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State of the art on graphene lightweighting nanocomposites for automotive applications

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Abstract

This work presents a state of the art SoA of recent developments related to the lightweighting of automotive structure by introduction graphene as material constituents. It proposes some innovative directions in terms of technological manufacturing and process for a better understanding of the graphene contribution for enhancing multifunctional properties of nanocomposites. Likewise, the general context of research trends is presented along with the novel composites solutions for the design of lightweight materials using graphene-based polymer composites. The need for developing a modelling strategy is discussed in terms of simulation and optimisation of the multifunctional properties as well as the hierarchical modelling of hybrid composites. The large-scale production is presented through the existing process and manufacturing engineering. And finally, the industrial feasibility of such composite is presented by a demonstrator development and the life cycle analysis.

Keywords: Graphene, Graphene composites, Automotive applications, Materials processing, Life cycle analysis.

1. Introduction

The need for reducing motors engines pollution has been generalized since the carbon footprint become an important design parameter for improving the fuel economy of conventional, gasoline-powered automobiles. Electric-based vehicles, advanced combustion and fuels technologies have been developed to improve energy efficiency of cars and trucks. Indeed, the energy efficiency is supported by variety of technologies, among which the weight reduction through multifunctional materials is one of the most promising. They are essential for boosting the fuel economy of modern automobiles while maintaining their safety and performance. Because it takes less energy to accelerate a lighter object than a heavier one, lightweight materials offer great potential for increasing vehicle efficiency. The vehicle's weight reduction and material substitution in vehicle subsystems comprise a variety of techniques that range from improved design and the enhanced integration of components to the deployment of lighter, higher-strength materials. For such purposes, many automotive manufacturers have focused their design efforts on new technologies.

Lightweight materials are an important technology that can improve passenger vehicle fuel efficiency by 6–8% for each 10% reduction in weight while also making electric and alternative vehicles more competitive [1]. Therefore, the new generation of vehicles must be lighter, less polluting and more fuel-efficient. Their design should be developed aiming for individual mobility whilst also retaining safety, environmental friendliness and affordability [2]. These issues can be overcome by a strategy combining light structures and multifunctional materials. However, significant hurdles remain with respect to improved performance, manufacturability, cost, and modelling for such materials [1]. As a consequence, considerable materials science effort and new discovery need to be developed to overcome these hurdles. The discovery of the graphene with its interesting physical and mechanical properties have opened promising window for designing novel light composites.

Graphene is at the centre of an ever growing academic and industrial interest because it can produce a dramatic improvement in mechanical properties at low filler content [3]. Indeed, one of the most immediate applications for graphene resides in composite materials [4]. To take a full advantage of its

properties, integration of individual graphene sheets in polymer matrices is important. Exceptional physical as well as thermomechanical properties, a high surface/volume ratio and low filler content of graphene make it a promising candidate for developing the next-generation of polymer composites [5-7]. Graphene has been used to increase stiffness, toughness and thermal conductivity of polymer resins by a large margin [8-11]. However, many challenges, including the lack of constitutive material modelling for high performance structural applications can affect the final properties and applications of graphene composites.

This paper aims to bring a contribution to research efforts by analysing advanced nanocomposite materials based on graphene and their potential applications on automotive. To this end, the generalised use of composite materials in automotive is presented as well as the novel composite solutions based on the graphene polymer composite materials. A description of the constitutive modelling of graphene based polymer composite materials is presented along with the hierarchical modelling of hybrid fibres reinforced graphene based polymer matrix composites. The large-scale production is presented through the existing process and manufacturing engineering. And finally, the industrial feasibility of such composite is presented by a demonstrator development and the life cycle analysis.

2. Design of Lightweight Composite for Vehicles:

2.1. Novel Composites Solutions

There has been significant worldwide research previously undertaken on lightweight technologies for vehicles. Figure 1.1 depicts light-weight components targeted by today automotive manufacturers [12].

