Bio-Polymers for Green Composites

Claudio Scarponi*

Dipartimento di Ingegneria Meccanica e Aerospaziale, Università "La Sapienza", Via Eudossiana 18-00184 Roma

*Corresponding author: E-mail: claudio.scarponi@uniroma1.it

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Abstract

The purpose of the paper is the description of the main applications of the so-called "Green Composites", composed by natural fibers, such as hemp and flax, embedded by bio-polymers and the contribution of such materials to the reduction of the environmental pollution. Since the Kyoto Conference at the end of 90's of the last century, there is higher and higher attention to the concepts of sustainability. The market of composite materials based on synthetic resins and synthetic fibers, such as glass and carbon fibers, is huge. Billions of tons worldwide are used in the fields of civil constructions, marine constructions, sport goods, automotive, aerospace constructions. After a short description of the most interesting bio-polymers and natural fibers of interest worldwide, the paper describes the products developed nowadays and future possibilities for more applications, with care to future applications in aeronautical constructions.

Keywords: Bio-polymers, composite materials, environment, applications, aeronautical parts.

Introduction

The growing global environmental awareness and social concern, high rate of depletion of petroleum resources, concepts of sustainability and new environmental regulations have together triggered the search for new products and processes that are compatible with the environment.

In order to avoid irreversible consequences, the most industrialized countries came together to realize some plans in order to reduce the concentration of gaseous compounds without affecting the economy [**1**]. Those are enclosed in many international agreements and conferences, such as the *Kyoto Protocol, the Copenhagen Agreements, Paris climate Conference and the so called "20-20-20" agreement* in European countries.

International environmental treaty produced by the *United Nations Conference on Environment and Development (UNCED)*, informally known as the Earth Summit, held in Rio De Janeiro, Brazil, June 1992. The treaty intended to achieve "Stabilization of Greenhouse Gas concentrations in the atmosphere at a level that would prevent dangerous anthropogenic interference with the climate system".

It establishes legally binding commitments for the reduction of four greenhouse gases (Carbon Dioxide, Methane, Nitrous Oxide, Sulphur Hexafluoride), and two groups of gases (Hydrofluorocarbons and Perfluorocarbons) produced by (industrialized) nations.

On the 23rd of January 2008, the European Union gave the green light to a program of legislation on reducing CO_2 emissions and increasing the sources and supplies of renewable energy.

The abbreviation "20-20-20" says it all: to reach before the year 2020:

20% of the energy production from renewable sources
20% improvement in energy efficiency.
20%. carbon dioxide emissions reduction.

With increasing number of applications and mass volume uses (aeronautical/automotive) in particular, recording double digit growth worldwide, disposal of composites after their intended life is already becoming critical, as well as expensive.

The increasing sensibility to the environmental issue has affected the market demands, which has veered towards the usage of Natural Fiber Composites (CFN).

Industries, laboratories and research facilities are working on those called "Green Composites", entirely bio-degradable, obtained through the impregnation of natural fibers such hemp and flax, embedded in a biodegradable polymer matrix.

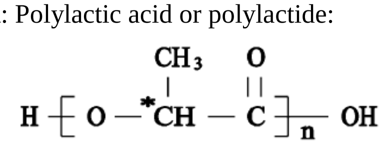
Its nearly complete biodegradability, the absence of toxic agents and the little energy costs in term of usage, defines it as protagonist of the immediate future.

Materials and properties: Biopolymers

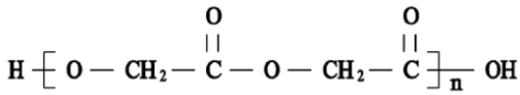
Despite the high number of biodegradable polymers in the market, hereby four basic polymers have been chosen. In the scientific literature, these four attracted more labs because of their characteristics [**2**].

For convenience reasons, from now on, these polymers will be denoted by their abbreviations and composition:

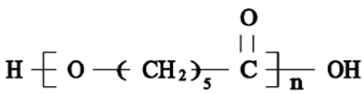
- **PLA**: Polylactic acid or polylactide:



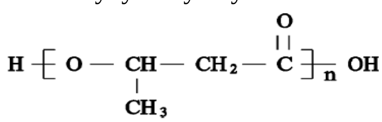
- **PGA**: Polyglycolic acid or polyglycolide:



- **PCL**: Poly-ϵ-caprolactone:



- **PHB**: Polyhydroxybutyrate



The selection is based on the determination of physical and mechanical characteristics as following:

- Polymer density (ρ, in g/cm^3)
- Tensile properties: tensile strength (σ, in MPa), tensile modulus (E , in GPa) and ultimate strain (ϵ , in %)

Specific tensile properties are obtained by dividing the original properties by the polymer density, leading to: specific tensile strength (σ^*, in Nm/g) and specific tensile modulus (E^*, kNm/g).

