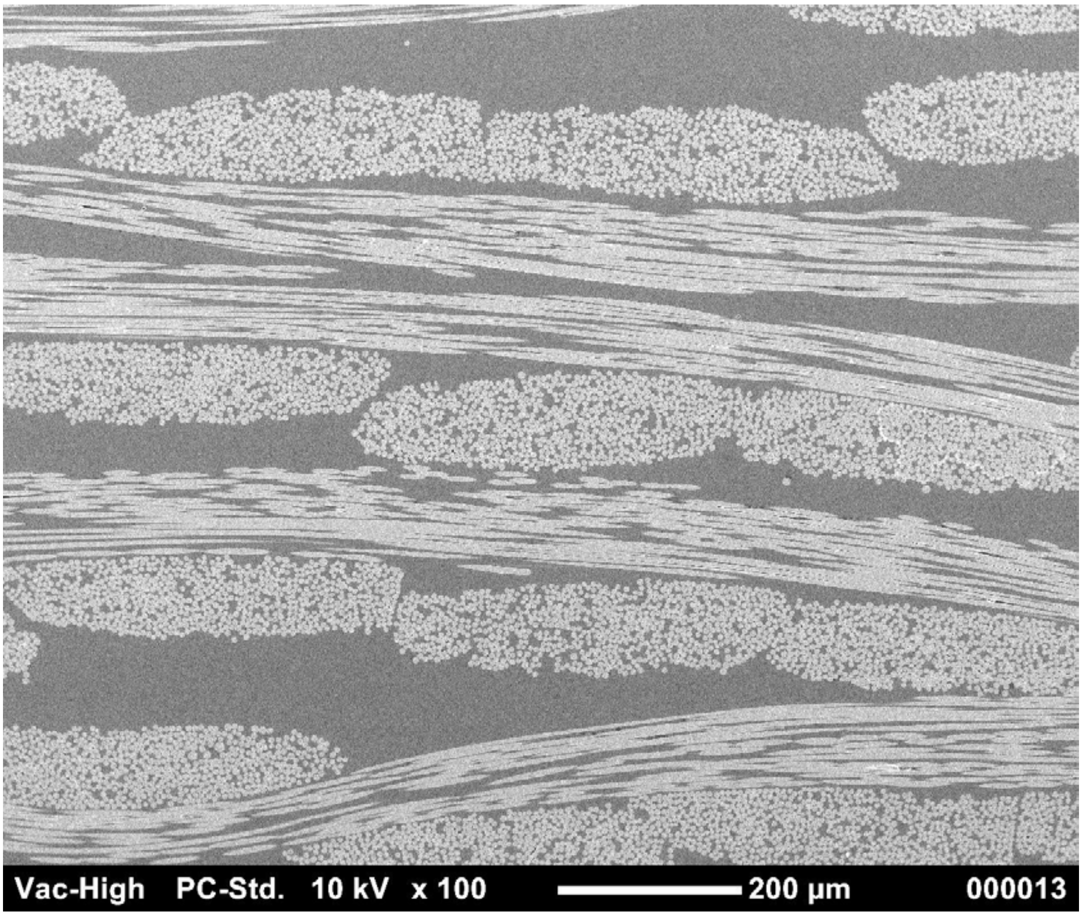
Platform Composites

Fiber Reinforced Polymers

First steps in the program definition



CONTENTS

1. INTRODUCTION

Unlike the situation in countries surrounding the Netherlands, the Dutch landscape of (continuous) fibre reinforced polymer based composites is characterised by many local initiatives supporting regional interests and local companies. Recently it has been realised that there is an urgent need for a concerted national effort joining the forces of all parties in the value chain. The combination of DPI and M2i with their industrial and academic partners offers an excellent starting point to organise this.

The industrial value chain varies per market segment, but in general it is characterised by a limited number of large material suppliers, a large number of small composite companies and again a limited number of large customers.

Dutch universities and research institutes have a good knowledge position and the industry basis is strong, especially in selected areas such as thermoplastic composites and the production of thermoset resins as well as Fibre Metal Laminates.

