ON LINE AND IN-SITU CONSOLIDATION OF THERMOPLASTIC

R. Dell'Anna[1], F. Lionetto[1], F. Montagna[1] and A. Maffezzoli[1]

[1]Department of Engineering for Innovation, University of Salento, via per Monteroni, 73100 Lecce, Italy
Email: riccardo.dellanna@unisalento.it Web Page: http://mstg.unile.it

Keywords: Ultrasound, Thermoplastic, Composites, Filament Winding, Welding, Finite Elements

Abstract

High power ultrasound was used in order to mould thermoplastic composites in a process including the simultaneous fibers impregnation and plies consolidation. An experimental equipment, made of an ultrasonic device implemented on a filament winding machine, was developed. During winding, a titanium horn is put in contact with thermoplastic layer and its melting point is reached. Several cylindrical speciments were fabricated and their physical, mechanical and morphological properties were investigated.

Temperature field obtained during in-situ consolidation through heat generated by ultrasonic waves propagation was modeled using finite element (FE) analysis in order to optimize process parameters.

1. Introduction

The strong potential of continuous fibre-reinforced thermoplastic composites for high performance applications needs to find fast and efficient manufacturing processes. In comparison to thermosets, the use of thermoplastic matrix produces several advantages in term of process speed, elimination of volatile organic compounds (VOCs) and recyclability. One of the most promising approaches consists in the simultaneous deposition and consolidation of the plies with the application of heat in order to melt the thermoplastic matrix [1, 2]. The heat sources currently more used are i) hot air jets or flames, which are characterized by an acceptable cost but also by a limited ability to control the temperature in narrow ranges or ii) laser sources, much easier to control but more expensive [3]. Ultrasound waves, although used for welding of thermoplastic composites[4-6], have been also recently considered as heat source for on-line and in-situ consolidation. Ultrasonic assisted consolidation can be easily automated for the deposition of prepreg tapes or commingled yarns with thermoplastic matrix, both on flat and curved surfaces. The ability of ultrasound to detect the viscoelastic behavior of polymers [7-9] could be exploited also for the on-line monitoring of change of state of the matrix during composite consolidation processes.

Ultrasonic consolidation technique provides energy transfer from horn to the composite semi-preg at low amplitude (5–100 μm) and high frequency (20-40 kHz). Wave propagation produces surface and intermolecular friction which melt thermoplastic matrix [10]. As observed by Tolunay et al. [11], the heating occurs over the whole volume under the sonotrode tip.

In this work, high power ultrasound has been used to fabricate thermoplastic matrix composites, in a process including the simultaneous fibers impregnation and plies consolidation. An experimental equipment made of an ultrasonic device (horn), implemented on filament winding machine, and a compaction roller to applied consolidation pressure, has been developed. Both crystalline and amorphous thermoplastic polymers have been tested. During winding, horn is in conctact with joining parts for a time sufficient to reach the melting of the matrix. First, solid state and then viscous friction mechanisms induced by ultrasound are able to heat and melt the thermoplastic matrix. At the same time, the horn pressure ensures impregnation of reinforcement fibers. Ply consolidation is then promoted by the use of a consolidation roller which cools down the matrix while applying additional

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

pressure. The heat transfer phenomena occurring during the in-situ melting and consolidation have been simulated by finite element (FE) analysis, solving an energy balance accounting for the heat generated by ultrasonic wave propagation and the melting characteristics of the matrix. A characterization of the physical and mechanical properties of samples obtained with this equipment has been performed.

1. Material and methods

The developed ultrasonic equipment, schematically shown in Fig.1, consists of an ultrasonic welding equipment for thermoplastic polymers (Sonic Italia srl, maximum power:2000 W, frequency: 20 kHz), whose titanium horn is mounted on a filament winding machine [12] in order to produce cylindrical prototypes (inner diameter: 150 mm; variable length: 20-200 mm). Two materials have been used: Twintex R PP 60 1870 N (Fiber Glass Industries Inc.) and 57G-L-PET-524 (Comfil ApS). The first material is a dry commingled prepreg made of continuous E-glass rovings (60 wt %) and Polypropilene (PP) filaments. The second one is a composite tape made of E-glass fibers (57 wt%) impregnated with amorphous L-Polyethylene Terephthalate (L-PET).

