 7: Summary of ESTELLA cellulose nanofibres properties**

	Tensile strength	Diameter (µm)	Length (µm)	Thermal decomposition T <sup>a</sup> (°C)
<b>ESTELLA Nanocellulose</b>	100% reinforcement of the tensile modulus	20 -1	1-10	318 -333

### 2.2.2. Hemp (continuous fibre)

Hemp fibre-reinforced composites have been successfully used for lightweight and low-cost applications in recent years, but significant barriers for structural applications of these composites still exist. These barriers include lack of confidence in the use and performance of natural plant fibres and their composites limited understanding of diffusion behaviour and poor resistance to moisture(22)(23). Table 8 shows the most relevant information obtained from the literature review related to hemp.

**Table 8. Continuous hemp fibres properties**

Type	Tensile strength (MPa)	Modulus (GPa)	Density (g/cm <sup>3</sup> )	Thermal decomp. (°C)	Stiffness (GPa)	Failure to strain (%)	Ref.
<b>Hemp fibre</b>	900	70	---	---	---	---	(24)
	20-1000	---	1.4-1.6	---	17.6-35.5	2.1-2.6	(25)
	---	---	---	280-500	---	---	(26)
	---	---	---	250-320	---	---	(27)

Making continuous reinforcement using hemp fibre is quite challenging and it will be tried in this ESTELLA project. Before that as a reference material another bast fibre flax will be used for the initial testing purpose.

As Table 8 shows, the mechanical properties of the hemp fibre are higher than other plant reinforcement. The ESTELLA hemp will be developed in order to achieve a tensile strength around 600-900 MPa, the flax will be between 350-1030 MPa and the modulus between 20-60 Gpa. These properties are related with the morphology of the fibres and the pre-treatment. The ESTELLA hemp has inherent hollow microstructure and the presence of cellulose or lumen shows extremely lower thermal conduction than conventional fibres. The diameter of the elementary fibre will be between 10-45 µm. Other relevant property is that it will have a density range between 1-45-1.55 g/cm<sup>3</sup>.

As mentioned earlier, the hemp yarns from bast is a demanding task hence the plan is to buy the commercially available yarn and to weave them in to a fabric by applying different patterns (Plain and Twill) for the composite application.

The surface modification will be carried out to improve the surface morphology and to remove the hemicellulose. The chemical modification methods like NaOH treatment, peroxide and anhydride treatment will be utilized. The atmospheric plasma treatment will be utilized to improve the wetting behavior and the surface morphology.

In addition, this modification will improve the flexibility as well as the thermal stability. The initial decomposition due to humidity is up to 150°C and the first stage of decomposition starts around 200°C hence the natural fibre should



withstand minimum of 200°C. Thermal characterization will be utilized to assess the thermal stability of the flax or hemp fibre.

Table 9 shows a summary of ESTELLA cellulose targeted properties:

**Table 9: Summary of ESTELLA cellulose nanofibres properties**

	Tensile strength (MPa)	Modulus (GPa)	Density (g/cm <sup>3</sup> )	Diameter (µm)	Thermal decomposition T <sup>a</sup> (°C)
<b>ESTELLA Hemp</b>	350-1030	20-60	1.45-1.55	10-45	200

### 3. RAW MATERIAL PROCESSING CAPACITY

**Thermoset composites could be processed by several techniques**(28) depending on the morphology of fibres, the product geometry,... They could be classified in two main types: autoclave moulding and OOA processes.