Supported by the Dutch ministry of economic affairs, the two Top sectoren HTSM and Chemie, DPI and M2i are setting up this new program with fundamental and applied research projects. The strategic objectives are to create an innovative network and to accelerate development and utilisation of composites in high tech applications and to make sure a strong and internationally recognised Dutch position in specific markets and expertise domains is created. The two topsectors HTSM and Chemie are prepared to contribute funds in excess of 2 MEuro for the start-up of such a research platform, provided it is well supported by the industry and contributes to the long term policies of both topsectors.

The current proposal focusses on the next 5 years but should be the stepping stone to a long term strategic Dutch program on Fibre Reinforced Composites.

1.1 Objectives

The strategic objectives of the program are to create an innovative network and to accelerate development and application of fiber reinforced polymers in selected market segments and to create specific fields of expertise in knowledge domains not yet widely covered in foreign programs.

1.2 Organisation

The programme will be carried out in a Public Private Partnership (PPP) of a type yet to be defined. DPI and M2i will take the lead in setting up this PPP. The three Dutch technical universities (UT, TUD and TU/e) will be responsible for carrying out the more fundamental research. Industry and research centres closer to the industrial practice such as NLR, WMC and TPRC will be responsible for the more applied research. Both M2i and DPI (including DPI-VC) will be responsible for the knowledge transfer to the industry at large.

The work will be divided in separate work packages.

1.3 Platform program definition process

The program definition process will take place along a two-step process:

1) Pre-definition of the program by a joint DPI-M2i committee of experts (the steering committee, see Appendix)
2) Program refinement on the basis of additional input from consulted industries and intended financial commitment of prospective partners

The actual program will be launched early 2015.

The current report contains the skeleton of the research program as defined by the steering committee.

1.4 Point of contact

The point of contact for correspondence will be Ronald Korstanje

Dutch Polymer Institute
The art of connecting

Ronald Korstanje, MSc.
Program Area Coordinator Performance Polymers

Visiting address: Kennispoort, 3rd floor, J.F. Kennedylaan 2, 5612 AB Eindhoven
Mail address: P.O. Box 902, 5600 AX Eindhoven (NL)
T +31 (0) 40 247 5119 M +31 (0) 6 47 48 55 69 F +31 (0) 40 247 24 62
E r.korstanje@polymers.nl W www.polymers.nl

2. APPLICATIONS OF COMPOSITES

2.1 Applications of composites

Utilisation of composites is characterised by a wide range of applications from sports, armour, electrical, biomedical and household applications to materials for the transport, construction and energy sector. The main applications can be classified as follows:

- **Automotive:**
  Large series, limited size, short processing time (carbon/bio, transition from thermosets to thermoplastics)
- **Aircraft:**
  Carbon, small series of integral constructions (thermosets), large series of building blocks / sub-constructions (thermoplastics)
- **Space:**
  Professional prototypes, assemblies, exotic materials, high secondary demands (thermal compatibility, damping, extreme circumstances, etc.)
- **Maritime:**
  Glass, thermosets, sandwich panels, from one offs (super yacht) to small series. From integral constructions and large series of standard parts and building blocks to modules for advanced and large series of standard parts and building blocks/ modules for advanced hulls and moving parts.
- **Infra/building and offshore:**
  Similar to maritime, for bridges, cranes and lock gates, (heavily-loaded), facades, smart anti-fouling coatings, cost driven, long life, maintenance free.
- **Energy production, transport en storage:**
  Blades for wind energy production. Oil and gas transport through endless pipes (on a reel or produced in-situ), cryogenic- and high pressure vessels, high performance fibers, low cost polymers, mass production, methods for life monitoring (remote, inspection), from safe-life to fail-safe (markers)

2.2. Ordering of composites/ the Dutch landscape

The most recent study of the VKCN (beginning of the 21st century) shows that the Dutch composites industry consist of ca. 350 companies [4]. This includes circa 40 material suppliers, 200 composite producing companies, 25 assemblers and 50 others (including research institutes and engineering firms). The investments required to start a composite company are low, this has led to many small composite producing companies Due to the global financial crisis several companies have recently been forced to stop their activities, the current figures may thus well be lower than as indicated in this study.

In 2000 14.000 tons of polyester resins were produced in the Netherlands, this is equivalent to a turnover of circa 210 M€.

