

Plasticized cellulosic composites for packaging materials (COMPAC)

FINAL REPORT				
Title of the research project	Plasticized cellulosic composites for packaging materials			
Coordinator of the project	Prof. Jukka-Pekka Valkama			
BASIC PROJECT DATA				
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Contact information of the coordinator (institute/unit, address, telephone, fax, e-mail)	Baden-Wuerttemberg Cooperative State University, Karlsruhe Erzbergerstrasse 121 76133 Karlsruhe Tel. +49 177 3494478 E-mail valkama@dhbw-karlsruhe.de			
URL of the project	_			
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<u>Norway</u> The Research Council of Norway (RCN)	[amount in EUR]	
<u>Slovenia</u> Ministry of Education, Science and Sport (MIZS)	[amount in EUR]	
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PROJECT TEAM (main participants)		
Jukka-Pekka Valkama, M Prof. DrIng., Professor Paper technology	Cooperative State University Karlsruhe	Germany
Tero Tervahartiala, M M.Sc.(Tech.), Academic staff member	Cooperative State University Karlsruhe	Germany

Mirja Illikainen, Prof. Dr.(Tech.) Professor Fibre and Particle En	F gineering	University of Oulu	Finland



Petteri Piltonen, Dr.(Tech.) Researcher	М	University of Oulu	Finland
Bo Westerlind, Dr.(Tech)	М	Mid Sweden University	Sweden
Jukka Silvennoinen M.Sc.(Tech.) & MBA Head of R&D	М	Powerflute OY	Finland
Sami Anttilainen, M.Sc.(Tech.) & eMBA, Vice President, Technology	М	VALMET Oy	Finland
Lauri Verkasalo, M.Sc.(Tech.), Director, Strategic Research	М	Metsä Board	Finland
Tord Gustafsson Dr.(Tech) R&D Manager	М	Blatraden AB	Sweden
Hans Grundberg, Dr.(Tech.), Dr.(Tech.) Development engineer	М	Aditya Birla Domsjö Fabriker AB	Sweden
Heikki Sojakka, R&D Manager	М	Fiber-X AB	Sweden
Christian Hössle Dr.(Tech.) Product development manager	М	Klingele Papierwerke GmbH & Co.KG	Germany
Berthold Aumüller, Dr.(Tech.), R&D Manager	М	BHS Corrugated Maschinen- und Anlagenbau GmbH	Germany
Maximillian Rauscher, B.Eng. Development Engineer	М	BHS Corrugated Maschinen- und Anlagenbau GmbH	Germany



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	S (if relevant) earned or to b		d within this project.		
2017(Exp)) Dr.Ing.	Μ	Tero Tervahartiala, 1987,	TU Darmstadt	Prof. Samuel Schabel
2018(Exp) Dr.(Tech.)	М	Nils C. Hildebrandt, 1988	University of Oulu	University of Oulu
2016	B.Eng.	Μ	Maximilian Rauscher, 1993	DHBW Karlsruhe	Dr. Berthold Aumüller & Prof. Jukka Valkama



PROJECT SUMMARY REPORT

The COMPAC project was a collaboration project between three universities and nine industrial partners of which three were SME's. Participants of the consortium are all connected to the paper and packaging sector. Geographically the consortium is divided evenly to Germany, Finland and Sweden. The COMPAC consortium was set up with the goal to research and develop a cost-effective solution for packaging industry to decrease weight of paper-based packaging products, especially corrugated board. Planned path to reach the goal was to adapt a known vulcanised fibre production process into a paper making process.

Vulcanised fibre is a composite material made from 100% cellulosic material, such as cotton linters, by a zinc chloride or sulphuric acid dissolution and regeneration process. Traditional process is three-fold: 1) Submerging of a paper web into a bath of aqueous solvent where dissolution of cellulose is initiated. 2) Removing of the solvent by subsequent water baths where re-crystallisation of the cellulose fibres takes place. 3) Drying of the fibre web. The disadvantages of the traditional process are the toxicity of the chemicals, low production speed, long lasting washing stage and high costs. With costs starting from about 4.000 EUR/ton are the application fields for vulcanized fiber products limited. Vulcanised fiber as material is all cellulosic material and shows very high strength and electrical insulation properties. The project was divided in three steps: 1) researching for bio based fibre source alternatives from paper pulps and investigated different chemical solvent systems (DES, Ionic liquids, etc.) as an alternative to traditional method. Using the gained understanding about the formation of cellulosic composites from different combinations in second working packaging the processing ability of the material (green vulcanized fibre) and the converting ability (corrugation, 3D forming) was investigated. Finally the new market applications were analysed and the recyclability and biodegradability of the material was researched. To highlight the difference between conventional vulcanisation process and the process developed in this COMPAC project, this report will refer to the developed process as Plafco-process and to the material as Plafco-material (Plasticised fibre composite). PLAFCO was registered as a trademark during this project and a patent application was left for the process.