In Europe, most advanced industrial applicable research has been carried out in the SuperLightCar, Futura and MyCar projects, achieving up to 35% body in white BIW weight reduction. In the USA, the major department of energy DOE financed light-weighting material activity reports annually, and describes in detail which barriers to large scale adoption persist: costs that need to be reduced, high strain-rate deformation and failure not yet reliably modelled for latest generation materials, including carbon fibre-reinforced polymers CFRPs. In Japan Toray is collaborating with Mitsubishi Rayon, Honda and Nissan towards CFRPs cost reductions and subsequent car body / chassis applications (funded with €18.5 million by the Japanese government). In terms of material developments, it is clearly identified today that high performance fibre-reinforced polymers composites (FRPCs) have the potential to bring stronger weight savings. The combination of FRPCs with advanced metal alloys, known as "multi-material", remains an important R&D area that requires considerable further works. In addition, their design should be developed aiming for individual mobility whilst also retaining safety, environmental friendliness and affordability [2]. Figure 1.2 presents multi material lightweight vehicles MMLV in the design of the body in white BIW with 45% weight saving of the new Ford Mach II [13]

In recent years, a number of series production models were introduced to the market, serving to demonstrate the technical feasibility of applying the multi-material BiW approach to lower production volume, high-performance vehicle segments. Examples include the Porsche Panamera, the Audi TT, latest Daimler E-class and the BMW5, i3 and i8 series. In USA, where the vehicles are typically significantly heavier than those developed in Europe, substantial public resources are being allocated to fund Research and Technological Development in the area of materials and technologies to reduce vehicle weight. The SotA is characterised by the ever-present trade-off of costs vs. weight. Generally speaking, the benchmark for the BiW of a typical 2011 compact car is around $\epsilon 4/kg$ for the complete assembled BiW. This very low cost per kg of such a complex assembly of hundreds of parts represents the starting point for feasibility assessment for any lightweighting initiative aimed at mass-

volume production (e.g. around 100 vehicles per day). Assuming that a higher cost per kg of weight saved is acceptable in this case, the aim is to strive to achieve a package of weight-saving measures that allow for the entire structure remaining well below ϵ 6/kg when considering large-volume, mass market-oriented vehicles. Such extremely hard-to-meet cost targets represent the principal barrier to the adoption of advanced lightweight material concepts today. Multi material lightweight vehicles MMLV in the design of the body in white BIW with 45% weight saving have been achieved at cost. Recent presentations of BMW's i3 and i8 vehicles, that reached the market in 2013, reinforce the need to extend the progress that has been made in Europe by conducting pre-competitive, collaborative research. The BMW i3 vehicle, which is between a Mini and a VW Golf in size, has a relatively high price tag (between 30- ϵ 40k depending on the country, the incentives and the taxes) since their production volumes are expected to be limited to tens of thousands of units per year in comparison, by targeting volumes of hundreds of thousands per year (i.e. mass production).



Figure 1.1. Light-weight components targeted by today automotive manufacturers [12]



Figure 1.2. Multi Material Lightweight Vehicles MMLV [13]

In fact, the growing trend to substitute conventional steel and cast irons in vehicles for lightweight purpose had led to the development of automotive components with lighter materials. Among them, conventional materials, for instance aluminium or magnesium alloy and ultra-high strength steel, are used in the engine block, cylinder heads as well as in transmission, cases, valves bodies and channel plates [14]. Beside these materials, others categories show most promise, for instance, fibre-reinforced polymer composites (including carbon and glass fibres) and advanced polymers (without fibre reinforcement) [1].

Jaguar Land Rover, which switched to all-aluminium bodies in 2009, led the way with aluminium construction with the XJ and XK models. Ford has already replaced certain heavier truck platforms with unibody designs, and is also using advanced high-strength steels, aluminium, magnesium, natural fibres and nano-engineered materials to reduce vehicle weight. Moreover, Ford has stated that the platform for the 2015 F-150 model will weigh up to 340 kg less than the current generation [15]. For 2012 model, Ford's combined car and truck fleet achieved an average of 8.5l/km, up from 7.8 l/km in

2011 [16]. General Motors plans to cut vehicle weight by 15 % by MY 2016 by investing in more extensive use of advanced materials such as aluminium, high-strength steel and carbon fibre-reinforced plastics [17]. Chrysler is using high-strength steel extensively in the Dodge Dart and Fiat 500L [18]. The 2013 SRT Viper, with a lighter steel frame and a body that is entirely constructed out of aluminium, plastic composites and carbon fibre, is roughly 100 pounds lighter than the previous iteration of the Viper. VW's Audi marque has focused on exploring the potential of aluminium and fibre-reinforced composites, among other advanced lightweight materials [19]. BMW and Toyota also recently agreed to cooperate on the development of lightweight technologies such as reinforced composites for vehicle bodies. It appears clearly that replacing cast iron and traditional steel components with lightweight materials such as high-strength steel, magnesium alloys, aluminium alloys, carbon fibre and polymer composites can directly reduce the weight of a vehicle's body and chassis by up to 50%. However, significant problems exist with regard to improved performance, manufacturability, cost, and modelling. The following hurdles are identified with regard to advanced materials used in the automotive weight reduction:

Advanced high- strength steels (AHSS)	Aluminium Alloys	Magnesium Alloys	Fibre-Reinforced Polymer Composites	Advanced Composites
No microstructures for adequate strength-ductility trade-off	Limited formability. Casting of complex parts difficult	Low formability of sheets. Casting of complex parts difficult	Long cycle times for processing	Long cycles times in processing
Susceptible to local failure	Relatively high costs	Higher costs than aluminium	High cost of carbon fibres	Precursors not yet mature
Complicated modelling for forming	Insufficient strength/stiffness	Insufficient strength/stiffness	No reliable modelling	Susceptible of degradation

Table 1.1: Identified hurdles for advanced materials

These technical hurdles can be overcome by providing new materials science efforts based on graphene which has attracted both academic and industrial interest because it can produce a dramatic improvement in properties at low filler content.

2.2. Graphene-based Polymer Composites

Graphene (and its derivatives-based polymer composites) has shown huge possible applications in several fields, including aerospace, automobile, defence, electronics and energy industries, etc., thanks to its extraordinary reinforcement in composites. To take full advantage of its properties for applications, integration of individual graphene sheets in polymer matrices is crucially important. Many factors, including the type of graphene used and its intrinsic properties, its dispersion state in a polymer matrix and its interfacial interactions, the amount of wrinkling in it, and its network structure in the matrix can affect the final properties and applications of graphene-based polymer composites [20]. Thermosetting polymer resins are widely used in applications as diverse as aerospace structures, coatings, automotive components, adhesives and electronic components, but many of these are limited by the strength, stiffness, toughness, electrical conductivity or thermal performance of resins currently available. Primary composite aircraft structures could, for example, be lighter if the current limitations of resin toughness and low conductivity were improved. Graphene has been shown to significantly improve stiffness, toughness and conductivity of thermosetting polymer resins [21]. In a recent review, Galpaya et al. [22] compared mechanical-property improvements afforded by graphene, functionalised graphene and graphene oxide with those of other nanofillers such as carbon nanotubes (CNTs), silicon dioxide (SiO2), titanium dioxide (TiO2) and aluminium oxide (Al2O3). They report a 65% increase in fracture toughness upon adding just 0.125% of graphene (produced by thermal annealing). To achieve similar improvements would require a 120-fold higher weight fraction of SiO2, a 30-fold higher weight fraction of Al2O3 and a 60-fold higher weight fraction of TiO2. For CNTs, the best reported increase in fracture toughness is only 48% and this is only achieved with a 4-fold higher weight fraction than that which delivers the 65% increase in the equivalent graphene-epoxy nanocomposites.

Despite these impressive figures, however, graphene-reinforced thermosetting resins are yet to find their way onto the market for large-scale applications due to a combination of high graphene cost, not constant reproducibility and standardisation of graphene based material supplied, lack of proven large scale dispersion techniques and the fact that property improvements have yet to be proven beyond the laboratory scale.

Though, several technologies are embedded in the next generations of multifunctional Graphene-based composites, there are still a lot of technological challenges to overcome, particularly in the area of the type of Graphene used and its intrinsic properties, the dispersion state of Graphene in the polymer matrix and its interfacial interactions, the amount of wrinkling in the Graphene, and its network structure in the matrix can affect the final properties and applications of Graphene-based polymer composites [20]. Hence, the present challenges for researchers are in the development of lightweight, high-performance, cost-effective and multi-material solutions:

