Piezoresistive thermoplastic film for composites embedded health-monitoring system

Authors: R. Volponi[a], P. Spena[a], F. De Nicola[a], P. Russo[b]

a *CIRA Italian Aerospace Research Centre- Composites Prototyping Lab.*
b National Council of Research -Institute for Polymers, Composites and Biomaterials.
Corresponding author: r.volponi@cira.it

Abstract

Adding nanotubes in a polymer is possible to change it in an electrical conductive materials that can show also a piezoresitive behavior.
The present work show a way to use the piezoresitivity property of a nanocharged polymer into a fiber reinforced composite to improve an integrated health-monitoring system.

1 Introduction

The use of fiber reinforced resin composites has continuously expanded expecially in aerospace. Inspection and maintenance are important aspects when considering the availability of aircraft for revenue flights.
An exciting developments in materials and structures to construct ever more efficient air vehicle able to enable ‘smart’ maintenance could be possible through materials with embedded systems able to monitoring their status.

Using a nanocharged resins is possible to increase the electrical conductivity of composites by means nanotubes, and it’s possible to use their tendency to form electrically conductive networks when embedded, even at low concentrations, in traditional insulating polymers thus giving rise to a new class of conductive composites and multi-functional smart materials tanks to the piezoresistive capability of these CNT-composite materials.[1],[2],[3],[4].

Unfortunately usual manufacturing process for composites becomes not ever practicable as the viscosity of a resin charged with nanotubes increase dramatically[5],[6],[7]

An alternative solution to chain the the piezosestivity properties of a nanocharged polymer into a fiber reinforced composite has been proposed to get an integrated health-monitoring system.

The system proposed is based on a nanocharged thermoplastic polymer sheet self-standing that can be easily integrated during the laminating phase in a manufacturing fibers composite process.

2 Materials

In order to find a thermoplastic polymer to manufacture a nanocharged thin film, the selection of the material has been done considering an useful method to charge a thermoplastic polymer with nanotubes and the compatibility of that material with the polymer used as matrix in the final composite.

The matrix in a composite is usually an epoxy resin, and is not even simple to find a thermoplastic one that bonds well with thermoset matrix.

In a first approach several experiments has been done with polymer as Polyvinylacetat, Polystyrene.
That kind of polymers are soluble in a solvent as acetone or tetrahydrofuran, and are chargeable without difficulty dispersing nanutubes into a solution of polymer and solvent by sonication.

However that polymers doesn't bond well with epoxy and the insertion of a thin sheet of that polymers into a fiber reinforced laminate are source for delaminations.

Excerpt from ISBN 978-3-00-053387-7

The selection for a suitable material as been done over a a class of thermoplastic polymers usually used in manufacturing of fiber reinforced composites.

Frequently thermoplastic polymers are included in the epoxy resin formulations to improve the toughness of the composites.

The selected one has been a polymer yarn used to weave unidirectional carbon fabric produced by EMS-Griltech and named Grilon MS®

Fig.1 Grilon MS® Yarn

That material is declared as based on a mixture of phenoxy resins and to be meltable with epoxy resins [8], [9], [10].

2.1 Fabrication of nanocharged thin sheet

The pristine material is a yarn that isn't soluble in any common solvent, so to add nanotubes to it it's necessary to use an extrusion process.

The process of extrusion is done by means a machine called extruder, that heat the polymer over his melting temperature and mix it through two screws that rotates in opposite direction.

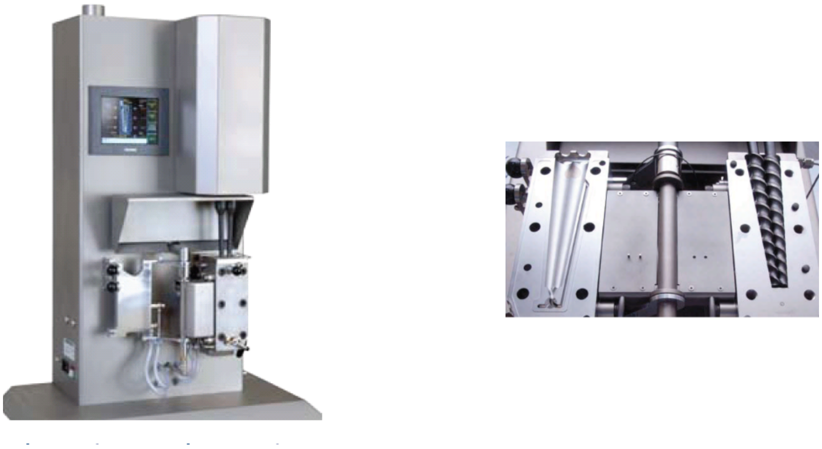


Fig.2 Extruder

Excerpt from ISBN 978-3-00-053387-7

The temperature at which the polymer melts has been determinate by an analysis done with a Differential Scannig Calorimeter (DSC).