COMPAC's goal was not only to integrate the production process into a paper making process, but to find more sustainable alternatives for dissolving of cellulose. Initially the plan of the project was to develop a process to pre-modify the fibre material (pulp) before making the paper itself. Based on laboratory tests with various pulp types and cellulose solvents it was identified that a variety of conventional paper pulps are applicable for making Plafco. However, to treat "single" fibres in the pulp preparation process effectively, without formation of fibre lumps, the pulp suspension would have had to been lowered below 1% and below resulting high chemical-and washing water consumption, not to forget energy to hold the right operating temperatures. Several solvents systems were tested and weighed for their effectiveness, price and hazardousness for the environment.

Integration of a solvent bath to a paper machine would require significant structural modifications of the machine and require excess energy for temperature controlling. A plan was devised to use a surface-sizing unit located in the drying section of a paper machine to apply the cellulose solvent. Based on our plan the required washing section can be built in the machine pit of a paper machine and for drying the existing drying cylinders could be used. Benefits of this approach is



very low chemical consumption and by insulation the energy losses in cooling can be minimized. The plan was successfully implemented and tested on a pilot scale (500 mm wire width) paper machine in Sweden. Corrugation tests on Plafco material was successfully conducted in Germany on a pilot corrugator (1000 mm corrugated roll width). Tests on Plafco-material show that there is potential of increasing the mechanical properties of corrugated board by up to one third (short-span crush test and calculated box crush test), resulting in a significantly lighter packaging products.

1.1 Introduction

1.1.1 Background

Future needs for the packaging paper industry are to develop more stable and functional packaging products from renewable resources with lower carbon footprint and lower costs. Through development of stronger structures, remarkable environmental and economical savings can be achieved. Stronger fibre matrix will allow using less material for the packaging products and saves enormous amounts of raw material but also improve the range of use by offering wider range of moist conditions where wood based packaging material can be used.

Wood containing composites have been successfully introduced to large variety of application like WPC (Wood Plastic Composites) panels. However, these materials still contain high amount of non-renewable raw materials despite the plastic substitution with e.g. PLA has shown good results. Furthermore WPC material production is made with plastic production equipment e.g. pelletizing and extrusion technologies.

1.1.2 Objectives

The objective of project COMPAC was to integrate the production of an entirely lignocellulosic composite into a paper making process. Furthermore the composite produced should be directly applicable to the industrial packaging product converting processes and other applications like press moulding applications.

Regarding material properties themselves the objective was to increase barrier properties for example against mineral oil migration, especially important for the food stuff industry. In addition, packaging products made from the composite material should bring more stability, strength and barrier properties without high investments in process equipment.

1.2 Results and discussion

Production process perspective:

Ultimate highlight of the project COMPAC was the pilot tests done at Fiber-X, Markaryd, Sweden. At Fiber-X the concept of producing Plafco material in-situ on a paper machine using both zinc chloride and urea-based systems. Both while producing paper as well as as an off-line concept. Tensile properties of paper produced increased up to 5 fold. Paper used was dissolved pulp provided by consortium member Domsjö Fabriker AB / Aditya Birla Group. Dissolution could be



decreased to under a second by using surface sizing press enabling quick treatment times and lays foundations to economical production. Figure below shows the process steps, the pilot machine used and the picture in insert the surface sizing equipment applied. As highlighted in the process steps plasticisation process can be also implemented as an off-line concept which has benefits in lowering the investment costs when rolling the concept out to the industry.



Figure 1: Pilot machine, pond size press used and process overview

The innovation was enabled by our invention to apply the surface sizing unit as the chemical application way. Ability to implement the Plafco-concept in existing paper making and corrugating processes lowers the barriers for companies to adopt the technology. Research done on pulp types and process parameters have given us understanding on how to optimally use conventional paper pulps - there is no need for high-value speciality pulps, keeping the production costs on a moderate level, increasing overall competitiveness. It is estimated that an industry relevant Offline Plafco machine, with wire width of 1500-2500 mm would cost around 1,5 to 3 MEUR when purchasing new machinery. The costs will be significantly lower, when retrofitting a paper machine or using a used surface sizing unit and a drying section. Solutions for washing section and cooling systems exist on the market. Production cost of Plafco-material is estimated to lay between 200 - 500 €/MT, not including the costs of the raw paper which normally lays between 500 - 1000 €/MT. Thus the pricing is competitive with conventional plastic materials. Low barrier for current paper producers to adapt the production process combined with competitive manufacturing costs give companies working in (or wanting to make a transition to) packaging paper products a possibility to create a competitive advantage by widening their product portfolio to more high value products and to enter new markets. Especially possibility to enter new markets such as food packaging has significant potential in increasing sustainability. Products, such as take away cups or plates are made from cardboard, but covered with plastics. Plafco-material has large potential



in increasing sustainability simultaneously raising awareness of the fact that in many products that are seemingly sustainable, might include "hidden" plastics.