- Lack of new methods of large scale production of Graphene based products mechanical exfoliation is not scalable to an industrial process;
- Lack of new methods of functionalization;
- Investigation of the exfoliation process of Graphene based material during the process;
- Expected low ductility of Graphene-based composites structure. Considering implementation on several vehicle components (i.e. front end), this will lead to high vehicles' deceleration, which minimising the vehicle safety;
- Insufficient knowledge on attainable strength/stiffness of Graphene thermosets/ thermoplastic polymer composites;
- No existed materials model on commercial explicit finite element software to model Graphene based composite materials for high performance structural applications;
- Graphene-based composite material characterisation and modelling still not fully investigated especially with regard to automotive applications and different loading conditions;
- Lack of knowledge on Graphene composites for high performance structural applications and interface properties between the Graphene and polymer matrix under severe loading condition (i.e. fragmentation and crash);
- Preparation of automotive composites-Lack of knowledge on how to design in Graphene composites automotive structures that can offer high stiffness, strength and predictable and safe failure modes;
- Nowadays vehicle and body architectures do not usually take advantage of the essential qualities of new composite materials;
- Some approaches to joining and bonding of Graphene-based composites parts insufficiently covered by simulation and modelling tools; no automotive experience available;
- The joining of dissimilar materials is not covered by an appropriate know-how and several critical points are not yet solved by the scientific community and researcher;
- Great attentions focused on embedded CO₂ in overall LCA within lightweighting process; however, no solid info on how to evaluate pro's and con's inside design process.

2.3. Large scale-production of graphene-based products.

Graphene promises to be the ultimate nanofiller having outstanding and often unsurpassed electronic, mechanical and thermal properties. However, to date true commercial applications have yet to be realised or implemented due to lack of understanding of material and its dispersions under melt conditions. Melt processing is crucial in determining the performance of the final consumer product. Comparable length-scales between graphene nanofillers and polymer chains provide a new challenge for composite formulation and processing: strong flows impact stretching of polymer chains and ordering, orientation and dispersion of the nanofiller. Control of these nanoscale phenomena by combining process-engineering technologies with new knowledge and methodologies from chemical and physical sciences provides a platform for realising the commercial potential of graphene nanocomposites: enhanced mechanical, anti-static and barrier properties would deliver consumer benefits through better product performance and extended product life and business and environmental benefits through less raw material being consumed and transported. Graphene's in commodity polymers should deliver consumer products with improved mechanical, thermal and electrical properties. Realising this requires a fundamental understanding of graphene's behaviour in polymer matrices, and its effect on polymer processing.

Graphene-related materials (GRMs) can be produced basically by different methods: micromechanical exfoliation (small-scale production, high cost), chemical exfoliation, chemical reduction of graphite oxide (low purity, high defect density, wastes in production), chemical vapour deposition (moderate scalability, high cost and high process temperature) and epitaxial growth on SiC (low yield, high cost, high process temperature, very expensive substrates). The outstanding characteristics of graphene (electrical conductivity, mechanical strength) were achieved in principle for mechanically exfoliated graphene and deposited on special substrates, but for many applications in composite materials, requiring high scalability and reduced price, high degree of purity and low defect density are not a major issue. In this contest, exfoliation techniques have a potential for large-scale production of low-cost graphene. Some of the key developments could be a microwave-assisted exfoliation technique, intercalation and exfoliation of graphite flakes with the aid of gases, a mechanical exfoliation technique for large-scale production of graphene nanoflakes by controlled ball milling of graphite flakes in a liquid medium or continuous rubbing of solid graphite block against rotating glass substrates in a solvent while simultaneously subjecting it to ultrasonication treatment. Challenges for new graphene supplier are strictly related to synthesis of a right material for a right application at affordable costs. The key is surely the possibility to tailor GRMs production towards final applications, optimizing as much as possible all the process steps (raw-materials selection, process steps and dispersion procedure) and the way to manage GRMs during each successive processing steps.

A key consideration when targeting the large-scale production of Graphene nano-composites is the extent to which we can extrapolate the laboratory scale results in order to predict the properties of Graphene-reinforced polymers produced on a large-scale. Often the laboratory scale composites are produced using "ideal" Graphene i.e. known to contain only one or two layers. In reality, as we look to scale the Graphene production process, the likelihood is that we will produce material with a distribution of layer thicknesses. The question then is: what effect does the number of layers have upon the properties of the composite? This has been studied recently by Gong et al. [23], who used Raman spectroscopy to firstly characterise the Graphene (in terms of number of layers) and also to monitor its subsequent deformation during atomic force microscopy (AFM) nano-indentation experiments on polymer composites, by utilising stress-induced shifts in the Raman bands.