- Characteristic temperatures: glass transition temperature (T_g, in °C) and melt point (T_m, in °C)

Such characteristics are reported in Table 1a and 1b and in **Fig. 1(a)**, **1(b)**, **1(c)** and **1(d)**.

In **Table 1a** you can read the main characteristics of the four selected bio-polymers; in Table 1b there are the mechanical characteristics of the four bio-polymers as well and the desired characteristics you can get in case of a unidirectional composite lamina, based on flax fibers.

In **Figs. 1(a)** and **1(b)** you can find respectively the bio-polymers tensile strength and ultimate strain, while in **Figs. 1(c)** and **1(d)** it is represented respectively the specific composite strength and specific Young's modulus as function of flax volume fraction in case of a unidirectional lamina.

Among the presented biopolymers, PLA seem to score well on all the discussed properties:

- Polymer and composite densities are low;
- Degradation behavior and mechanical properties are acceptable and their melt points are almost ideal in order to produce fibers reinforced composites;
- PLA is already commercially available;
- PLA is used in 3D printers technology.
- The most commonly used process is polymerization. It is termed as ring-opening polymerizations of lactide with various metal catalysts (tin octane) in solution, melt form or even as suspension. This process results in metal catalyst combining with lactide to form larger PLA molecules.

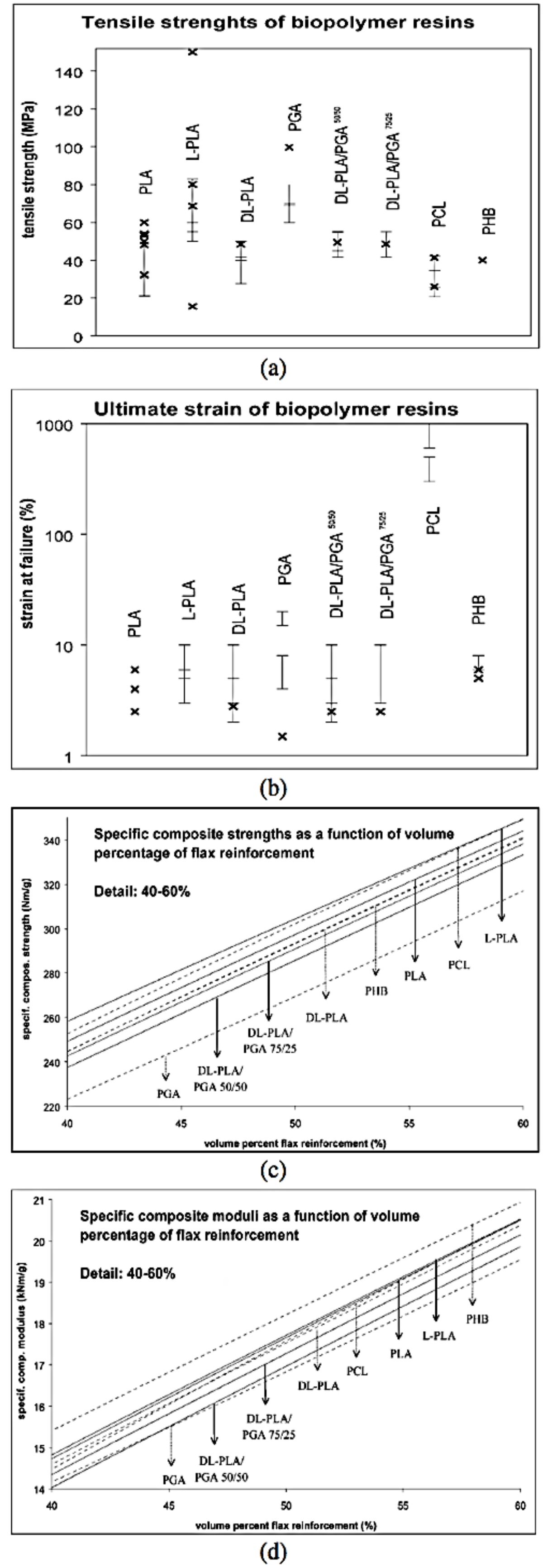


Fig. 1. (a) bio-polymers tensile strength; **(b)** ultimate strain; **(c)** and **(d)** specific tensile strength and specific Young's modulus.

Table 1. (a) and (b) - Bio-Polymers physical properties (a) and mechanical properties (b).