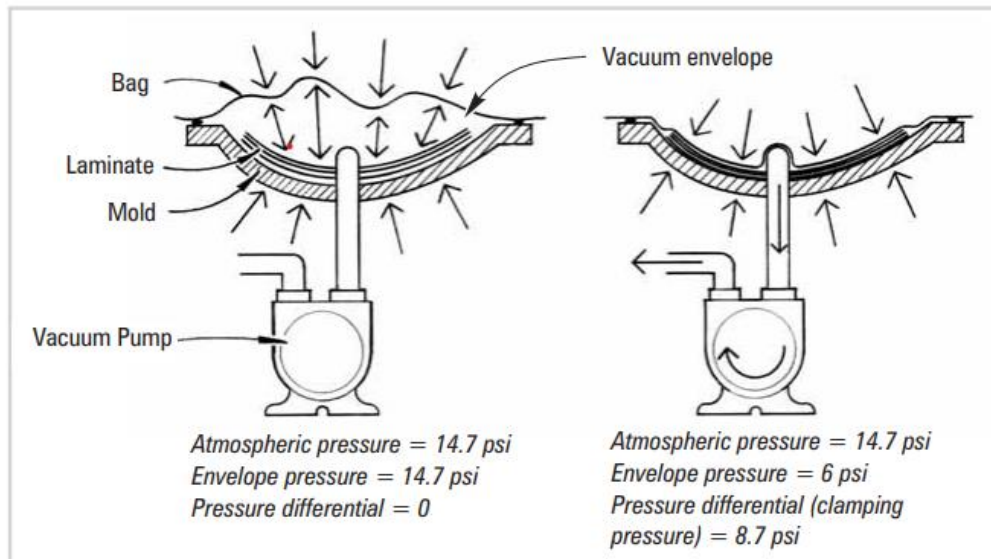
#### 3.1. Autoclave moulding

**Autoclave moulding** is one of the open moulding processes where the moulded part is cured by application of the vacuum, heat, and pressure of the inert gases. The moulded part (or piece) is placed in a plastic bag, where the air is exhausted by a vacuum pump. This removes air inclusions and volatile products from the moulded part. Then, heat and inert gas pressure are applied in the autoclave causing curing and densification of the material. Finally, autoclave curing enables fabrication of consistent homogeneous materials. The method is relatively expensive and is used for manufacturing high-quality aerospace products. This process has some advantages such as the pressure that helps bond composite layers, the ability to manufacture pieces with high fibre loads, as well as high-quality products. However, it **requires long cycle times and high energy and environmental costs**. For this reason, composite materials manufacturing processes are nowadays aiming to avoid this process (29).

#### 3.2. OOA processes

##### 3.2.1. Vacuum Bagging

The bagging process is a process in which a flexible, transparent film is used to enclose and compact wet laminates that are cured under atmospheric pressure. This method uses a vacuum pump to draw air from inside the vacuum bag and then compresses the part under atmospheric pressure (Figure 7). The resin is squeezed and sucked out of the wet laminate in the bleeder. This is a simple process that can use a wide variety of moulds. However, when it comes to large parts it requires longer set-up times and the manufacturing rate is low (30).



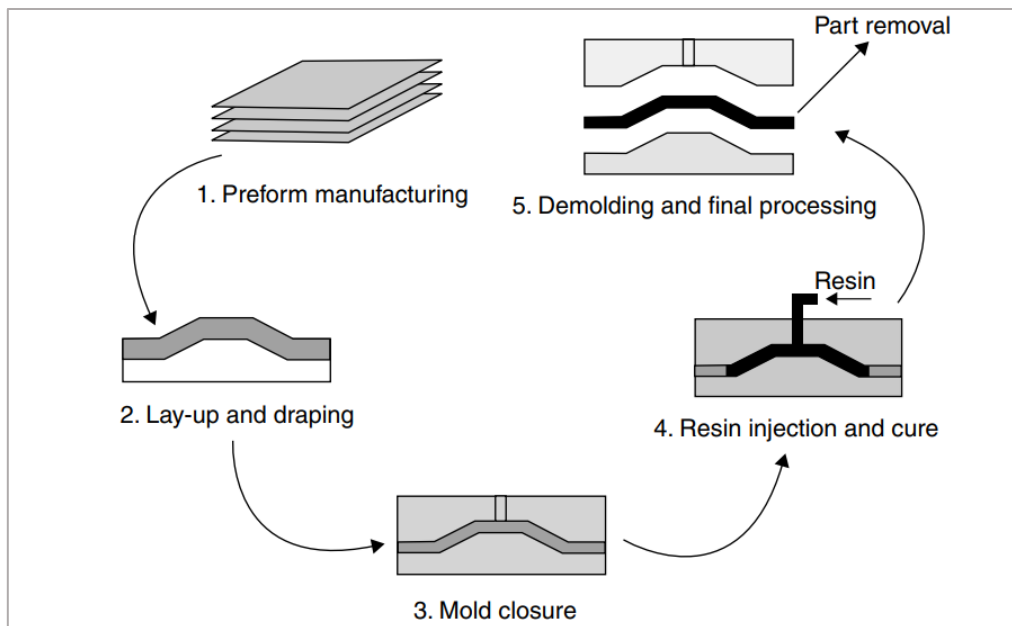
**Figure 7. A typical vacuum bagging lay-up before and after vacuum is applied**

### 3.2.2. Vacuum Bag Only/Oven Cure

Vacuum bag-only curing (VBO) uses the vacuum bag as the previous process and cures inside a non-pressurised oven. In the absence of high pressure, it is important to consider the property of the OOA resin, the architecture of the fibre bed and the prepreg system. Proper fabrication requires the removal of bubbles and volatiles before the gel time in order not to generate voids and thus defects. The mechanical properties of the composites are similar to those manufactured with the autoclave but the manufacturing times are longer.