This method has previously shown that individual flakes of Graphene have the capability to reinforce a polymer matrix but, interestingly, the work by [23] has shown that bi-layer Graphene would be equally as good as monolayer Graphene, whilst tri-layer Graphene would only show a 15% reduction in reinforcing efficiency. Furthermore, it is only when the number of layers is >7 that the reinforcing efficiency of Graphene falls to less than half that of monolayer Graphene. If we consider the Graphene interlayer spacing in relation to the dimension of the polymer coils (which would

separate the layers in a polymer composite), however, it can be shown that higher volume fractions of Graphene can be obtained for the multi-layer material than for monolayer Graphene. Therefore, there is a balance to be struck between the ability to achieve higher loadings and the reduction in reinforcing efficiency as the number of Graphene layers is increased.

In addition, the best method of producing Graphene to be introduced in composites for industrial applications is the reduction of graphite oxide in controlled conditions. The chemical or thermal reduction of Graphene oxide can produce Graphene with a large number of defects, reducing the quality of the final product and worsening the expected properties. Without introducing functional groups, reduction of graphite oxide produces Graphene with different percentages of oxygen, a good quality Graphene need to have less than 2.5% of oxygen and preferable less than 1% of oxygen. However, the presence of oxygen helps to introduce new functional groups through chemical reaction such as covalent reactions (nucleophilic substitution, electrophilic substitution, and condensation and addition reactions) and non-covalent reactions [24]. The direct chemical reaction in pristine Graphene involves the use of powerful chemical reactions like radical reactions or the use of dienophiles to react with the C-C bond [25].

3. Modelling, Simulation and Optimisation of Multifunctional Properties

The number of theoretical/numerical works published on Graphene-based composites has so far been limited. The understanding of deformation and fracture of graphene reinforced polymer composite materials GPCMs and fibres reinforced GPCMs denoted FGPCMs under conditions typical for high-performance structural applications is still in their infancy. There are limited established theoretical studies available in the technical literature on their dynamic and crash analysis, and fracture and failure behaviours under severe loading conditions [26], typical for automotive applications

A variety of techniques could be used in modelling the graphene-based composite materials; however, the clear integrations between these techniques are still not clear for some applications (i.e. high performance structural applications). Much of the previous works has focused on modelling the elastic modulus at a set temperature. Since temperatures vary widely in space, a temperature-dependent model is required for the elastic modulus. There are two main approaches taken when modelling graphene composites. The first focuses on the molecular level interactions. Molecular models use simulations to determine local interactions among atoms or chemical reactions between the matrix and nanotubes [27]. Molecular models are limited to very small systems due to their high computational cost. The second is continuum modelling which considers overall deformations. Continuum models include the Mori-Tanaka model, rule of mixtures, and the Halpin-Tsai model [28]. Multi-scale models, which combine molecular and continuum models, have also been developed. Models by Wang et al. [29] should be cited.

Examples of existing works in this field are, for example, the molecular dynamics-based simulation techniques employed by Awasthi et al. [30], who studied the load transfer mechanisms between polyethylene and a graphene sheet. Cho et al. [31] employed a Mori-Tanaka approach to study the elastic constants of nano-composites with randomly distributed graphene sheets. Most recently, Montazeri and Rafii-Tabar [32] developed a multi-scale finite element model to study the elastic constants of a Graphene-based polymer nano-composite. Parashar and Mertiny [33] also proposed a multi-scale model using finite elements to characterise the buckling phenomenon in Graphene-based polymer composites.

Besides, the modelling aims at developing and applying methods, algorithm and tools for supporting the graphene-based polymer composites simulation. This procedure is depicted by Figure 1.3. Specifically, the goal of the modelling is connected with the development of a multi-scale modelling of graphene-based polymer composites for automotive light-weighting and crashworthiness purposes. The material selection and thus their constitutive law is accounted for by a comprehension derivation of their mechanical properties. Indeed, a starting point of such modelling will focus on the derivation of GPLs' mechanical properties. At the atomistic scale, the graphene sheet is considered undergoing non-linear deformations [34].



Figure 1.3. The multiscale modelling approach (MMA)

For the polymer matrix, a elasto-plastic behaviour is selected to account for the material nonlinearities. Thereafter, the progress toward improved **crashworthiness simulation** is addressed by an integrated numerical methodology for predictive modelling of **damage and failure** through a multiscale modelling [35] based on the constitutive models including the process history and the material state at required scale during the loading.