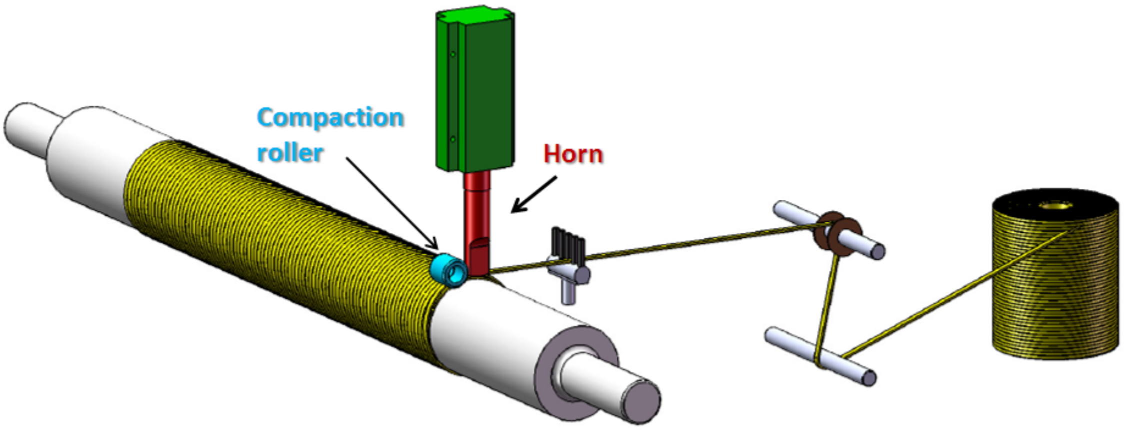

Figure 1. Ultrasonic consolidation set-up.

The temperature evolution during this innovative manufacturing process has been simulated using Comsol Multiphysics 4.4 Software. The effectiveness of the ultrasonic consolidation has been also assessed by physical, mechanical and morphological characterization [13] of the obtained cylinder prototypes.

3. Results and discussion

3.1 Modeling of the process

Simulation analysis has been used to obtain the temperature distribution in the composite, thus providing an efficient tool for process parameter optimization. The model has been solved with the finite element method using the commercial software Comsol Multiphysics 4.4.
Since composite thickness was very small compared to the mandrel diameter, a 2D Cartesian geometry has been used. Four domains have been considered: mandrel, horn, compaction roller and composite (Fig 2). The latter has been put in moving along X-direction to promote the conctact with other domains in a different times. The governing equations used to resolve the non-isothermal problem are the conservation equation for energy coupled with the equations accounting for the heat generated by ultrasonic waves and the heat absorbed by the melting of the matrix:

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

$$\rho Cp \frac{\partial T}{\partial t} = k\frac{\partial^2 T}{\partial x^2} + k\frac{\partial^2 T}{\partial y^2} + Q - \rho \dot{H}_m \tag{1}$$

where ρ is the density, Cp the specific heat capacity, T the Temperature and k is the thermal conductivity. Q represents the heat generation produced by ultrasonic heating according to the following equation [14]:

$$Q = \frac{\omega * \varepsilon_0^2 * E''}{2} \tag{2}$$

where $\omega = 2\pi f$, with f the ultrasonic frequency (20 kHz). The strain amplitude ε_0 is obtained as the ratio between the maximum displacement amplitude of the ultrasonic horn and the thickness of the commingled roving or composite tape under consolidation. The loss modulus E’’ is a measure of the energy dissipated through intermolecular friction.
The term $\dot{H}_m$ in equation 1, representing the heat necessary to promote the polymer melting, is a function of the degree of melting X_m, which can be expressed by the statistical approach of Greco and Maffezzoli [14]. Obviously, the term $\dot{H}_m$ in equation 1 is equal to zero in the case of E-glass/LPET tape sicne LPET is completely amorphous.