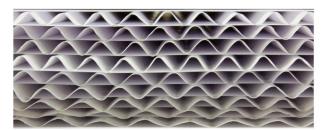
Project COMPAC was also successful in proving that Plafco-material is corrugatable using existing process equipment. This work was done in collaboration with BHS Corrugated Maschinen- und Anlagenbau GmbH and Klingele Papierwerke GmbH & Co.KG. Conversion of corrugated board further was studied by Andreas Hering Papertec Greiz and our assumptions were confirmed, that Plafco-material is also further convertable in cutting and die cutting processes. Figure below shows the setup used to test various Plafco-sheets and their corrugation properties.



Figure 2: Corrugation of Plafco-material

Several other converting techniques were tested within the project to evaluate the potential of Plafco-material to be used in packaging products with more complex geometries. Techniques used with fibre reinforced composites, such as wet layering of laminate structures and resin infusion process were successfully tested. In addition form pressing proved out to be a very viable method to form Plafco, as it is significantly more elastic than paper. Success in application of above mentioned techniques give also possibilities to replace plastic products within consumer product sector, such as housing for electrical equipment (Plafco-material is an electrical insulator).





Example 1. Corrugating





Example 2. Laminating to form

Example 3. Deep drawing

Figure 3. Examples of forming methods tested.

Material perspective:

Significant amount of effort has been to understand the plasticisation process with various solvents, most importantly with zinc chloride and urea-based systems. Most important result is that conventional softwood and hardwood pulps, both bleached and unbleached are applicable for making Plafco-material.

Following points will give an overview on the material properties: Tensile properties (softwood & hardwood pulps)*

Tensile strength
Tensile index
Elastic modulus
Elongation at break
st (softwood & hardwood)*
SCT
SCT Index
st (softwood & hardwood)*
CMT(0)
CMT(0) Index
(High dissolving & Eucalyptus)*



64 - 181 g/m² (water) 7 - 53 g/m² (oil) Airtight & Compostable *80 g/m² isotropic sheets

As can be seen the mechanical properties are on-par with many plastics commonly used in various packaging products and for the most part surpass the paper used for corrugated board products. Mechanical properties, airtightness and moisture resistance and moisture regulative properties show potential for Plafco-material to be used also in building industry.

1.3 Conclusions

Most important contribution to the state of the art is the innovative Plafco-process, which enables the industry scale use of a sustainable cellulose solvent. Result of this project has proven, that a paper machine, with minor adjustments, can be used to produce a fully cellulosic composite. As raw material normal hardwood and softwood pulps can be used. In addition the convertability to packaging end-products has been proven, and the preliminary testing shows that significant advantages can be gained by applying Plafco-material compared to conventional corrugated board material.

1.4a Capabilities generated by the project

Thesises:

B.Eng. Thesis (DHBW / BHS Corrugated), Maximillian Rauscher (2016): Corrugation properties of plasticized fibre material.

Ph.D. Thesis (DHBW / TU Darmstadt), Tero Tervahartiala (expected 2017) Ph.D. Thesis (University of Oulu), Nils C. Hildebradt (expected 2018)

European Patent: 17168108.3 (Verfahren und Anlage zum Herstellen eines flächigen imprägnierten Faserstoffproduktes) extended on 26.04.2017. Patent pending. *German Patent:* 102016205954.8 (Verfahren und Anlage zum Herstellen eines flächigen imprägnierten Faserstoffproduktes). Applied 8.4.2016, corrected and extended on 26.04.2017.

New registered trademark: PLAFCO, Nr. 30 2017 201 265

New company: PLAFCO FIBERTECH OY VAT: FI28383218



1.4b Utilisation of results

The results of project COMPAC have been used for four H2020 project applications:

COCO-FUNPAC	BBI-VC2-2015	25.08.2015
PLAFCO-MAP	BBI-JTI-2016	08.09.2016
SAMA-PACK	SFS-2017-1	14.02.2017
SUB-ZERO	BBI-D5-2017	07.09.2017

Next H2020 application will be BBI.2017-D5. H2020 applications focus on building a pilot Plafcomachine and to make demonstration products for food packaging and corrugated board packaging. In addition a German AiF application process has been initiated with the focus on using Plafco-material in building industry. It is also planned to apply for the upcoming WoodWisdom program. In each application the perquisite has been to take as many of the COMPAC consortium members as possible to the applications - when fitting to the call.