A first run is done heating the polymer at a rate of 10°C/min till a temperature of 190°C

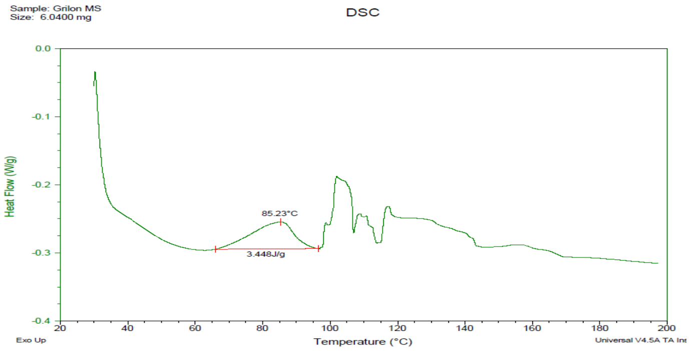


Fig.3 DSC First heating

It's possible to see that around 65°C the polymer starts to reacts and over 140°C became stable.

In a second run the reactivity there isn't but is possible to clear the melting temperature at 92°C and a clear thermoplastic behavior.

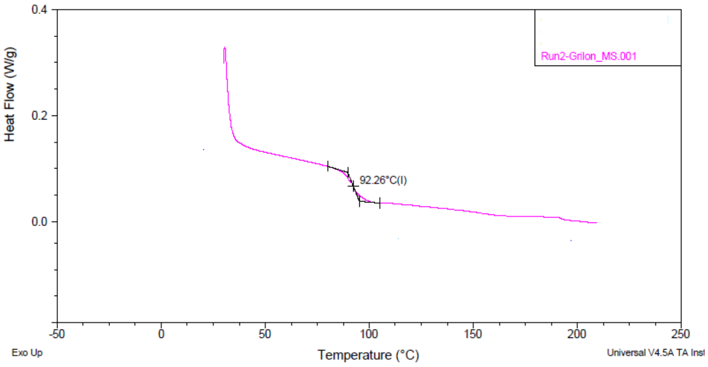


Fig. 4 DSC Second heating

So the extrusion process have to be done at an higher temperature than the that of melting.

The temperature used to mix and to extrude polymer with nanotubes has been settled at 160°C, where the viscosity of the nanocharghed polymer is appropriate to be extruded. Finally has been obtained pellets of polymer charged with 5% and 10% whit nanotubes.

Excerpt from ISBN 978-3-00-053387-7

Fig. 5 Nanocharged Pellets

Fig. 6 Hot Press

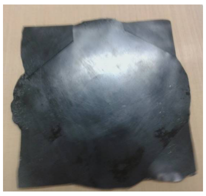


Fig. 7 Nanocharged thin layer

From each kind of pellets has been manufactured several thin sheets by means of an hot press The thickness of the films are about 150µm for that charged with 5% and about 300µm for films obtained from pellets charged with10% of CNT.

2.2 *Characterization of thin films*

The films have been evaluated over different aspects: their electrical properties and their compatibility with epoxy resin.

2.2.1 *Piezoresistivity*

From different sheet several rectangular samples have been cut and electrical contacts have been done at the edges of each sample:

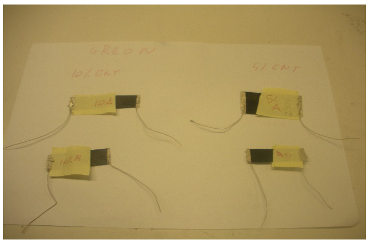


Fig.8 Nanocharged Samples with electrical contacts

In that manner the electrical conductivity has been evaluated:

Excerpt from ISBN 978-3-00-053387-7

Sample	Width	Lenght	Thickness	Resistance	Conductivity
	mm	mm	mm	Ω	S/m
Grilon +10%CNT A	57	14	0,302	667,5	20,19
Grilon +10%CNT B	47	16	0,288	412,2	24,74
Grilon +5 %CNT A	50	26	0,188	1443	7,08
Grilon +5 %CNT B	43	15	0,169	1752,2	9,68

The characterization of the electrical proprieties of the sheets has gone on to evaluate the piezoresistivity of the sheets.