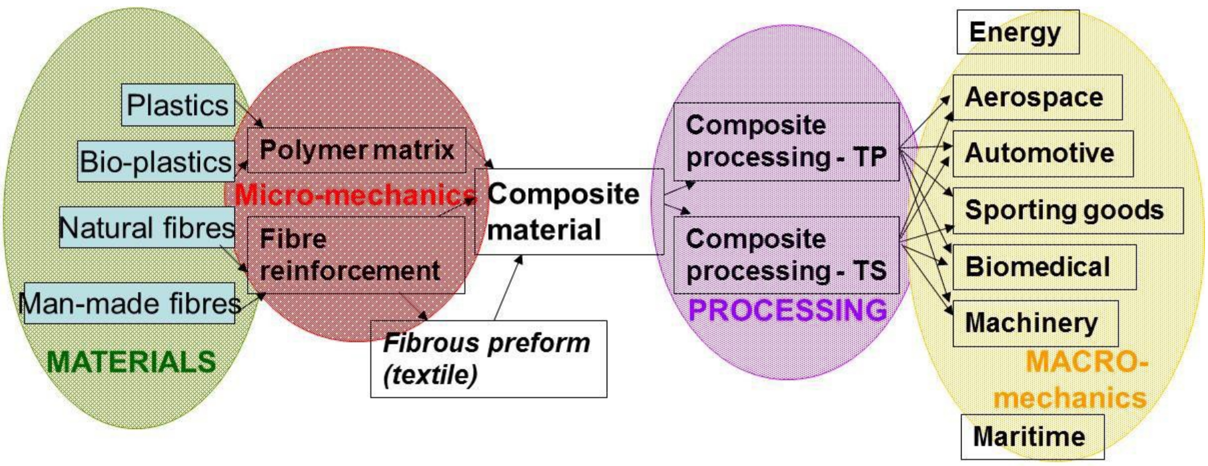
The study “Global Market Scenario” from the JEC composites predicts that the use of composites will grow with 6% per year in upcoming economies.

The Dutch landscape for composite applications is very diverse, and can be ordered according to the following variables/ parameters:

- *Constructive role*: primary or secondary constructions/ heavily of lightly loaded/ fail safe / damage tolerant/ safe life.
- *Product morphology* (open vs. closed sections, straight vs bent sections, thick vs thin walls).
- *Environment*: hot / cold = cryogenic-artic / aggressive.
- *Productsize* and *size of series*.
- *Fiber morphology*: whiskers / chopped fibers / long fibers / tapes / fabrics / woven structures / nitted structures /3D preforms /stitching / tufting etc.
- *Fiber materials*: glass, carbon, polymers, biofibers
- *Matrix materials*: thermoplastic, thermosetting, commodity / high end / specials (e.g. for very high temperatures)

2.3 Value chain for composites

The value chain for fiber reinforced composites involves material suppliers (fibers & matrix material), composite producers (combining the fibers and the matrix), processing (forming and joining), the OEMs (making the final constuction) and the end users (interested in non destructie damage monitoring and repair). Finally the recycling of composite materials will be a topic of serious interest. A complete program should preferably involve all aspects of the value chain in selected market segments. The focus in the program will be determined by the needs of the participating industries.



Fugure 1: Composites production and applications, from prof. Verpoest KUL

3. IDENTIFIED WORK PACKAGE THEMES

3.1 Optimisation and Control of the Composite Performance and Manufacturing Process

A major topic encountered in the production of all forms of composites irrespective of size and chemical composition is the issue of property and dimensional control. The composite manufacturing process requires many steps at different length scales, yet the final performance of the composite is that of the structure as a whole.

Aspects of fibre alignment, fibre wetting, matrix flow, consolidation of plies all play a role in achieving optimum properties. In contrast to sheet metal and bulk polymer production, the tools to tune and monitor the relevant processes are underdeveloped resulting in a higher uncertainty in product performance than in other materials classes. Furthermore, the micro and meso scale are relatively close to the macro scale, such that variability on these smaller scales often show up at the larger scale, inherently leading to a relatively large variability in properties and performance as measured in coupon tests.

Scaling up the composite market requires automated, fast and robust manufacturing processes with predictable and reproducible component properties and performance. It is needed to get away from manual lay-up and the related human induced variability, to reduce the amount of trial-and-error iterations in product development, and to eliminate the necessity of repeated time consuming non-destructive inspections with e.g. ultrasonic techniques.