4. Process and Manufacturing Engineering Graphene-based Polymer Composites

Several manufacturing techniques have been used to elaborate the graphene-based polymer composites. Among them, strategies for surface modification of graphene with polymers are focused on covalently or non-covalently modified graphene. In addition, a series of effective processing routes for producing high-quality graphene-polymer nanocomposites, such as melt compounding, solution blending, in situ polymerisation, latex mixing and electro-polymerisation have been developed [36, 37]. However, two main challenges, namely the dispersion behaviour and the interfacial interactions between the graphene and polymers, should be overcome. Indeed, the surface modification of graphene with polymers is used to deal with such challenges. It is based on the covalent modification which gathers all "grafting to" method such as amidation and esterification reactions, nucleophilic ring-open and substitution reactions. The "grafting from" method consists in atom transfer radical polymerisation (ATRP), reversible addition fragmentation chain transfer (RAFT) polymerisation, ring opening polymerisation (ROP) and in-situ condensation polymerisation. The non-covalent modification is based on the weak interactions between the used stabilisers and graphene. Its main modes of action are π - π interaction, electrostatic force and hydrogen bonding as well as π - π interaction and hydrogen bonding.

For fabrication purposes, thermoplastic polymers like polyethylene terephthalate PET or polyamide 6 (PA6) are combined with surface functionalised graphene in appropriate mixing methods like melt compounding, compounding with TRGO, compounding with functionalised GO (fGO) or solution blending, blending polymers with RGO, blending polymers with functionalised graphene.

This process aims to elaborate the graphene-based composite by an appropriate selection of suitable materials (both polymer matrix and graphene-related materials as low content filler). Thermoplastics present a number of advantages to parts manufacturers over their more commonly used thermoset counterparts. They harden when cooled yet retain their plasticity; they re-melt and can be reshaped by reheating them above their processing temperature. This makes thermoplastics much easier to recycle than thermosets. In addition, once melted, thermoplastics harden quickly at relatively low temperatures, meaning that reinforced thermoplastic parts could be **produced rapidly in short cycle** times. They tend to be lighter than thermosets, they are highly resistant to impact and they stand up well to strain. Due to the **recyclability** [38] and interesting properties for structural applications, a preference is given to thermoplastic polymer matrix. Among them poly-arylene ether nitrile (PEN) or polypropylene (PP) or polyamide PA6, polybutylene terephthalate (PBT) have been already used in automotive applications. GPLs will be derived from the so-called "top-down" approach which demonstrates a great opportunity for large scale production for composite applications. Indeed, manufacturing will use the reduction of graphite into graphite oxide by a modified Hummers method. This procedure is followed by the exfoliation of graphite oxide. Exfoliation can be done by ultrasonication of graphite oxide in water leading to a colloidal dispersion of individual graphene oxide nanosheets. They represent graphene functionalised with hydroxyl groups and remain electrically insulated to its parent graphene. Finally, the graphene oxide nanosheets are chemically reduced to graphene also called reduced graphene oxide (RGO). In addition, a characterisation can be use X-ray photoelectron spectroscopy to quantify the oxygen's degree leftover of graphene. The above reduction of graphene oxide can be implemented by environmentally friendly techniques, for instance, photoreduction and hydrothermal dehydration as well as catalytic/photo-catalytic reductions. Also, the elaborated GPLs can be characterised using microscopy methods as well as spectroscopy tools for investigating the GPLs' structure at different length scale. This analysis will be carried out by atomic force microscopy (AFM). Spectroscopy tools will enable to quantify the energy/momentum distribution within the GPLs and based on Raman spectroscopy and Auger spectroscopy as well as Xray photoelectron spectroscopy. For two-phase composites (GPLs/polymer matrix), advantage can be taken of the high-density surface defects of GPLs that are derived from graphite oxide as filler within polymer matrix for providing high-energy adsorption sites for polymer chains. The process technology for achieving uniform dispersion of GPLs within the matrix will be based on the solution mixing via ultra-sonication followed by high speed shear mixing as dispersion technique to produce GPLs in situ directly in epoxy matrix.

5. Hierarchical Composite FGPCMs

Hierarchical advanced graphene-based polymer composites constitute nowadays efficient materials to replace conventional composites in structural applications. In fact, hierarchical glass-fibre-reinforced graphene nanoplatelets polypropylene composites [39] have shown that the combined effect of the two fillers of rather different size scales, e.g., micro- and nanoscale, can lead to significant improvement of the tensile modulus and impact strength, while the dispersion of the nanofiller in the PP matrix promoted the formation of a stronger interface between the matrix and GF. Moreover, carbon-fibre and GPL-reinforced poly-arylene ether nitrile (PEN) PEN/CF/GPLs composites [40] have demonstrated the synergic effect of combining reinforcements to deliver excellent mechanical properties 1.7, 4.5 and 6.4 times larger than those of PEN/CF composites, PEN/GPLs composites and PEN host, respectively.