The samples has been glued with an structural adhesive over an aluminum plate, in that manner the structural adhesive bonds the sample over the support and isolate it.

Fig. 9 Sample bonded on a rigid support

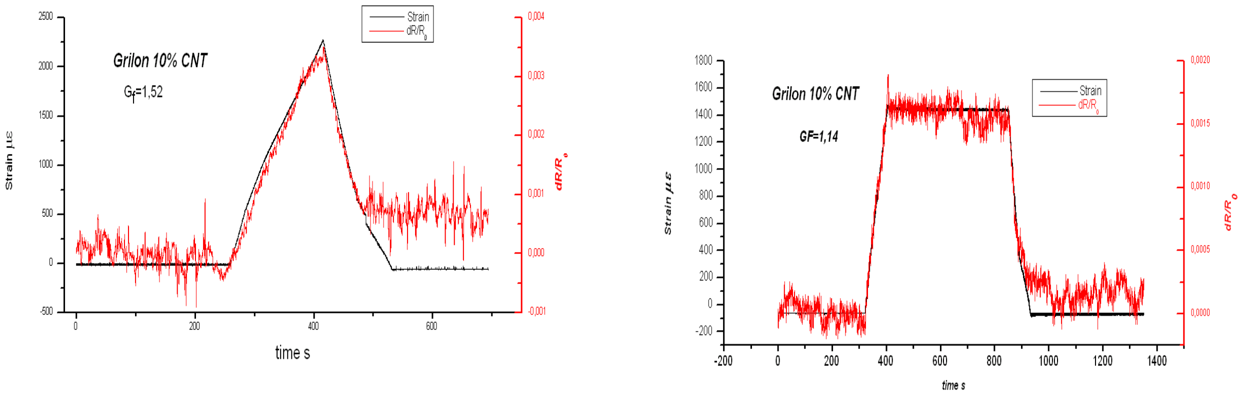
Strains have been applied to the sample by a mechanical testing machine INSTRON 4505 and in the same time a constant current flows across the sample .

Fig. 10 Electro-mechanical test

Excerpt from ISBN 978-3-00-053387-7

Simultaneously strains and tension have been recorded in the time:

For the sample charged with 10% of nanotubes has been found:



a

b

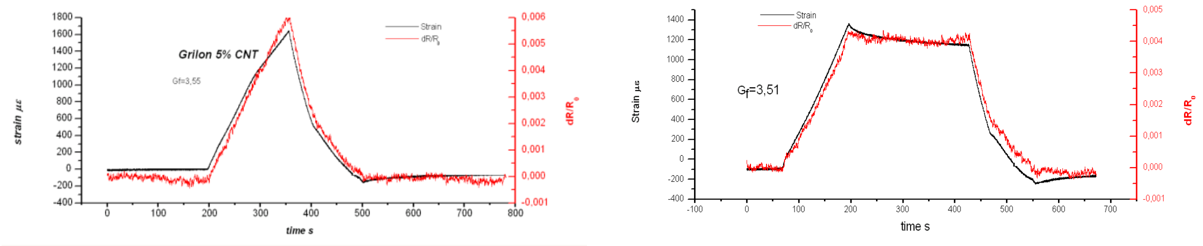
Fig. 11 ***a -b*** *Tests for 10% CNT charged layer*

It's possible to see how the variation of resistance of the sample follows the strain applied but Gauge factor in a bit more than 1.

The Gauge Factor is defined as the ratio between the strain and the variation in percentage of the electrical resistance: $G_f = \dfrac{\Delta R/R_0}{\varepsilon}$

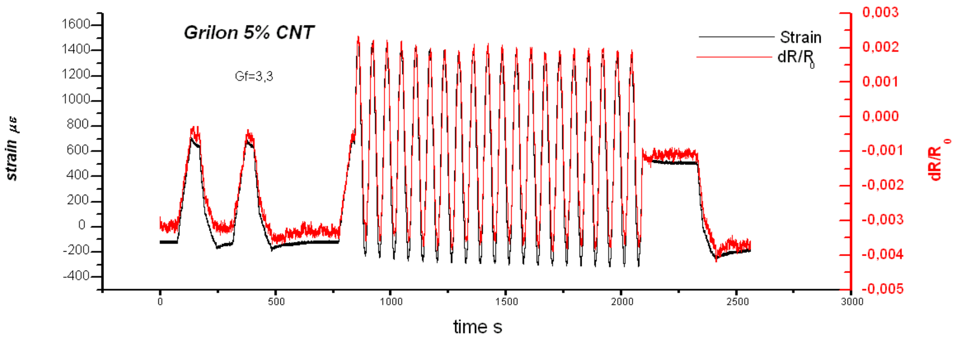
It's desiderble to have an high value for G_f.