### 3.2.3. Resin Transfer Moulding (RTM)

The RTM process involves using a closed mold to fabricate a composite part. Fiber preform is cut according to the mold shape and placed in a closed mold cavity. A low-viscosity thermoset resin is injected through the injection port into the mold cavity, usually with a 3.5–7 bar pressure. The injected resin impregnates the preform evacuating entrapped air bubbles until complete wetting is reached. Once the resin starts exiting from the vent ports, the resin injection is stopped, and vent ports are closed. The resin is allowed to cure by heating the mold or the initial addition of inhibitors to the resin system. After the resin is cured, the mold is opened, and the part is de-molded. Some variants of the RTM process are VIPR, FASTRAC, light RTM (LRTM), structural reaction injection moulding (S-RIM), and co-injection resin transfer moulding. Figure 8 shows the scheme of the process.

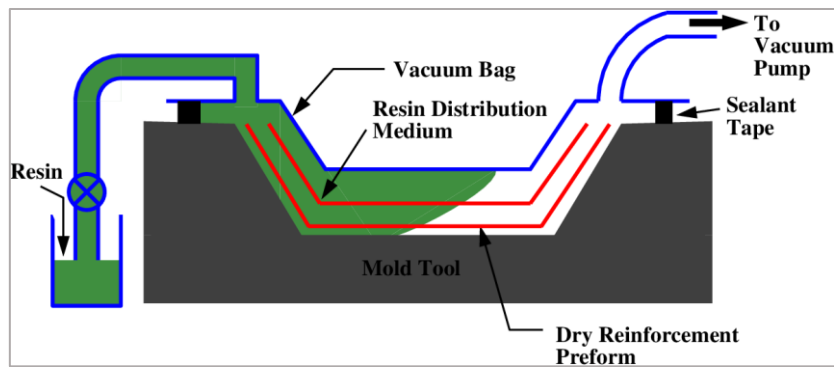


**Figure 8. RTM process scheme**

Some advantages of RTM are that the process can produce parts with close dimensional tolerance and an improved surface finish. Parts made by RTM have a high-volume fraction of about 60–70%. RTM can manufacture complex-shaped composite parts. Consistent reproducibility of composite parts can be achieved using the RTM process. Due to high resin pressure and faster mould opening and closing, a fast-manufacturing cycle is reached, further improved by process control. Some drawbacks of the RTM process are the limited size of parts that can be manufactured. Fibre wash can occur due to high resin pressure and loose fibre compaction. Furthermore, improper location of injection gates and vents can lead to a macro void in the composite (31).

### 3.2.4. Vacuum Assisted Resin Transfer Moulding (VARTM)

In the VARTM method, the reinforcement is placed on a one-sided mould and sealed with a vacuum bag to form a closed mould. A vacuum is applied at the vent, which drives the resin under atmospheric pressure to impregnate the reinforcement while evacuating the air bubbles and compacting the fibre preform. The resin flows through the porous preform and arrives at the vent. The injection is closed, but the vacuum is maintained until the part is completely cured and de-moulded. Figure 9 shows the scheme of the process.



**Figure 9. VARTM process scheme**

The VARTM process is used to produce large composite parts at a low cost with a low production volume. This process is widely used in the energy, aerospace, marine, defense, and infrastructure building industries. Variations of VARTM have been invented to cater to the manufacturing of complex parts with better quality at a reduced cost. The VARTM process has some advantages: flexibility of mould tooling and selection of mould materials, resin and catalyst can be stored separately and mixed before infusion, low emission of volatile organic compound, and visible inspection of the process to identify and manage dry spot occurrence. However, some drawbacks of this process are that consumables such as sealing tape, peel-ply, and vacuum bags may not be reusable. The low resin injection pressure can limit void compressibility resulting in high void content and low fiber volume fraction. The process may be susceptible to high chances of air leakage, depending on the operator's skill level (32).

### 3.2.5. Seeman Composite Resin Infusion Moulding Process (SCRIMP)

The SCRIMP process is a modification of the VARTM process. It is an improved version of the VARTM process to efficiently and effectively distribute resin during impregnation using a distribution media. Therefore, it is used for making high-quality and repeatable parts with minimal volatile emissions. Composite parts made by the SCRIMP process have a high fiber volume fraction typical of about 60–75%. The distribution media is a highly permeable material placed between the vacuum bag and the topmost layer of the fabric. It helps to distribute the resin quickly, thereby reducing the fill time. The resin initially flows through the DM layer before wetting the reinforcements through the thickness direction (see Figure 10)(33).