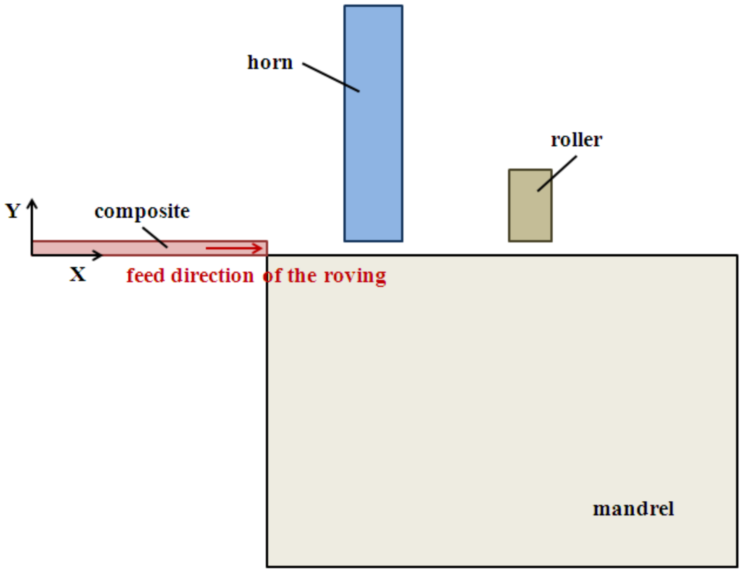

Figure 2. bidimensional geometry of the FEM model

The thermal conductivity k and heat capacity Cp of the composite in equation 1 are anisotropic properties with different values in the longitudinal and transversal direction. Moreover, k and Cp significantly vary with the temperature [12]. The values of thermal conductivity and heat capacity along the longitudinal axis have been determined by the rule of mixtures, while those along the transversal axis have been determined by the inverse rule of mixtures, commonly used also for the determination of mechanical properties of composite materials [16].
In this system, four boundary conditions have been set: 1) convective flux on the side surfaces of the horn, the mandrel and the roller and on the upper and lower surfaces of the composite; 2) constant temperature on the lower surface of the mandrel and on the upper surfaces of horn and roller; 3) continuity of temperature and heat flow between the moving surfaces in contact. The last condition is substituted to the first only when the composite is in contact with the mandrel and / or the horn and / or the roller. A mesh with triangular elements with the maximum dimensions smaller than one tenth of

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

the smallest composite thickness has been used. A time dependent study has been used with the Heat Transfer and moving mesh modules in Comsol Multiphysics 4.4 (Comsol Inc.).

The FE analysis has provided the temperature distribution in the composite during the impregnation and consolidation process. The prediction of the temperature distribution in the composite during processing is very important since it strongly affects the quality of the thermoplastic matrix composites. Three different winding velocity have been considered using Twintex-PP. The temperature maps for a velocity of 0.7 rad/s at different times, corresponding to different positions of the composite, are reported in Figure 3. In this figure, “H” and “R” indicate horn and roller respectively.