Tests performed over the the sample charged with 5% of nanotubes have given:



b

a



c

Fig. 11 a – b- c *Tests for 5% CNT charged layer*

The Gauge Factor found for the sample with 5% of nanotubes is around 3.5, an interesting value useful for develop a strain sensing system.

2.2.2 Adhesion to epoxy matrix

The last test performed over the nanocharged thin sheet needs to verify the adhesion to the epoxy resin matrix in a fiber reinforced composite.

The properties of adhesion have been evaluated following the standard ASTM D 2344 that describe how to perform a Short Beam Strenght test on a laminate.

A layer of pristine Grilon of thickness of 147µm and another one of Grilon charghed with 5% of nanotubes and thick 156µm are put in the middle of a laminate of 20 carbon prepreg plies (Hexcel M18), in different zones and leaving a zone without any kind of layer for reference.

The laminate has been cured in autoclave under a vacuum bag as states in the fabrication guidelines of prepreg.

From each of three different areas has been cut 4 sample:

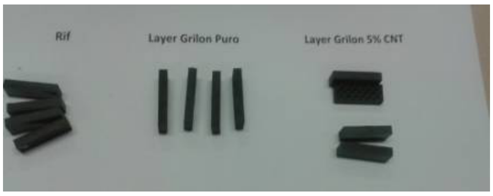


Fig.12: Samples for SBS tests

The tests have been performed using a mechanical test machine INSTRON 4505, the samples have been put over two edges and in the middle has been applied a load as shown in the following scheme.

Excerpt from ISBN 978-3-00-053387-7

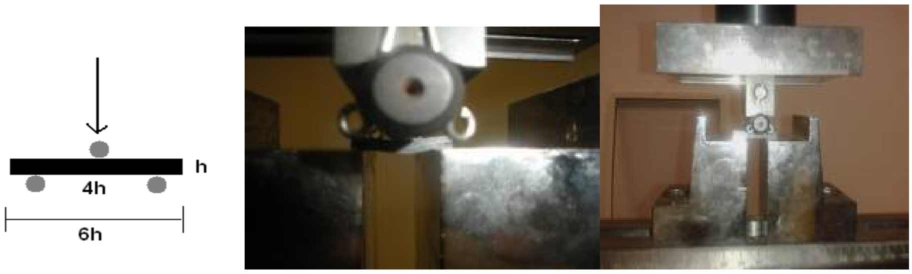


Fig13 SBS test procedure

From the maximum load applied during the test is possible to evaluate the Strength of short beam F_{SB}:

$$F_{SB} = 0.75 \frac{P_M}{b * h}$$

where P_M is the maximum load allied, b the width of the sample and h the thickness.

Results gives:

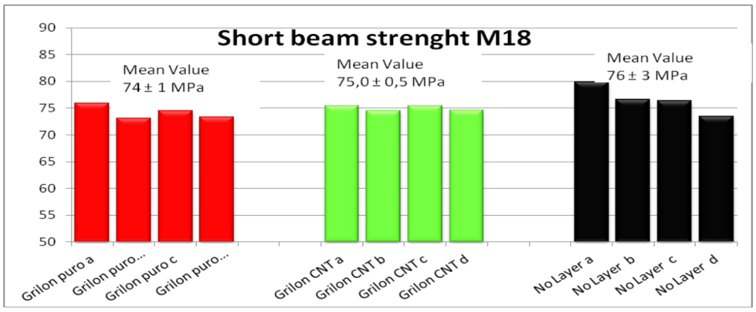


Fig.13 SBS Tests results

That values highlight a good adhesion of the nanocharghed layer to the matrix of the laminate and isn't source for delamination.