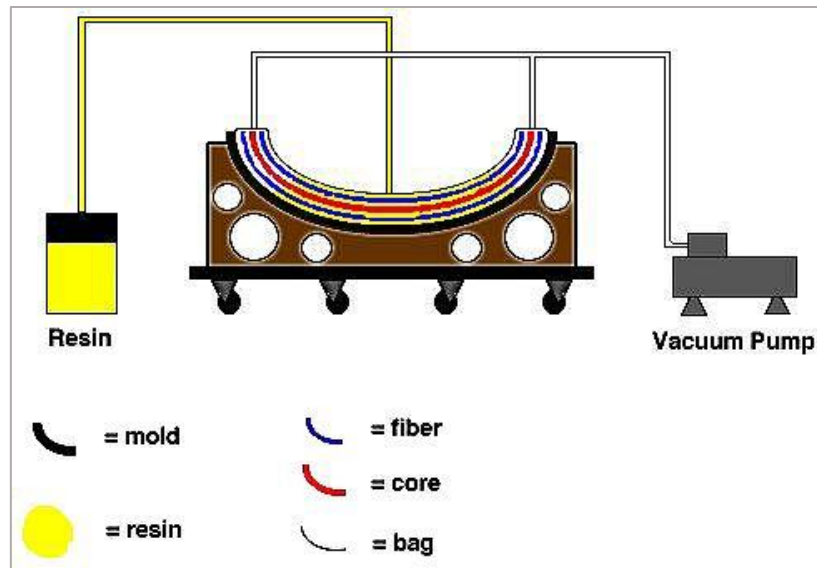


Figure 10. SCRIMP process scheme

### 3.2.6. Compression Moulding

Compression moulding is among the oldest materials processing techniques. For plastics, it was one of the first industrial methods, and is also known as matched die moulding. The basic process consists of heating a thermoset resin, under severe pressure, within a closed mould cavity until the resin cures through a chemical reaction of cross-linking polymeric chains. Under pressure, the resin liquefies and flows, taking the shape of the mould cavity, and then hardens into the desired product (Figure 11). Once sufficiently cooled and strong, the part is removed from the mould. The curing reaction continues while cooling to ambient (room) conditions.

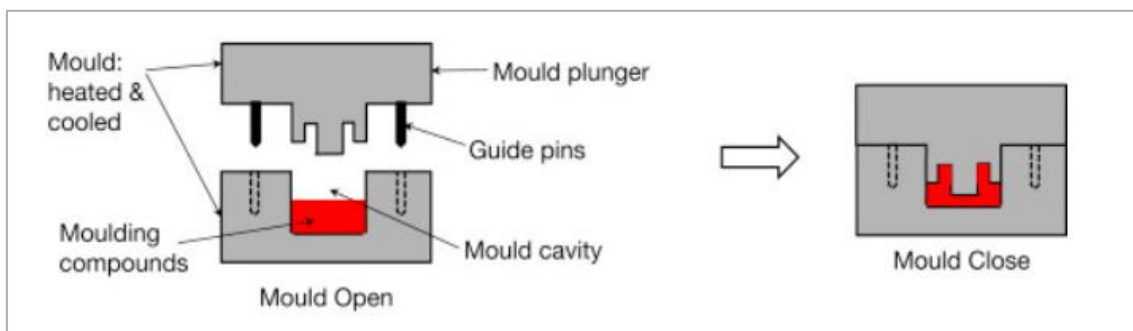


Figure 11. Compression Moulding process scheme

### 3.3. Processability requirements

The ESTELLA project is committed to the manufacture of composites using environmentally friendly processes, i.e. outside the autoclave. This reduces energy consumption and environmental emissions.

The specific process in which they are to be manufactured will be defined when the prototypes are designed, as they depend on their geometric

complexity. However, regardless of the selected process, all of them have in common a series of parameters that will make the designed materials capable of being manufactured by OOA processes.

The most relevant parameters for the processing of the matrices are:

#### - Viscosity

The matrices must penetrate the fibre and be injected or infused easily, so the viscosity of the matrices must be as low as possible. According to commercial resins the optimum viscosity should be 10-600mPa.s.

#### - Curing time

The curing curve relates the processing temperature to the time required for all the matrix bonds to be established and to obtain maximum properties.

In the case of the resins designed in the ESTELLA project; these are not fast curing resins (cured in a few minutes) as they lack catalyst. The curing time of the resins varies from 30 min to a few hours, usually up to 6 h, and the process follows a multi-step approach at different temperatures, ranging from 40 to 140°C(34) .

Table 10 shows some examples of commercial epoxy rheological properties and Table 11 the targeted ESTELLA epoxy properties:

**Table 10. Commercial epoxies rheological properties**

Reference	Viscosity at 120°C (mPa.s)	Curing time at 120°C (min)	Reference
RTM6_HexFlow	30	>240	(35)
Resoltech 1500	16	2	(36)
Araldite LY3585	40-60	2	(37)

**Table 11. ESTELLA epoxies properties**

	Viscosity at 120°C (mPa.s)	Curing time at 120°C (min)
ESTELLA bio matrices	10- 500	360
ESTELLA fossil matrix	10-500	15-90

With regard to fibres, the parameters for their processability are ease of handling and compatibility with resins. Both are achieved thanks to the weaving of the fibres (in the form of fabric for hemp and mat for cellulose) and the compatibilisation pre-treatments described above.

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