For an innovative design, it is proposed here that the hierarchical composites **FGPCMs** are prepared by an economically and environmentally viable method of melt-mixing and moulding melt-mixing thanks to its simplicity and compatibility with existing industrial polymer-processing techniques extrusion and injection moulding. A first step in that process concerns the two-phases composite. It is related to the dispersion of GPLs inside the polymer matrix. Then, the GPLs/polymer blend can be painted layer-by-layer on the fibres. A second way is the use of graphene sheets GNPregs as bulky material for direct production of nanolaminate composite materials. Therefore, it is possible to design the composite laminate based on the fibre plies. The obtained fibre plies could be laid up and wetted with the GPL/polymer. Finally, a vacuum bag is placed over the system and the sample is allowed to cure for 24 hours at room temperature followed by another high-temperature cure in oven at 90°C during four hours.

6. Sustainable Technologies and Processes

Among challenges on the way of CFRPCs into production vehicles are high-speed manufacturing systems providing hundreds of thousands of units per year part counts. To achieve such a target, existing production processes have been optimised by car makers. Indeed, BMW has used the resin transfer moulding (RTM) of carbon fibre/thermoset composites for the 2013 i3 and 2014 i8 models. However, the cycle time, which is an important parameter, remains unknown and less documented. 10 minutes to de-mould is reported for that process. Plasan Carbon Composites and Globe Machine have developed a pressure press moulding process for a carbon fibre/thermoset composite roof and hood for 2014 Corvette Stingray with a cycle time of 17 min part-to-part, while a cycle time of 10 min partto-part was obtained by Gurit using the compression moulding for carbon fibre/thermoset composite parts. The most promising cycle time was obtained for fibre-reinforced thermoplastic composites - 60 s part-to-part - developed by Teijin/General Motors using the compression moulding process. By capitalising on the advantages that thermoplastics present over the thermoset matrices in terms of cost and recyclability, Toyota/Toray claimed to be the first mass-produced vehicle to feature a carbonfibre-reinforced thermoplastic (CFRTP) structural components with the Mirai model using a press moulding process. Another important research development after the manufacturing process is the joining technologies. Joining solutions to combine parts made of polymer and metal are existed. However, they are either not cost effective enough to apply them in mass-produced economy vehicles, or their performance does not allow using them beyond semi-structural applications. Moreover, with the increase of lighter-weight materials, traditional joining techniques face new challenges. For instance, heat input, which becomes a much bigger problem for fibre-reinforced polymer composites. As a result, lower temperature techniques are often needed.



Figure 1.4. Manufacturing technologies (MT)

Herein, it is proposed a manufacturing technology shown by Figure 1.4. It is based on a high degree of automation, short cycles times, good reproducibility and an excellent dimensional stability. Indeed, **it aims** to design components with hierarchical graphene-reinforced polymer composites based on the latest low-temperature and hybrid joining processes. Adhesive bonding and mechanical fastening (folding and riveting) processes as well as ultrasonic welding can be investigated for thermoplastic to melt plastic locally and press parts together. For structural applications, the adhesive bonding presents several advantages of connecting dissimilar materials without re-melting or altering the mechanical properties of joined materials. Moreover, **this technology aims to address** the hybrid joining techniques. They are based on the combination of adhesive bonding and structural joining process such as self-piercing rivets or spot welding to provide additional strength until complete curing and overall high stiffness in the final vehicle construction and also to reduce the "light-weight index" that measurers weight in relation to the overall mass of the vehicle.

7. Industrial Feasibility and Demonstrator Development

Innovative materials such as CFRPs are nowadays generalised in the concept of new automotive. They are combined with conventional lightweight materials to address interior as well as exterior applications. For instance, the architecture of the BMWi3 comprises two elements: aluminium drive module, which incorporates the powertrain, chassis, battery, and structural and crash functions and the Life module, or passenger cell, made from CFRPs.