3 Panel within healt-monitoring embedded system

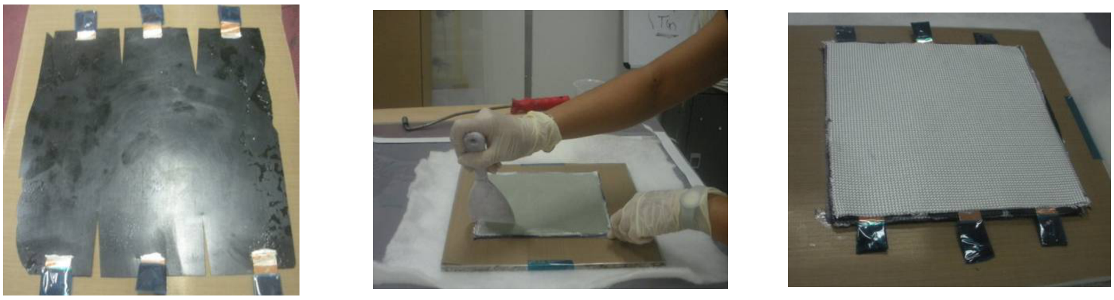
The integration of a nanocharged thin sheet in a composite has been the last step to verify the possibility to use it as an integrated healt-monitoring system.

A 20 x 20 cm panel has been manufactured by means of hand lay-up process, using glass fiber plies.

The nanocharged film with 5% of nanotubes has been put in the middle of the 24 plies of the panel.

The piezo-resistive sheet has three electrical contact in each lateral sides.

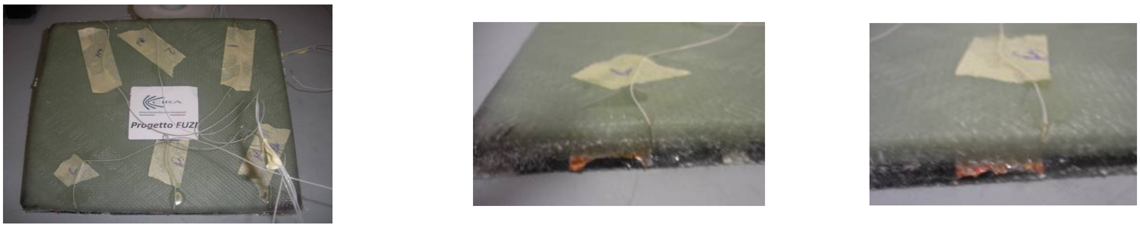
Excerpt from ISBN 978-3-00-053387-7



a) *b)* *c)*

Fig.14: Phases of manufacturing of glass fiber reinforced panel with nanocharged layer

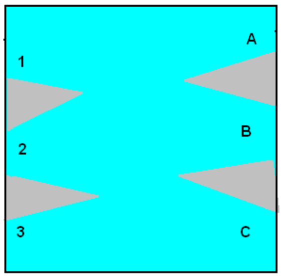
After the laminating phases the panel has been cured in autoclave under a vacuum bag. The final result is:



a) *b)* *c)*

Fig. 15 Final glass reinforced panel (a) contacts part. (b),(c)

The resistances between the different contacts have been measured:



Electrical resistance between different points of contacs (ohms)	
R1A=934,7	R1B=879,4
R2A=779,6	R2B=677,8
R3A=1051,1	R3B=919,8
R12=770,6	R13=1086,7
RAB=641,5	RAC=1172,4

Tab1:Values of electrical resistance between different points

The panel has been tested with a mechanical testing machine model MTS 810 with a load cell of 250 KN

Tests have been done bending the panel in a three point flexure mode.

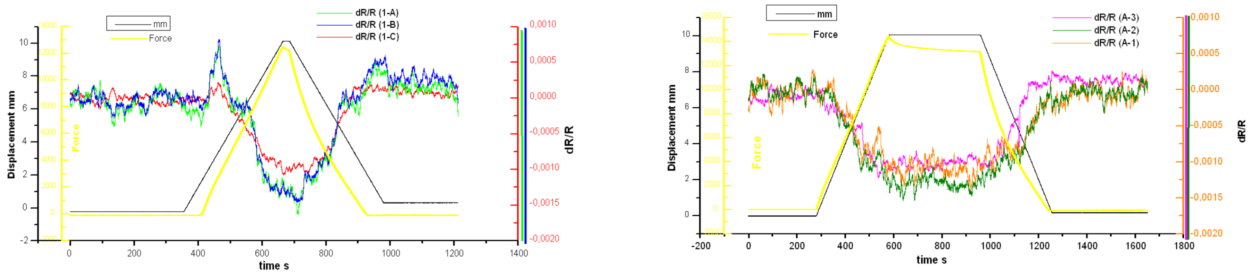
Excerpt from ISBN 978-3-00-053387-7

a ***b*** ***c***

Fig16 a b c: Electro-Mechanical set-up

During the tests a continuous current of 0,5mA flew between the points 1-C of the panel, and simultaneously were recorded the tension between the point 1-C, 1-A and 1-B, the displacement of the load cell and the force applied.