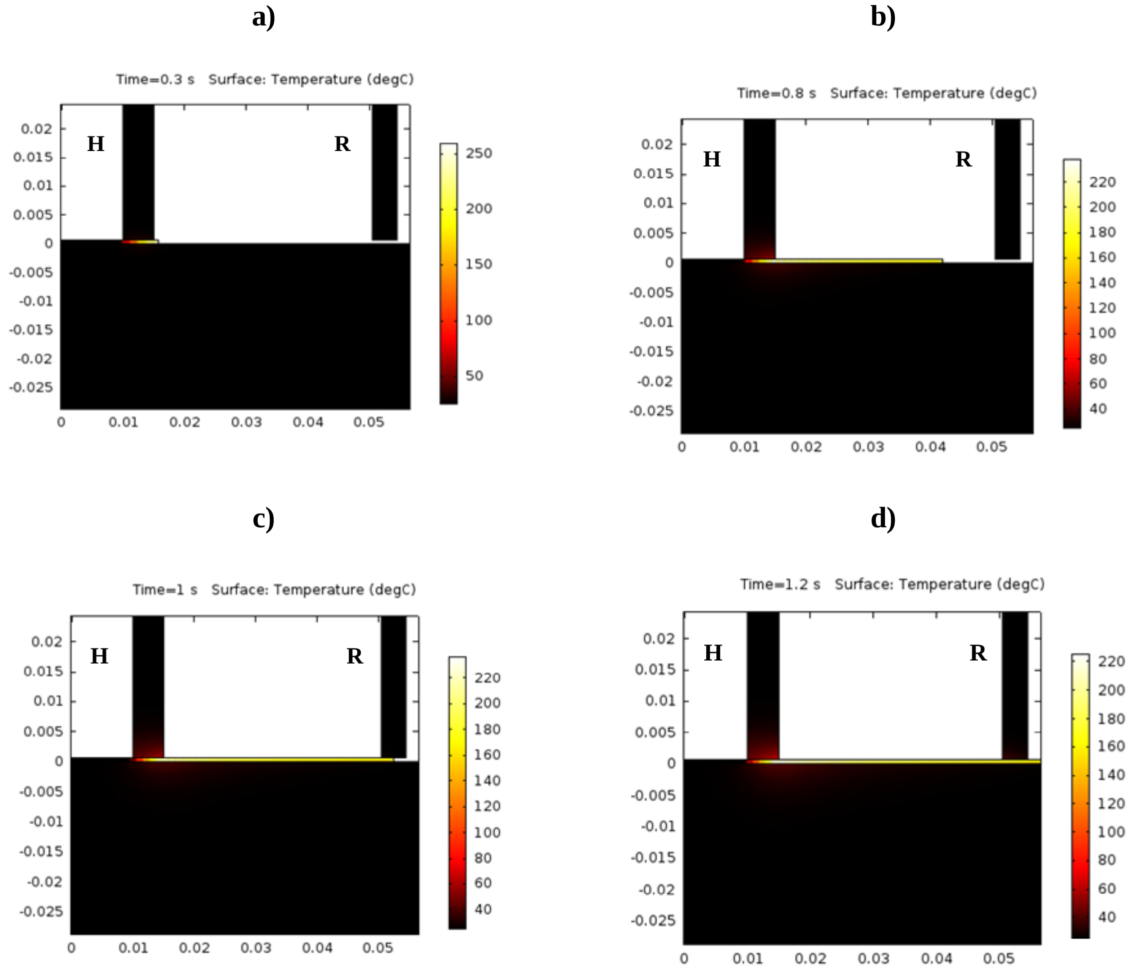

Figure 3. Temperature maps at different times for a composite speed of 52.5 mm/s.

As shown in Figure 4, for Twintex-PP, the temperature of composite domain starts to increase when the roving comes in contact with the horn, reaching a maximum values of 224°C at 52.5 mm/s at the exit of the contact with the sonotrode. Then, composite starts to cool down and a change of slope is observed when the sample goes under compaction roller, which is at room temperature. Simulations with the higher or lower winding speeds provided maximum temperatures too high or too low, respectively, in relation to the melting temperature of the polypropylene matrix. The FE results have been validated by the experimental measurement of the temperature. A good agreement between experimentally measured and simulated values has been found.

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

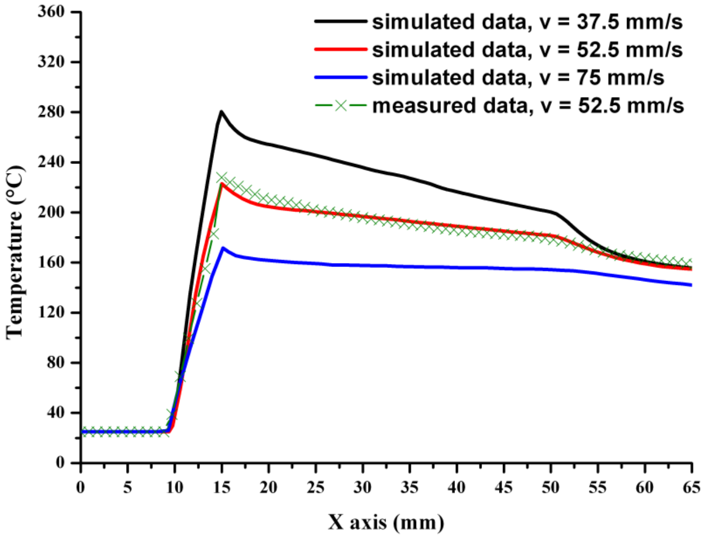

Figure 4. Simulated temperature distribution calculated in Twintex middle plain and along x direction at different winding speeds.

3.2 Results of sample manufacturing and characterization

Composite cylinders produced by ultrasonic assisted filament winding have been characterized by density measurements and dynamic mechanical analysis in torsion on specimens cut parallel to prototype axis (90° to fiber direction) so that the shear modulus G_{12}’ was measured. This represents a property dominated by the matrix and by fiber/matrix adhesion, providing useful data about the results of impregnation and consolidation processes. For comparison purposes, samples with the same number of layers have been consolidated by compression molding at 200 °C and 17 bar for 30 seconds.