There is a urgent need for resources to build a pilot Plafco-machine as the resources within COMPAC were not sufficient to produce Plafco-rolls for production of corrugated board on fullscale corrugating machines. Also for press moulding lines, rolls with even quality are required. A company, PLAFCO FIBERTECH OY, has been established to serve as a way to open ways to attract private financing for realising the Plafco-concept. Members of COMPAC consortium are very interested in supporting the realisation of the concept as it supports their business. Companies in consortium serve as pulp suppliers, raw paper producers and converters. In a way PLAFCO FIBERTECH OY is a result of filling in the missing block within our consortium.

Once the resources to build a Plafco-machine is gathered, the goal is to work in tight cooperation with converters and material suppliers to get the first Plafco-products to the market. Currently one sourcing contract is under formulation, where a company is agreeing to buy Plafco-material if the price is right.

1.5 Publications and communication

1. Articles in international scientific journals with peer review

Publication: Piltonen P, Hildebrandt NC, Westerlind B, Valkama JP, Tervahartiala T, Illikainen M (2016). *Green and efficient method for preparing all-cellulose composites with NaOH/urea solvent.* Composites Science and Technology, 135, 153-158.

Publication: Hildebrandt NC, Piltonen P, Valkama JP, Illikainen M: Self-reinforcing composites from commercial chemical pulps via partial dissolution with NaOH/urea, Industrial Crops & Products 109C (2017) pp. 79-84.



Publication: Yapar, Ö.; Piltonen, P.; Visanko, M; Farooq, M; Valkama, J-P.; Illikainen, M.: Allcellulose compositesprepared with aqueous NaOH/urea solvent system based on a new impregnation method. (Submitted to Composite Science and Technology.)

Publication: Tervahartiala T, Hildebrandt NC, Piltonen P, Schabel S, Valkama JP. Corrugated board from all-cellulose composites: Comparison of chemical pulp raw materials. (Submitted to Packaging Technology and Science)

Publication: Westerlind, B., et.al., Through-the-thickness shear, fibre and tensile properties of plasticised paperboard.(not submitted yet.)

2. Articles in international scientific compilation works and international scientific conference proceedings with peer review

Presentation: Hildebrandt NC, Valkama JP (2016). Production of plasticised fibre composites (PLAFCO) with a paper machine via partial dissolution of paper. Cellulose Workshop, Örnsköldsvik, Sweden.

Poster: Continuous production of plasticised cellulosic composites with a paper machine, T. Tervahartiala & J., Valkama, Progress in Paper Physics Seminar August 22-26, 2016, Darmstadt, Germany.

Poster: Westerlind B (2016). Plasticised cellulosic composites for packaging applications (COMPAC). Science Innovation day. MiUn, Sundsvall, Sweden.

3. Articles in national scientific journals with peer review

4. Articles in national scientific compilation works and national scientific conference proceedings with peer review

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5. Scientific monographs

6. Other scientific publications, such as articles in scientific non-refereed journals and publications in university and institute series



a) Other dissemination

Making of H2020 applications that base on the Plafco-concept has been a very effective way to attract attention to the COMPAC project. In addition, attending various trade fairs (Interpack, Düsseldorf 2017; FachPack, Nürnberg 2016; PTS Seminar, München 2016; and alike) have been good ways to spread knowledge of the concept further and to gather partners for the next stages.

1.6 National and international cooperation

All together the consortium has officially met during 6 meetings to exchange information and ideas on project goals and what extra resources each partner had access to progress further. As the project progressed, some participants initiated "side projects" around the project theme that were relevant for their business. Example of this is the development of a measurement system to quantify packaging performance or finding new possible customers outside the normal value chain. As many partners belong to larger company groups/concerns, discussions went up to the central mother company, and information was then relayed further to other companies in the group.

The fact that the discussions got so far within the companies` internal structures was due to us being capable of forming a real business case out of our project result very early in the project. In a way this created a small snow-ball effect as we got more parties interested within partners` organisations and thus also better input in form of cost calculations and evaluation of synergy possibilities between stakeholders. All of this came in hand when formulating of our concept.

Collaboration between DHBW, University of Oulu and MiUn has strengthened significantly. In addition, new research collaboration has been initiated between DHBW and TU Darmstadt and TU Dortmund. Within new project applications around COMPAC collaboration has also been made with institutions in the Netherlands. Collaborations add value in making the overall concept of cellulosic composites closer to a wider market entrace as the theme is worked upon from many aspects.