Properties	Limits	PLA	PGA	PCL	PHB
ρ (g/cm^3)	Upper	1.21	1.50	1.11	1.18
	Lower	1.25	1.707	1.146	1.262
σ (MPa)	Upper	21	60	20.7	40
	Lower	60	99.7	42	
E (GPa)	Upper	0.35	6	0.21	3.5
	Lower	3.5	7	0.44	4
ϵ (%)	Upper	2.5	1.5	300	5
	Lower	6	20	1000	8
σ^* (Nm/g)	Upper	16.8	40.0	18.6	32.0
	Lower	48.0	>45.1	36.7	33.9
E^* (kNm/g)	Upper	0.28	4.00	0.19	2.80
	Lower	2.80	4.51	0.38	2.97
T_g (°C)	Upper	45	35	−60	5
	Lower	60	45	−65	15
T_m (°C)	Upper	150	220	58	168
	Lower	162	233	65	182

(a)

Biopolymer type	Typical polymer properties			Composite (σ=400 MPa and E=23 GPa) properties			
				Based on desired σ		Based on desired modulus	
	σ (MPa)	E (GPa)	ρ (g/cm^3)	v_f (%)	ρ (g/cm^3)	v_f (%)	ρ (g/cm^3)
PLA	50	2.5	1.24	50.0	1.35	48.2	1.34
PGA	75	6.5	1.61	48.1	1.53	42.9	1.54
PCL	30	0.35	1.13	51.4	1.29	50.7	1.29
PHB	40	4.0	1.24	50.7	1.35	46.3	1.34

Typical flax properties: σ=750 MPa and E=45 GPa.

(b)

Table 2. (a) Some characteristics of natural fibers; (b) Mechanical characteristics of natural fibers, compared to synthetic fibers.

Fibre type		Length (mm)	Diameter (µm)	Moisture content (wt.%)	Cellulose (wt.%)	Hemi-cellulose (wt.%)	Lignin (wt.%)
Synthetic	Carbon HS	-	<17	-	-	-	-
	E-glass	-	<10	-	-	-	-
Fruit	Coir	20-150	10-460	8	32-43.8	0.15-20	40-45
	Cotton	10-60	10-45	7.85-8.5	82.7-90	5.7	<2
Bast	Flax	5-900	12-600	8-12	62-72	18.6-20.6	2-5
	Hemp	5-55	25-500	6.2-12	68-74.4	15-22.4	3.7-10
	Jute	1.5-120	20-200	12.5-13.7	59-71.5	13.6-20.4	11.8-13
	Kenaf	-	-	-	31-72	20.3-21.5	8-19
	Ramie	900-1200	20-80	7.5-17	68.6-85	13-16.7	0.5-0.7
Leaf	Sisal	900	8-200	10-22	60-78	10-14.2	8-14
Grass	Bamboo	1.5-4	25-40	-	26-65	30	5-30

(a)

Fibre type		Density (kg/m^3)	Price (USD/kg)	Young's modulus (GPa)	Tensile strength (MPa)	Elongation (%)
Synthetic	Carbon HS	1800–1840	124–166	225–260	4400–4800	0.0
	E-glass	2550–2600	1.63–3.26	72–85	1900–2050	1.8–4.8
Fruit	Coir	1150–1220	0.25–0.5	4–6	135–240	15–35
	Cotton	1520–1560	2.1–4.2	7–12	350–800	5–12
Bast	Flax	1420–1520	2.1–4.2	75–90	750–940	1.2–1.8
	Hemp	1470–1520	1–2.1	55–70	550–920	1.4–1.7
	Jute	1440–1520	0.35–1.5	35–60	400–860	1.7–2
	Kenaf	1435–1500	0.26–0.52	60–66	195–666	1.3–5.5
	Ramie	1450–1550	1.5–2.5	38–44	500–680	2–2.2
Leaf	Sisal	1400–1450	0.6–0.7	10–25	550–790	4–6
Grass	Bamboo	600–1100	0.5–0.5	11–32	140–800	2.5–3.7

(b)

In **Fig. 2(a)** is shown the global production of PLA, while in **Fig. 2(b)** and **2(c)** are reproduced the leaders in PLA production and the countries.