Various steps are necessary to achieve these objectives:

- Development of virtual (CAE) multi-scale modelling tools that adequately capture the influence of processing conditions on the local and on the overall (product scale) mechanical behavior. This will allow for optimization of material, product and processing designs necessary to achieve the required application properties.
- Material property data for model validation needs to be determined Process monitoring techniques need to be identified and/or developed to acquire the relevant process information;
- Strategies need to be developed to employ these techniques in order to correlate the sensor output to the product quality with good accuracy and specificity;
- Possibly, to develop corrective strategies by adapting the process setting, based on both process analysis tools and the output of the process monitoring sensors, in order to reduce scrap rate and increase productivity.

The outcome of this programme should result in

- A shorter time to market due to a more efficient composite development process by using these process analysis tools;

- In-line process monitoring technology by which the need for non-destructive inspection is reduced significantly or eliminated in total;
- Smart processing methods which interactively respond to the process monitoring signals to prevent (or significantly reduce) scrap production.

3.2 failure and repair of composites

While composites generally have excellent (initial) properties, they can have small damages directly after production that are not easy to detect. They are sensitive to damage-evolution due to time dependent mechanical macroscopic and local loads, thermal loads, electrical loads and chemical loads. In particular the combination of chemical attack and mechanical load is insufficiently known.

Failure in composites is a poorly understood phenomenon, and expertise on remedies is limited. There are a multitude of failure mechanisms and modes, some of which have been researched extensively (e.g. fibre fracture, crack initiation and propagation, delamination), but most of which cannot yet be adequately modeled and predicted (fatigue). Environmental failure mechanisms, such as damage due to (combined) influences of humidity, UV-radiation, biofouling, and erosion, become more relevant for e.g. new developments in infrastructural, architectural, and civil applications. A great challenge is prediction of long-term performance, both mechanical and environmental. Understanding of interaction between failure mechanisms is lacking even more.

While damage is essentially unavoidable is important to realise that the repair strategies for composite structures are grossly underdeveloped. Non-destructive testing of structures is still insufficiently developed and the reparability of composites after damage sets new demands for composite architecture and chemistry.

Composite structures are integral structures, meaning that local failure can often not be mitigated by part exchange. Repairs in composite structures are closely related to connection methods, notably adhesive connections, long-term performance prediction of such connections is still a popular research topic. Nevertheless, repairs in composites become increasingly relevant, for various reasons. In some industries, damage tolerant design is becoming more prevalent, meaning that inspections and repair become an integral part of the product's life cycle. Furthermore, in various industries, the need for life extension programmes requires adequate condition monitoring and repair methods.

Failure and repair are adjacent to other fields in the sense that influences of manufacturing and repair preparation are quite significant, and that there is ample scope for interaction with structural health monitoring developments.

3.3 connecting composites to metal structures

Light weight structures with optimal durability, can be obtained by combining dissimilar materials such as composites and metals. The combination of such materials requires hybrid joints. The joints are form locked or adhesively bonded constructions based on composites in combination with steel, aluminum or titanium and can be applied in demanding applications such as in aerospace or in marine environments.

Adhesive bonding is the most promising technology for joining hybrid structures. Mechanical and chemical mismatch during the joining process form considerable hurdles. Stresses and (physical) aging during the lifetime of joined parts, as well as influences of the diverse environments these materials have to perform in, intrinsically lead to sites of likely early failure.

One of the main challenges is thus to achieve a durable adhesive bond, to guarantee the interface integrity composite/adhesive/metal and the adhesive material integrity during the service life of the structure. This knowledge is essential to make the correct choices in the initial design, such as surface pre-treatments, coatings, geometries, etc.

Adhesive bonding of composites with any surface is influenced by the surface free energy of the matrix materials used. Composites based upon thermosetting or thermoplastic polymeric matrices, and containing glass, aramid, or carbon fibres, are usually based upon epoxy or polyester resins and are, therefore, more polar than fluorocarbon polymers or polyolefins. This leads to epoxy and polyester matrices (and surfaces) possessing relatively high values of surface free energy and this makes them more receptive to adhesive bonding. This is because relatively high values of the surface free energy of the composite substrate will lead to good 'wetting' by the adhesive and to good intrinsic adhesion via the 'adsorption' mechanism of adhesion. The 'adsorption' mechanism of intrinsic adhesion simply proposes that, providing intimate molecular contact between the substrate and adhesive (i.e. good 'wetting') is established, then the intrinsic adhesion may arise from interatomic and intermolecular forces acting across the adhesive/composite interface. Examples of such forces are dispersion forces, dipole forces, hydrogen bond forces and primary chemical (e.g. covalent bonding) forces, which will lend itself very well to bonding of "like" materials, but describes the hurdle if bonding to other materials is required [1].