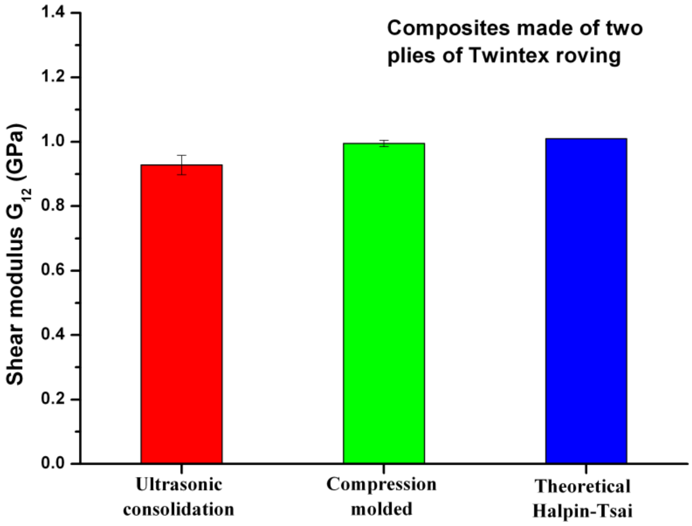

Figure 5. mechanical properties of Twintex consolidated (DMA analysis)

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

As observed from Figure 5, for composites made of 2 plies of twintex roving and consolidated with a velocity of 0.7 rad/s, the experimental values of G_{12}' are close to the theoretical one, 1010 MPa, which has been obtained using the Halpin-Tsai equations [16, 17] and those obtained for compression molded samples.

The void content of ultrasonically consolidated samples for Twintex-PP/E-glass composites obtained with a mandrel speed of 0.7 rad/s (3.6%) is within the typical range found in literature for composites processed by filament winding [1, 2, 18, 19] and compression moulding (20-22). For composites based of Comfil tape the degree of void is reported in Table 1.

Table 1. Void content of composites producted with Comfil tape.

Consolidated speed (rad/s)	Void content (%)
As received Comfil tape	1.91 ± 0.02
0.13	1.70 ± 0.04
0.15	2.86 ± 0.04
0.18	3.22 ± 0.03
0.26	4.32 ± 0.05
0.52	6.80 ± 0.10

It is clear that degree of void increase with mandrel speed. The results of mechanical and physical characterization indicate that the consolidation obtained by ultrasonic exposure has been satisfactory. During processing, ultrasonic heating has been able to melt the thermoplastic matrix and the contact time and pressure applied by horn and compaction roller have been able to obtain the impregnation of the fiber and consolidation of the laminate.

4. Conclusions

An experimental set-up, integrating a laboratory filament winding machine with the horn of an ultrasonic welding head, has been developed. The system has been equipped with a compaction roller to promote the impregnation of the fibers and the consolidation of plies. The propagation of ultrasonic waves has been used to achieve melting of the thermoplastic matrix, impregnation of the reinforcement fibers and, finally, consolidation of the different plies of different fiber reinforced thermoplastic composite materials.

The temperature distribution in the composite during exposure to ultrasound has been simulated by a finite element model accounting for the heat generated by ultrasound and for polymer melting. These modelling results are validated by experimental monitoring of the temperature using a thermocouple. The reliability of the developed system has been tested by producing some prototypes of composite with cylindrical shape. Characterization of these samples has shown low void content and high mechanical properties.

References

[1] F. Henninger, K. Friedrich. Thermoplastic filament winding with online-impregnation. Part A: process technology and operating efficiency. *Compos. A-Appl. Sci. Manuf.*, 33:1479-1486, 2002.

[2] R. Gennaro, F. Montagna, A. Maffezzoli, F. Fracasso, S. Fracasso. On-line consolidation of commingled polypropylene/glass roving during filament winding. *J. Thermoplast. Compos. Mater.*, 24:789-804, 2011.

[3] J. Tierney, J. W. Gillespie. Modeling of in-situ strength development for the thermoplastic composite tow placement process. *J. Compos. Mater.*, 40:1487-1506, 2006.