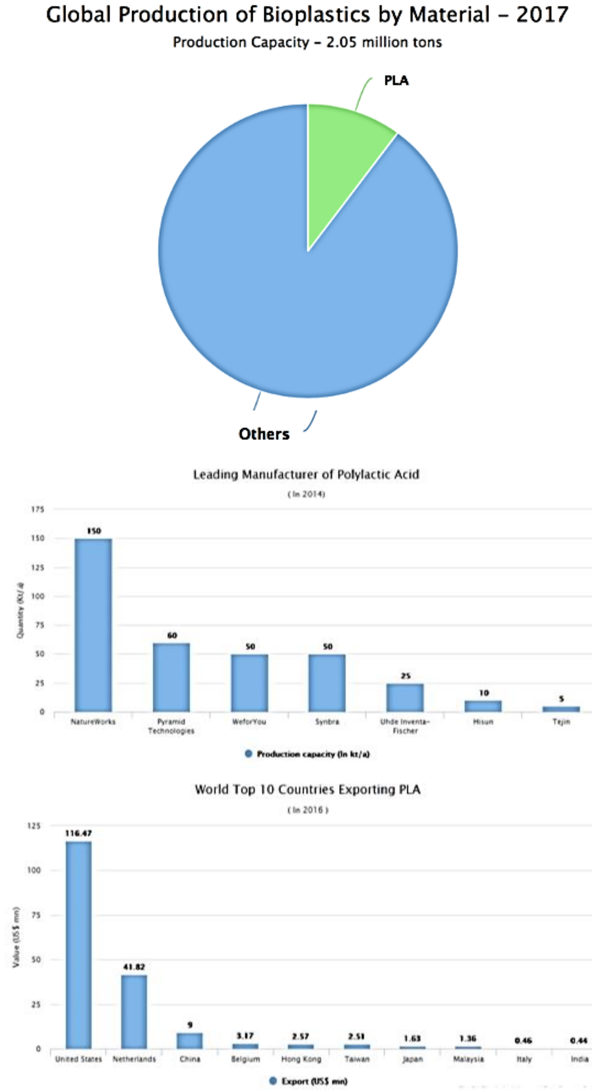


Fig. 2. (a) Global production of PLA; (b) and (c) Companies leaders in PLA production and countries.

Materials and properties: Natural fibers

The number of natural fibers worldwide is very impressive and of course, typologies and crops depend of environmental conditions of five continents.

For instance, Europe is interested to flax (France, Belgium) and hemp (Germany, Italy, Poland), India and Bangladesh have jute, China gives attention to bamboo, Brazil has an incredible variety of fibers, such as caruà [**13, 14, 15**].

Natural fibers have origin by minerals, (asbestos), by animals (wool, silk, spiders) and by plants. Among them, composite materials operators use the last one for their productions.

In **Table 2(a)** and **2(b)** there is the description of the main natural fibers studied worldwide, their main characteristics and the comparison with the glass fibers, obvious competitor for their applications.

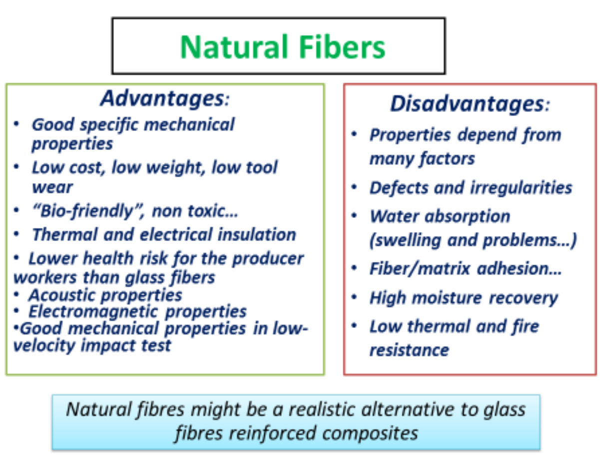
In **Fig. 2(a)** is shown the global production of PLA, while in **Fig. 2(b)** and **2(c)** are reproduced the leaders in PLA production and the countries [**19**].

In **Fig. 3(a)**, **(3b)** and **3(c)** are represented some plants of interest in Europe, Asia and USA: flax, hemp and bamboo, while in **Fig. 3(d)** there is the description of advantages and disadvantages concerning the use of natural fibers.

(a)

(b)

(c)



(d)

Fig. 3. (a) flax; (b): hemp; (c): bamboo; (d) advantages and disadvantages.

Green composites

As seen, polymer composites have been widely used for many years and their market share is continuously growing. Nevertheless, this also bring out one of the main their main limitations: the reuse and recycling of the different components is quite difficult.

It is often preferred to perform the direct disposal in a dump, or incineration.

Though, this way is often considered unsatisfactory because of the high cost, technical difficulties and environmental impact.

These problems have begun to be evident for about 10 years, leading the scientific research to look for new alternatives, able to replace traditional polymers with substitutes having lower environmental impact, referred as *green composites*.

The first attempts toward this class were composites based on recyclable polymers (e.g. polyolefins) filled with natural-organic fibers:

The use of natural-organic fillers allows a considerable reduction in the use of non-renewable resources.

These fillers are drawn from relatively abundant plants, therefore are very cheap.

They are much less dangerous for the production employees in case of inhalation, easy to be incinerated, lead to final composites with lower specific weight and allow obtaining interesting properties in terms of thermal and acoustic insulation.

Despite that, even these composites are not fully eco-compatible, since their recyclability has some limitations and their biodegradability regards only the filler.