Composites based upon thermoplastic polymers typically employ matrices such as poly(ether-ether ketone), poly(ether sulphone), polypropylene, etc. These polymers tend to have low values of surface free energy and hence are difficult to bond using standard engineering adhesives. Hence, for these composites more complex surface pretreatments are often needed prior to bonding when using standard engineering adhesives [1].

Next to adhesive bonding other approaches like mechanical fastening, welding and solvent bonding are employed in industry, each coming with their own design aspects, performance and effectiveness in the final construction [2].

Mechanical fastening presumes the use of additional parts (fasteners) such are polymeric or metallic screws, bolts, washers, rivets or it relies on integrated design elements such as snap-fit or press-fit joints. This method lends itself well for joining of dissimilar materials (like polymeric with metallic surfaces).

Solvent bonding, the application of a solvent at the bond line to induce sufficient mobility of polymer chains to interdiffuse, mostly finds its place in the connection of composite materials to materials of comparable chemical character (solubility).

Welding, or fusion bonding, uses heat to melt the polymeric material at the contact surface enabling flow of material to form a bond by chain entanglement or with surface roughness with a substrate. Also this method is mainly used for connecting materials of similar physical properties.

All these methods only illustrate the range of possible approaches, which will also heavily depend on the design aspects and the expected performance and tolerated failure modes of the final structure. Next to the moment of connecting, and evaluating the strength of the formed connection, the lifetime and performance under extremes as well as different response of the materials that form the structure under environmental influences (like heat, humidity, chemical attack, stresses) will determine the real lifetime and durability of the formed connection.

In this part of the platform focus should be on:

- Provide a design guideline for hybrid structures, where the loads are transferred efficiently tor the composite part.