The industrial feasibility is oriented to interior as well as exterior applications by developing new FGPCMs based on thermoplastic/thermosetting matrices with higher stiffness and lower density. These composites are expected to withstand structural loads at low temperature enabling the exploration of new interiors applications oriented to IP carriers, which includes seamless tear seam for airbag, knee bolster and centre console. The new FGPCMs are oriented to take into account occupant safety in the event of any crash based on the development in material development and modelling. FGPCMs with a thermoplastic matrix will also address exterior components, for instance fenders, exterior door panels, tailgates, tank flaps, claddings and spoilers. Depending of the body panel applications, the demonstrator development deals with the appropriate manufacturing feasibility and process method. In addition, the demonstrator development encompasses the BIW stage as well as the assembly. The BIW area will follow the process stage. In the assembly shop, the BIW will meet its engine leading to a gradually completion of the vehicle by fitting the wheels, the brakes, the seats, the dashboard, the steering wheel, the lights, the electrical system and the on-board technologies. The cost-effective analysis will be used as predictive engineering tools to perform detailed industrial manufacturing during the design phase and before tooling. It will be carried out by solving the processing challenges and help in identifying the appropriate choice of materials for an easy processing. This strategy will be based on virtual process technologies (for instance, injectionmoulding) consisting of pre-processing, analysis and post-processing in the goal to achieve optimum results.

8. Life Cycle and Economic Analysis

Among key regulations related to the sustainability of the automotive industry, the weight reduction of car and recycling technologies is a massive priority and key trend. Automotive segments face major sustainability challenges regards to the invention and employment of appropriate technologies that enables to shift from non-renewable to renewable resources for energy and materials. These trends are mainly concerned with (i) light-weighting to improve fuel efficiency and reduce carbon emissions, (ii) deployment and use of renewable sourced materials and (iii) efficient recovery, reuse and recycling at the end of life of automotive.

Several studies have demonstrated that 85% of the automotive greenhouse gas emissions occur in the use phase, and about 10% in the production phase and about 5% in the end of life or recycling phase [41]. It has been proven that light-weighting reduces carbon emissions during the use phase and hence represents the most impactful action in the reduction of carbon footprint of an automotive [42]. Today, Life cycle analysis LCA tools are often used at the end of the design work and therefore it happens that the chances to improve products and/or technologies in an early design stage, where changes are still possible with minimum costs, are missed. if LCAs take place too early, future changes in important pre-chains like energy supply leading to a different picture at the time of series production are not correctly considered. Today's LCA approaches tend to consider environmental impacts for production of materials, manufacturing of parts, utilisation phase and end-of-life for vehicle.



Figure 1.5. LCA approach

As an innovative approach, LCA considers prospective boundary conditions, as shown by Figure 1.5, meaning that it will include forecasted conditions for the series production in future by applying estimated energy supplies, e.g. for the years 2020 and 2030. Especially for light-weight and graphenebased composite materials, this may have significant influence on the overall results, as, according to experience, good performance of light-weight materials in the utilisation phase increases the relevance of the production phase in the overall life cycle. Another novel approach, beside production of materials and manufacturing of parts, is that LCA will consider the automobile's end-of-life (EOL), meaning the recycling processes and the influence of changes due to composite materials studied in previous FP7 projects (ALIVE, Evolution, Urban-EV, etc.) will be included in the analysis by the partners. In addition, light-weighting with FGPCMs is the target for reducing the carbon footprint of automotive. This methodology will be totally based on the cradle-to-grave approach, i.e. a closedup loop production. A critical factor for design for sustainability will be the material selection. Thus, the light-weighting as a concept will address mainly two elements (i) the design for resource conservation, e.g. to minimise material usage and (ii) the design for energy efficiency. The above material selection will be used as input database inventory for a developed software tool that will encompass an algorithm based on environmental impact and integrate individual component of the FGPCMs within the software core.

9. Conclusion

Lightweighting becomes an important issue for energy efficiency in automotive. It arises the need for developing a novel generation of materials that will combine both weight reduction and safety issues. Throughout this work, the applicability of graphene-based polymer composite materials is discussed regards to the fulfilment of these requirements. For such a composite, open challenges concerning graphene reinforcements are addressed in terms of materials modelling as well as the process and manufacturing engineering. They are also related for instance to the large-scale production based on exfoliation techniques. From modelling view point, this work presents strategies to overcome the above limitations by developing appropriate constitutive models to integrate the macro-scale behaviour. These strategies bind combination of several techniques form Molecular mechanics to Continuum mechanics. Finally, the developed constitutive is candidate for an implementation within a finite element code.

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