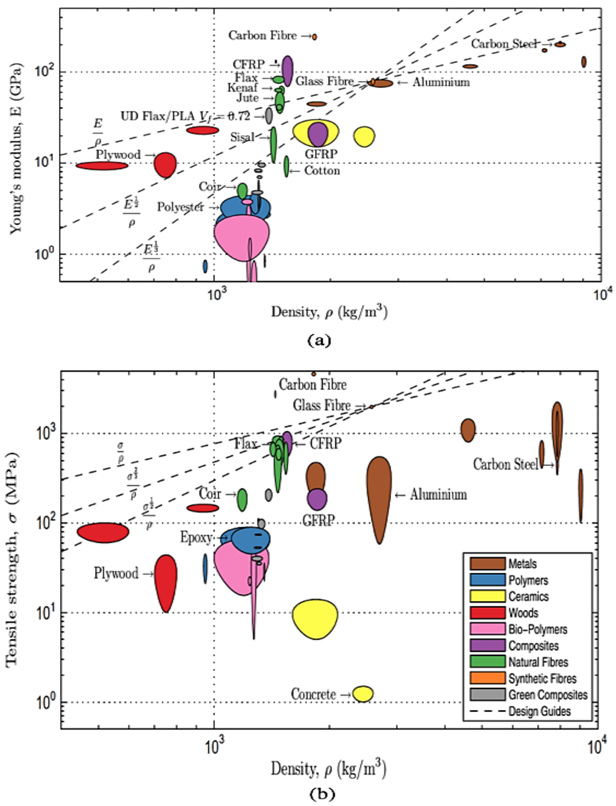
For this reason, research have been focused on the production on 100% eco-sustainable and "green" composites, by replacing non-biodegradable polymer matrices with biodegradable ones.

These bio-polymers are derived from a variety of renewable sources and include both thermosetting and thermoplastic polymers. These include:

- Poly(lactic acid) (PLA), as already seen
- Polyhydroxybutyrate (PHB), polysaccharides of plant origin
- Polyglycolic acid or polyglycolide
- Poly-ε-caprolactone
- Cellulose and Alginate, animal origin

In **Fig. 4(a)** and **Fig. 4(b)** the graphs show the position of green composites respect to other materials (4(a) Young modulus against density; 4(b) tensile strength against density).

The majority of traditional polymer matrix are derived from non-renewable petroleum which is formed from biomass over the course of 10^6 years when consumed as plastic products or fuel, it is usually converted into CO_2 within 1-10 years. Thus the use of this distinctly finite resource is unsustainable.



Figs. 4. (a) and (b) Young's Modulus and tensile strength against density.

This is a large incentive for pursuing green composites where both the reinforcement and matrix materials are derived from plants – usually in the span of less than a year.

Using renewable resources in this way: whereby the rate of CO_2 sequestered is balanced with the rate of consumption, contributes significantly to developing carbon neutral materials.

Materials are defined as biodegradable if they degrade through actions of living organisms.

Natural fibers are inherently biodegradable, as are many polymers; for example polyanhydrides and polyesters both degrade through hydrolysis but at significantly different rates: 0.1h and 3.3 years respectively.

Biodegradation is a desired quality it prevents accumulation of solid waste: green composites could allow composite materials to enter the market of products with limited service life, as their biodegradability could offer a serious advantage over synthetic composites.

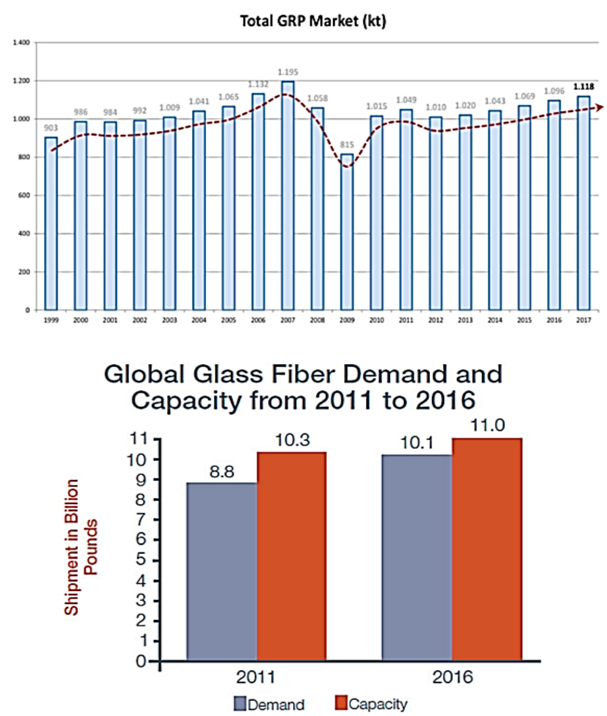
PLA degrade to carbon dioxide, water and methane within two months, whereas petroleum-based plastics require hundreds of years.