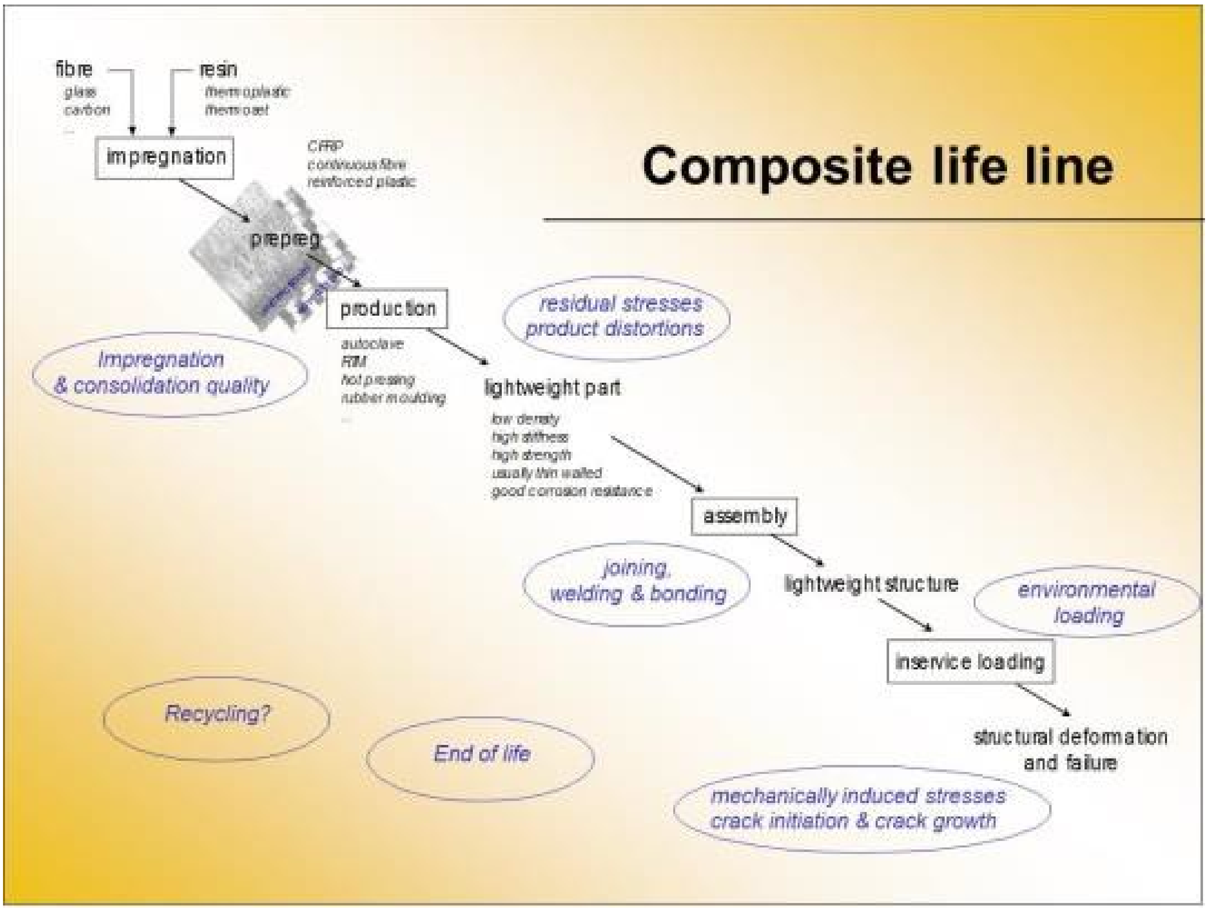
- Select environmentally friendly surface treatments for metals before adhesive bonding to the composite material.
- Determine the right adhesive bonding techniques and process to achieve successful connection between polymer composite and other parts of structures (like metals, plastics etc). Compare adhesive bonding to mechanical bonding and include combinations of chemical and mechanical bonding.
- Develop an understanding of mechanisms of combined chemical and physical degradation (= mechanical degradation) of interfaces in hybrid joints to increase the reliability and predictability of the lifetime of hybrid joints and bonded structures. The results will be validated by Aging studies under different environmental (e.g. moisture, temperature, UV-light) influences.
- Implementation in production lines, focusing on reduced cycle times, cost, etc.

The unusual expertise embedded in M2i and DPI on both metals and polymers will give us a very strong competitive advantage on the international market. It was the earlier expertise in metal-polymer bonding which formed the crucial technological basis under the development of the GLARE and other fibre metal laminates.

[1] A.J. Kinloch, B.R.K. Blackman and W.S. Teo, THE ADHESIVE BONDING OF POLYMERIC MATRIX COMPOSITES, ICCM17proceedings, 2009
[2] M. Sercer, P. Raos, Joining of plastics and composites, Journal of Plastics and Composites.
Related M2i projects:
[3] J.M.C. Mol & H.A. Terryn: Adhesion and delamination mechanisms of multilayer organic field joint coatings on carbon steel
[4] R. Akkerman: In and Out-of-plane strength of Hybrid Joints between Metals and Thermoplastic Composites/ Titaan in thermoplasten
[5] T.J. Dingemans: Hybrid Polymer Composites (HyPoComp)

3.4 recycling and end-of life treatment

Material re-use is the key facilitator in the 'circular' economy. While already very difficult for monolytic materials such as metals and polymers, the recycling and more general the end-of-life treatment of intrinsically heterogeneous composite materials is even more complex.



Composite structures often have a better Eco-footprint compared to their equivalent in traditional materials like steel or aluminium. Reduced emissions through lower weight in transportation, easier installation and reduced maintenance in construction, all contribute to this great performance. However, the increasing use of composite materials has also has raised environmental concern over the waste produced. This has lead in recent years to so-called Eco-composites based on biobased resources such as natural fibres that are biodegradable or can be incinerated, as well as fully recyclable single-polymer-composites such as polypropylene matrices reinforced with high strength polypropylene fibres. However, most of today's composites use either glass or carbon fibres together with thermosetting matrices, which are not easily recycled because they are cross-linked and cannot be remelted, in contrast to thermoplastics.