[4] I. Fernandez Villegas, H.E.N. Bersee. Ultrasonic welding of advanced thermoplastic composites: an investigation on energy-directing surfaces. *Adv. Polym. Technol.*, 29:112-121, 2010.

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

[5] I. Fernandez Villegas, L. Moser, A. Yousefpour, P.Mitschang, H.E.N. Bersee. Process and performance evaluation of ultrasonic, induction and resistance welding of advanced thermoplastic composites. *J. Thermoplast. Compos. Mater.*, 26:1007-1024, 2013.

[6] F. Ciampa, G. Scarselli, M. Meo. Nonlinear imaging method using second order phase symmetry analysis and inverse filtering. *Journal of Nondestructive Evaluation*, 34, 2:1-6, Springer US, 2015

[7] F. Lionetto, A. Sannino, G. Mensitieri, A. Maffezzoli. Evaluation of the degree of crosslinking of cellulose-based superabsorbent hydrogels: a comparison between different techniques. *Macromol. Symp.*, 200:199–207, 2003.

[8] F. Lionetto, A. Tarzia, A. Maffezzoli. Air-coupled ultrasound: A novel technique for monitoring the curing of thermosetting matrices. *IEEE Trans. Ultrason. Ferroelectr. Freq. Control*, 54:1437–1444, 2007.

[9] F. Lionetto, A. Maffezzoli. Monitoring the cure state of thermosetting resins by ultrasound. *Materials*, 6:3783-3804.

[10] C. Ávila-Orta, C. Espinoza-González, G. Martínez-Colunga, D. Bueno-Baqués, A. Maffezzoli, F. Lionetto. An overview of progress and current challenges in ultrasonic treatment of polymer melts. *Adv. Polym. Technol.*, 32:E582-E602, 2013.

[11] M.N. Tolunay, P. R. Dawson, K. K. Wang. Heating and bonding mechanisms in ultrasonic welding of thermoplastics. *Polym. Eng. Sci.*, 23:726-733, 1983

[12] F. Lionetto, R. Dell'Anna, F. Montagna, A. Maffezzoli. Ultrasonic assisted consolidation of commingled thermoplastic/glass fibers rovings. *Front. Materials*, 2:1-9, 2015.

[13] F. Lionetto, M. Frigione. Mechanical and natural durability properties of wood treated with a novel organic preservative/consolidant product. *Mater. Design,* 30:3303-3307, 2009.

[14] A. Benatar, Z. Cheng. Ultrasonic welding of thermoplastics in the far field. *Polym. Eng. Sci.,* 29: 1699–1704, 1989.

[15] A. Greco, A. Maffezzoli. Statistical and kinetic approaches for linear low-density polyethylene melting modeling. *J. Appl. Polym. Sci*., 89:289–295, 2003.

[16] F. Lionetto, E. Calò, F. Di Benedetto, D. Pisignano, A. Maffezzoli. A methodology to orient carbon nanotubes in a thermosetting matrix. *Compos. Sci. Technol*., 96:47-55, 2014.

[17] L. E. Nielsen, R. F. Landel. *Mechanical properties of polymers and composites*. Marcel Dekker, New York, 1994.

[18] L.A. Dobrzanski, J. Domagala, J.F. Silva. Application of Taguchi method in the optimisation of filament winding of thermoplastic composites. *Archives of Materials Science and Engineering*, 28:133-140, 2007.

[19] M. Stefanovska, B. Samakoski, S. Risteska, G. Maneski. Influence of some technological parameters on the content of voids in composite during on-line consolidation with filament winding technology. *Intern. J. Chem., Nuclear, Mater. Metallurgic. Engin.*, 8:398-402, 2014.

[20] A.C. Long, C.E. Wilks, C.D. Rudd. Experimental characterisation of the consolidation of a commingled glass/polypropylene composite. *Compos. Sci. Technol*., 61:1591–1603, 2001.

[21] C. Santulli, R.G. Gil, A.C. Long, M.J. Clifford,. Void content measurements in commingled E-glass/polypropylene composites using image analysis from optical micrographs. *Sci. Engin. Compos. Mater.*, 10:77-90, 2002.

[22] A. Greco, C. Musardo, A. Maffezzoli. Flexural creep behaviour of PP matrix woven composite. *Compos. Sci. Technol.,* 67:1148–1158, 2007.

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