Main drawback is the poor durability. Exposure to environmental conditions can lead to a rapid degradation of the material: accurately predicting the lifetime of green composites is a major challenge to their widespread implementation.

Many labs are working on this item.

Recent studies have demonstrated how, by means of a novel combination of chemical and heat treatments under tension on karanja protein (KP) and mango starch (MS), tensile and thermal properties of sisal fiber can be enhanced. Which means it's possible to improve strength and modulus along a low decrease in fracture strain for both KP and MS composites. The cost of manufacturing of these composites would be low, as resins are derived from agricultural waste [**7**].

Some other modified fibers such as liquid crystalline cellulose (LCC) and micro-fibrillated (MFC) showed a good improve in tensile properties orientation and crystallinity, which led to advanced green composites. These kind of composites could be used in primary structural and would represent a green alternative to petroleum-based composites [**8**].



Figs. 5. (a) Overview of Composites Market; (b) Europe Glass Fibers Market.

Composites market

Since 1960, the U.S. composites industry has grown 25 times, whereas the steel industry has only grown 1.5 times and the aluminum industry is three times larger, according to market research firm *Lucintel* [**17, 20**].

In 2016, the U.S. composite materials market grew by 3.7% to reach 8$ billion in value. It is expected to reach 10.6$ billion by 2022, with a compound annual growth rate of 4.9%.

In Europe the composites industry is now enjoying its fifth successive year of growth. Production volume of GRP has been growing continuously during 2017. As in 2016, the European GRP market is expected to grow by 2% to an estimated total of 1.118 million tons.

GRP production in Europe continues to grow but is expected to lag behind the global trend, well over 2%.

Glass fiber, the predo minantly used reinforcement, is expected to reach 9.3$ billion worldwide by 2022, with a CAGR (Compound Annual Growth Rate) of 4.5% since 2016.

The global demand for clean energy and infrastructure upgrades also will help to boost glass fiber demand in the future (**Fig. 5(b)**).

Natural fibers composite material market is evaluated of the order of greater of 150 x 10^3 tons. Glass fibers, in consideration of the Mechanical and physical characteristics, represent the natural competitor.

There is a huge possibility for renewable composites to gain space in the international market.

Composites applications - automotive

Automotive body parts by composites are mainly composed by CFRP (Carbon Fiber Reinforced Polymers) and GFRP (Glass Fiber Reinforced Polymers):

- GFRP, CFRP, SMC, C/C for seat structures
- Formula 1 uses CFRP extensively
- Fuel Tanks made up of Kevlar reinforced rubber
- CFRP and Honeycomb composites for Chassis

Composites applications - Wind energy systems

The blades of the giant wind energy systems are mainly composed by CFRP and GFRP.

Composites applications - Aerospace

The aerospace industry continues to remain at the forefront of composite adoption, primarily due to the market's proactive shift toward light weighting to meet emission reduction goals and increase fuel efficiency.

Over the years, key aerospace OEMs understood the significant advantages of composites over metals in terms of design flexibility, vibrational damping and a high strength-to-weight ratio.

Therefore, have since used composites for primary load-bearing structures and high-volume components such as wings, fuselages, elevators, rudder, ailerons and nacelles.

Within the market, carbon fiber reinforced polymers have been the primary beneficiary of the industry's answer to light weighting.

The share of CFRP is likely to further increase in the coming years, owing to the tremendous potential to replace aluminum and steel in primary load-bearing structures.

Natural fibers composites - Applications

The applications of natural fibers composites in Europe started about the middle of 90's of the last century.

First market of importance is the automotive field, thanks to the European law which determined that each new car had to have a defined percentage of renewable material. In this sense, any FIAT, AUDI, MERCEDES vehicle has about 30 to 40 kg made by natural fibers (**Fig. 6(a)**).

In **Fig. 6(b)** are represented marine applications, while in **Fig. 6(c)** are illustrated many items built by natural fibers composites, such bamboo catamaran skulls, tables, chairs and honey comb for sandwich constructions [**2, 16**].

An application concerning rotorcraft interiors by means of replace E-Glass fiber with Hemp-fiber has demonstrated the advantage in weight [**9**].