Glass fibre reinforced plastics (GRP) continues to account for by far the largest portion of the composites market, i.e. the reinforcing fibres in over 95% of all composites are glass fibres. The overall GRP market in Europe is an estimated 1 million tonnes, with the transport and construction sectors each consuming one third, while other large markets include electro &

electronics sector (E&E) and the sport & leisure segment. Growth rates for carbon fibre reinforced plastics (CFRP) are very high at around 15%, with currently approx. 70,000 tonnes of CFRP being produced for various market segments, the most important being rotor blades for wind turbines, closely followed by aerospace & defence and sport & leisure, while the automotive segment, is viewed as the sector with the largest growth potential.

Current and impending waste management legislation is putting pressure on all these industries to address the options available for dealing with composite waste. Such waste legislation focuses on dealing with waste through the waste hierarchy and is therefore favouring composite waste management strategies involving recycling and reuse. This not only affects end-of-life waste but also manufacturing waste. For example, Airbus has targeted for 95% of their CFRP manufacturing waste to go through a recycling process by 2025, with at least 5% of the waste to be recycled back into the aerospace sector.

At present most composite materials are destined for landfill at the end of their lifespan. As a result, globally, there could be over 1 million tonnes of glass fibre recycled out of end-of-life GRP and in 20 years time this is expected to grow to 6 million tonnes. To assist in the transition from disposal in landfill to recycling, the composites' industry needs to consider (re)designing materials and components for easier deconstruction, reuse and recycling at the end of the product life. EU directives such as End of Life Vehicles (ELV) and Waste Electrical and Electronic Equipment (WEEE) are already putting pressure on solving composite waste management through recycling and reuse. The ELV directive states that by 2015, 85% of ELVs will have to be reused or recycled (excl. energy recovery), with only 10% incinerated with energy recovery, and only 5% going to landfill. Whilst this new legislation does not yet impact on the construction industry, a proposed EU recommendation on Construction and Demolition Waste is currently under negotiations, which will have a significant effect as composite suppliers could lose their market share to metal and other industries if they cannot ensure that their components can be reused or recycled at the end of their life.

Thermoset plastics, unlike thermoplastics, have the reputation of not being recyclable. Thermoplastic composites can be recycled by remelting and remoulding. However, this is not the case for thermoset composites which dominate the large aerospace, wind turbine and construction markets. The recycling of such composites has been a growing area of research with mechanical, thermal and chemical methods of recycling all being considered, but few being applied successfully commercially. To add complexity, a recycling process for composites must be tolerant of contamination if post consumer scrap is to be recycled without costly cleansing operations. A process which requires a pure waste stream requires a high-value final product whose value is sufficient to off-set the initial cost of sorting and separation.

One option for thermoset composites is grinding, with the resultant recyclate being used as filler in new composite materials. However, mechanical breakdown and grinding often leads to low-value products. More advanced mechanical recycling has therefore focused on retaining fibrous regrind as reinforcing material to partly replace virgin reinforcing glass fibres in new moulding compounds.

Thermal recycling is currently the most technologically advanced process for recycling of composites, while chemical degradation is also considered to reclaim fibres. Thermal processes that have been applied to plastic scrap include: incineration, pyrolysis, gasification, hydrogenation and combustion. Combustion processes are generally optimised to work at high temperatures for high thermal efficiency, which leaves a low value incombustible residue as a hard glassy slag. Thermal decomposition of composites can however also be used to recover the

more valuable fibres. Whilst the calorific energy value of polymers is only around £30/tonnes, reclaimed glass and carbon fibres posses a much greater economic value at £1,000/tonnes and £10,000/tonnes, respectively. Processes to recover fibres typically need lower temperatures to preserve the strength of the reclaimed fibres and can be economically viable, providing that the thermal efficiency is not too much compromised. The most notable thermal recycling process with fibre recovery developed so far is using a Fluidized Bed process. Fibre reclamation processes seem particularly suitable to carbon fibres as they have not only greater intrinsic economical value, but also a high thermal and chemical stability, which implies that their excellent mechanical properties are not easily degraded. However, so far most of these reclamation processes produce short carbon fibres recyclate for non-critical structural markets, whilst only few closed-loop recycling technologies exist that are able to recover structural fabrics for advanced composite applications.
In summary, current and future legislation has driven industry to develop end-of-life solutions for composite products and currently a number of technologies are under development focusing on either material recycling or thermal processing. Key barriers to commercial success so far have proven to be the identification of suitable markets for composite recyclate at the right cost. Current recyclate is often too expensive to compete in available markets and because of this there is a need to develop higher grade recyclates for more valuable markets.