The results:



a ***b***

Fig. 17 ***a, b*** *Response of electrical resistance while a load has been applied*

Shows the variation of resistance follows the displacement and this means that the method could be used as an embended healt-monitoring system

4 *Conclutions*

It ' was manufacured a piezoresistive thin film based on a thermoplastic polymer which is suitable to the process of realization of composite materials, allowing to overcome the difficulties associated with the use of nanocharged resins.

Finally was tested the capability of strain sensing of that system and its real integrability into a composite.

Excerpt from ISBN 978-3-00-053387-7

5 *Acknowledgment*

The research leading to these results has received funding from the Italian *Ministry of* Education, *University* and *Research* (MIUR) under grand agreement DM 02_00029_3148467 called "Progetto FUZI" and managed by IMAST scarl.

Reference

[1] R. Volponi · P. Spena · F. De Nicola · A. Grilli «Multiscale composites within health-monitoring capabilities.» *ECCM16 - 16TH EUROPEAN CONFERENCE ON COMPOSITE MATERIALS, Seville, Spain, 22-26 June 2014*

[2]Alexopoulos, N.D., Bartholome, C., Poulin, P., Marioli-Riga, Z., 2010. "Structural health monitoring of glass fiber reinforced composites using embedded carbon nanotube (CNT) fibers". *Composites Science and Technology* 70, 260–271.

[3] Kang I, Schulz MJ, Lee JW, Choi GR, Jung JY, Choi JB, et al. "A carbon nanotube smart material for structural health monitoring". *Solid State Phenom 2007; 120: 289–96.*

[4] Gao, S.-L.; Zhuang, R.-C., Zhang, J., Liu, J.-W., & Mader, E. (2010). Glass fibers with carbon nanotube networks as multifunctional sensors. *Adv. Funct. Mater.*, Vol. 20, (May 2010), pp. 1885–1893, ISSN 1616-301X

[5] L. Guadagno, U. Vietri, M. Raimondo L. Vertuccio, G. Barra, B. De Vivo,P. Lamberti, G. Spinelli, V. Tucci, F. De Nicola R. Volponi, S. Russo "Correlation between electrical conductivity and manufacturing processes of nanofilled carbonfiber reinforced composites" *Composites Part B Engineering 05/2015; 80.+*

[6] Barra · F. De Nicola · B. De Vivo · L. Egiziano · L. Guadagno · P. Lamberti · M. Raimondo G. Spinelli · V. Tucci · L. Vertuccio · U. Vietri · R. Volponi " Enhanced electrical properties of carbon fiber reinforced composites obtained by an effective infusion process" *014 IEEE 9th Nanotechnology Materials and Devices Conference, NMDC 2014 12/2014;*

[7] L. Guadagno · M. Raimondo · U. Vietri · L. Vertuccio · G. Barra · B. De Vivo · P. Lamberti · G. Spinelli · V. Tucci · R. Volponi · G. Cosentino · F. De Nicola "Effective formulation and processing of nanofilled carbon fiber reinforced composites" *RSC Advances 12/2014; 5(8).*

[8] L. L. P. T. M. T. P. P. J. H. Doris W.Y. Wong, «Improved fracture toughness of carbon fibre/epoxy composite laminates using dissolvable thermoplastic fibres,» *Composites Part A: Applied Science and Manufacturing, pp. Pages 759-767, Volume 41, Issue 6, (June 2010)* .

[9] S. M. G. C. a. A. L. Gianluca Cicala, «Novel polymeric systems for high performance liquid molding technologies,» *Recent Res. Devel. Polymer Science, 11: ISBN: 978-81-7895-538-4, pp. 77-97, (2012).*

[10] K. B. W. M. Henne, "Reduction of Process Cycle Time and Improvement of Mechanical Properties of Composite Parts Manufactured in Resin Transfer Molding by Application of Grilon MS Binder Yarn", *SAMPE EUROPE 30th. International Jubilee Conference and Forum 2009.*

Excerpt from ISBN 978-3-00-053387-7