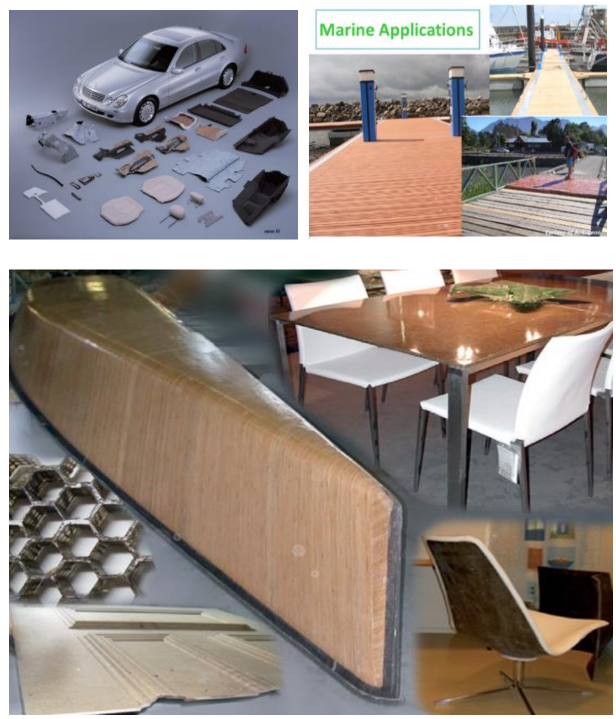


Fig. 6. (a) Natural fibers composite parts for E-class Mercedes car. (b) Natural fibers composites for marine applications. (c) Natural fibers composites for sandwich panels honey comb, hemp tables, flax chairs and catamaran bamboo skulls.

Natural fibers composites in aviation

The application of green composites in aviation is a very difficult item. The application of a new material in the fabrication of aeronautical parts needs complex, long and expensive procedures of certification, controlled by the Aeronautical Authorities [**3, 4**].

Of course, for the moment, the only possible application for the future is the fabrication of interiors parts (semi-structural parts), such as panels or caps.

For this reason, some parts have been built for Ultralight Aviation, because in such class of airplanes there is no certification.

Requirements to be demonstrated for the use of composite structures in interiors are:

- Mechanical characteristics:
  - Strength and stiffness in case of tension, compression, bending, torsion loads;
  - Good low-velocity impact behaviour;
  - Fire, smoke and toxicity (FST behaviour);
  - Crashworthiness;
  - Durability;
  - Low weight

Fabrication process consists on an about fully automated fabrication process. Fabrication environment is very clean, so the process is made in clean rooms, using only pre-preg materials or resin in form of sheets.

Classical fabrication process is lay-up in clean room, vacuum bag and autoclave.

Nowadays, in Europe the industry produces only flax and epoxy pre-pregs, in form of unidirectional tapes and tissues. The data concerning strength, stiffness, impact and weight are very promising, but FST behavior is not compatible with the necessary requirements.

Last December 2012, European project called "Cayley" brought together Boeing Research and Technology Europe (Madrid, Spain), Invent GmbH (Braunschweig, Germany), Aimplas (Valencia, Spain) and Lineo (St.-Martin du Tilleul, France). The aims are to industrialize environmentally friendly interior panels made with renewable polymers or recyclable thermoplastic sheets and natural fibers, namely flax.

Concerning the activities, Boeing produced flax sandwich panel made with epoxy, to be used for cabin sidewalls, while other partners investigated the difference between two different thermoplastic matrices, polypropylene (PP, synthetic) and polylacticacid (PLA, natural).

Flax fabrics have been treated with halogen-free flame-retardants and used to produce a full-scale sidewall panel for a 737 interior in a vacuum bag process. At laboratory scale, the bio-composite achieved compliance with FAA and EASA fire resistance requirements. In **Figs. 7(a)** to **7(f)** are represented respectively the use of composites on the airplane Boeing 787, the application of hemp/epoxy for the engine cover of the ultralight airplane MAG 1, caps and panels to be designed by green composites, CAYLEY project and interiors market previsions.

Market previsions for airplanes composite interiors

Market previsions for airplanes composite interiors are very interesting (**Fig 7(b)**).