Ultimately LCA tools will also have to provide answers regarding the real environmental impact of newly developed recycling processes. Moreover such tools can highlight areas for optimisation to achieve economically feasible industrial processes. For example, established waste management hierarchies and EU directives favour mechanical recycling over thermal or chemical recycling, but it remains to be seen if these are the best from an environmental point of view. For this, detailed LCA is needed to identify the environmental impact of alternative processes.

3.5 educational programs

While composites have been around for some time, the educational programs on composites at all industrial levels of expertise are underdeveloped.

While composites have been around for some time, the educational programs on composites at all industrial levels of expertise are limited to specific applications, hampering cross-overs between industries as well as expertise levels. For example, composite applications for the aerospace industry are based on different implementations of similar materials than for instance in the wind industry, while eventually cost and high reliability are important design drivers in both industries. Often, educational programmes focus on distributing the knowledge required for these specific applications or competence, ignoring similar approaches in other fields. This leads sometimes to the need for 'reinventing the wheel', e.g. experience in composite repair has developed in parallel in naval, aerospace, and energy applications.

There are already initiatives, e.g. in the industry associations, to provide basic knowledge on composites and to coordinate different levels of expertise, so that eventually VO-MBO-HBO-WO courses are interlinked and students can follow a continuous, coordinated, programme of composite education. Next to this coordination, technical content that reflects the state of the art in composite research must be available to continuously optimize this education.

This Platform will contribute to the development of connected sets of courses and other educational programs at MBO, HBO and academic level (both at designer and material processing level) to make sure that project results (other than the IP marked for limited distribution) become available to society and the end-users, as well as to warrant a sufficient influx of well-educated young professionals to improve the competition potential of the composite and composite using industries.

There are options for European cooperation with clusters like: EMC2, Bayern Innovativ, M.A.I Carbon and The Knowledge Transfer network.

4. Concluding remarks

4.1 Preliminary program

A group of experts from industry and academia have set up a preliminary program on the technological and scientific aspects of composites as well as on the application of these materials in practice. The program is aiming at selected topics enhancing the fundamental and applied knowledge for the application of composites. The main program themes we would like to focus at are:

1. process monitoring during composites production;
2. failure and repair of composites;
3. hybrid composites;
4. composite recycling and end-of-life treatment.

The aim is to set up a national research program of more than 4 MEuro involving both large companies and SME's. To organise this a scheme will be developed allowing the industry to participate on different levels. Apart from the financial support from the industry and the ministry the intention is to organise additional funding via NWO and participating research institutes. .

4.2 Input meetings

Composites are used in a variety of fields. In order to obtain a better insight in the needs of the industry and to gain principle commitment from industrial partners, input meetings have been planned on January 26th and 27th 2015 at SKF in Nieuwegein.

APPENDIX : COMPOSITION OF THE PLATFORM PREPARATION COMMITTTEE

- Remko Akkerman, UT, professor Composite Technology and scientific director of TPRC
- Martin van Dord, DPI-VC, NRK, VKCN, and link to SME's working in the field of composites
- Bert van Haastrecht, M2i, platform program coordinator Composite Materials at M2i (vice chairman)
- Jan Henk Kamps, Sabic, research chemist for new composite matrix polymers.
- Rogier Nijssen, WMC, program manager large scale composites and contact person for the Colleges of Higher Education
- Ton Peijs, TU/e and Queen Mary University (UK), professor and cluster leader M2i for the domain composites
- Bert Thuis, NLR, leader program additive manufacturing and chairman of Compuworld
- Sybrand vd Zwaag, scientific chairman DPI and professor Materials Science at the faculty Aerospace Engineering TU Delft (chairman)

REFERENCES:

1. Materials (Metals) for Additive Manufacturing, NLR-TNO Proposal , June 2014
2. M2i workshop Composietmaterialen voor structurele toepassingen, 3 december 2010
3. Workshop M2i Composieten, 19 mei 2011
4. Biocomposieten 2012, Marien van den Oever en Karin Molenveld, DPI Value Centre
5. Wikipedia