Green composites applications

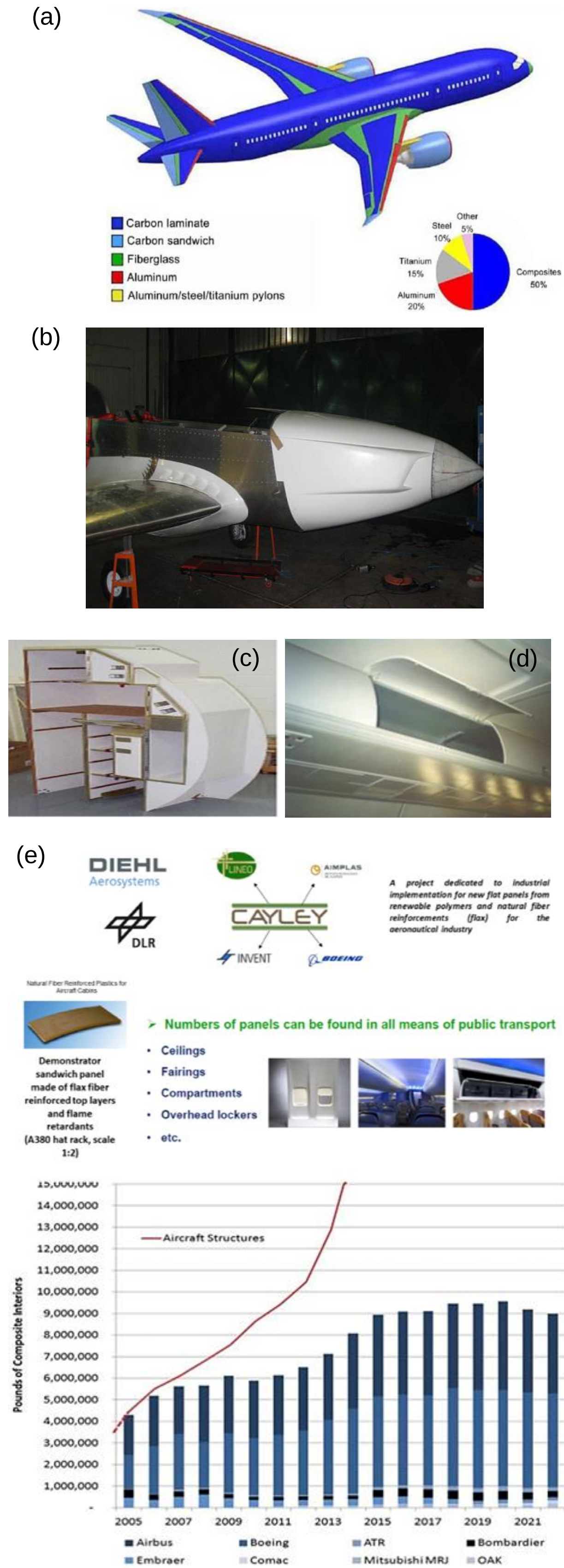
Green composite can be applied in the same fields described for natural fibers composites by synthetic resins, but in addition they can be used for short life-span products, typically thought of as those that are disposable, such as plastic cutlery and packaging, due to their easy disposal [**5, 6**].

In addition, let's consider also sporting equipment and biomedical applications [**11**].

Besides short life-span products, considered disposable, there are also items such as consumer electronics.

The company NEC has been working with green composites since 2004 when they used a PLA/kenaf composite in dummy cars in personal computers.

In 2006 NEC developed an 'eco' phone with the same material and in 2007 began using some green composites in the housing of personal computers.



(f)

Fig. 7. (a) Boeing 787; (b): Ultralight airplane MAG 1 engine cover; (c): panels for galleys; (d): airplane caps; (e): "Cayley" Project; (f): market previsions for airplanes composite interiors.

US mobile carrier Sprint declared in January 2012 that it would require all phones it sells to meet a minimum environmental standard.

Same year, a report by Juniper Research stated that 392 million ‘green’ handsets will be shipped by 2018.

Another potential product are toys, although no current example were found.

This shows that there’s both consumer and business interest in using materials with improved environmental credentials.

In the field of biomedical applications, the hydrophilicity of green composites facilitates interactions with other hydrophilic surfaces and substances such as living cell tissue.

This property of bioactivity, coupled with their biocompatibility and biodegradability, distinguishes green composites from their synthetic counterparts for use in biomedical applications.

Among their advantages we have:

- Superior mechanical properties without sacrificing weight
- Bio-degradation kinetics
- Cell permeability
- The possibility of incorporating other materials within the cell support such as growth factors or nutrients
- Shapability

One biomedical application for which green composites have demonstrated these advantages is the interdisciplinary filed of tissue engineering: specifically green composites have shown promise as scaffolds for soft tissue growth. Human mesenchymal stem cells (MSCs) can be seeded onto a scaffold and directed to differentiate down a desired cell lineage to grow tissues and organs. A trachea has been successfully grown using this technique and the subsequent transplant proved groundbreaking since the procedure allowed the patient an immunosuppressant-free life, unlike normal transplantees. The trachea was grown ex vivo on a donated extracellular matrix and differentiation was induced using growth factors.

Conclusions

In conclusion, green composites are the future of composites applications in many fields, thanks to their good mechanical characteristics, good thermal and acoustic insulation characteristics, low cost and environmental friendly aspects. The standards of natural fibers and bio-polymers are appreciable and in continuous evolution. Main competitor are the glass fiber composites, also because the price is low, but the increasing amount of natural fibers applied in structures will reduce the gap.

Future prospective

The rate of applications is estimated will be about 10% per year for the next five years. The improvement of the PLA characteristics will give the real possibility to have a complete renewable composite part in secondary structures, Some problems are in way of solutions, such as repetitiveness of the fibers characteristics, definition of trade marks, good interface between fibers and matrix, development of new green matrices, both thermoplastics and thermosets, the FST behavior compatible with the standards. Of course, new application in aeronautics must respect FAR and JAR standards. This means to improve a considerable effort in terms of time and money.

Acknowledgements

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