ISSN 1392–1320 MATERIALS SCIENCE (MEDŽIAGOTYRA). Vol. 20, No. 1. 2014

Influence of Coatings on Tensile Properties of Glass Fiber

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crossref http://dx.doi.org/10.5755/j01.ms.20.1.3432

Received 04 February 2013; accepted 03 September 2013

Glass fibers (GF) are widely used as a reinforcing material for many polymer products; to form very strong and light weight materials. Surface flaw generated by contact makes the glass fiber highly sensitive. Defects or cracks in the surface of glass fiber threaten the mechanical strength of the fiber that deteriorates the durability of glass fiber. Coating can play crucial role by forming single or multiple molecular layers on the glass fiber in rectifying the surface flaws and modify surface properties of glass fiber either online or offline. In this experiment, coating was introduced on the fiber surface by continuous impregnation of fiber with a solution of polymer in a padder in order to improve the mechanical strength of glass fibers. The tensile properties of GF roving after coating were studied. The experimental results showed that the tensile properties of coated GF roving were improved significantly compared to uncoated GF roving.
Keywords: glass fiber, coating, polysilazane, polysiloxane, composite.

1. INTRODUCTION

In high performance composite manufacturing industries, glass fibers are one of the most important reinforcements with excellent mechanical properties [1]. This properties offer huge advantages over other types of construction materials. Glass fiber is a versatile and cost-effective reinforcement for composites. It is lightweight, corrosion resistant [2]. The interfacial bond between the glass fibers and the matrix resin is optimizing composite properties [3]. They are mainly using for manufacturing technical textile such as automotive textiles, sport textiles, aerospace industry, wind turbine blades, textile reinforcement concrete, due to their high profile physical and chemical characteristics [4]. Many researchers [5 – 7] have studied the influence of polymer-glass fiber composites where some glass fibers are used as the reinforcing materials. Glass fibers whose surface has been modified by first using a coupling agent and then mixing with the polymer can improve the composite impact resistance [8] and shear strength. The level of adhesion between fiber and matrix affects the ultimate mechanical properties of a composite. To achieve optimum mechanical properties they were coated with different polymer materials. The prime requirements for optical fiber coatings were protection against micro-bending and static fatigue. This necessitated that cured coatings be concentric about the fiber, be continuous over the length of application, be of constant thickness, be abrasion resistant and moisture retardant [9]. Lot amount of various attempts were made to investigate and to increase resistance of glass fibre/polymer composites, using various kinds of polymers, such as epoxy resins [10], vinyl- [11] and polyesters [12]. Fiber fatigue is an important mechanical property of optical fibers. Fiber fatigue is thought to occur by crack growth of existing flaws on the glass surface, due to interaction between the Si-O bond and the moisture in the environment, when the fiber is subjected to stress. The studies on failure mechanisms [13 – 16] showed that under loading of tensile stress, the cracks start at the fiber ends and propagate along the fiber-matrix interface or cross through the matrix and finally the failure takes place. The coating is thought to contribute to fiber fatigue in that basic compounds, present in the composition, accelerate glass corrosion. Conversely, acidic components have been shown to improve fatigue resistance. Coatings with strong adhesion facilitated a greater n-value fatigue parameter for coated fibers [17]. It is imperative to apply concentric coating layers, to prevent damage to the fiber during the drawing operation, and to maximize fiber strength and micro-bending resistance. Non-concentric coating application puts differential stress on the fiber when cured. An unevenly coated fiber will experience non-uniform forces during periods of coating expansion and contraction and is susceptible to greater attenuation of light signals passing though the fiber. Silane coupling agents are applied to glass fiber surface to promote their adhesion to the polymer matrix. They have the general formula $Y\text{-}Si(X)_3$, where X represents alkoxy groups or chlorine which, after hydrolyzing to silanol, react with silanol groups present on the surface of glass fibers to form siloxane linkages and Y represents nonhydrolyzable organo functional groups such as amino and methacrylate that react with the matrix functional groups. Organic coatings undergo thermal oxidation, photo-initiated oxidation and chemical attack when exposed to the environment resulting in a degradation of properties, which in final coatings will include gloss and color loss as well as embrittlement and potential loss of adhesion to the base coating. Inorganic coating polymers such as polysiloxane and polysilazane on the other hand are much more resistant to these degradation mechanisms. Polysilazane polymers whose backbones

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consist of alternating Si-N bonds with pendent carbon-containing groups are widely used as precursors of silicon carbonitride ceramics. Thermal, mechanical and electrical properties of the obtained SiCN ceramics render them very suitable for high temperature applications. Polysilazane can for example be used as protective barrier for heat exchanger and on steel against oxidation [18]. Polysiloxane, with Si-O-Si backbone shows excellent heat and UV radiation stability, low-temperature flexibility, good hydrophobicity, and excellent moisture resistance [19]. In this experimental study, four types of polymer from three different companies were used to coat the surface of GF. Finally, coating percentage and stress-strain behaviour of the GF is analyzed.

2. MATERIALS AND METHODOLOGY

2.1. Polymer

The GF roving was used, which was received from Saint-Gobain IVW Cem-FIL GmbH, Düsseldorf, Germany. The basic yarn type alkali resistant GF roving LTR ACR 640 5325 of 640 tex, having f1600 in each roving (filament diameter 14 μm), was used as a substrate in the whole work. The material was used without any pre-treatment.

2.2. Coating materials

In this experiment, polysiloxane and polysilazane based polymers shown in Table 1 were used for coating of GF roving by continuous dip coating process. The chemical structure of these two polymers is shown in the Figure 1.

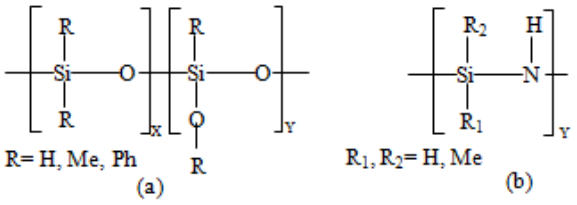

Fig. 1. Chemical structure of polysiloxane (a), polysilazane (b)

Table 1. List of polymers used for coating

Polymer	Type	Characteristics	Manufacturer
Silikophen P80/MPA	Phenylmethyl polysiloxane resin	Liquid (solids content 80 % by weight)	Evonik Tego Chemie GmbH
Silres K	Methyl silicone resin	Solution at toluene (50 % by weight)	Wacker Chemie AG
Silres IC836	Phenyl polysiloxane	100 % Solid	Wacker Chemie AG
KiON® HTT 1800	Polysilazane	100 % solids as liquid	Clariant GmbH

2.3. Solvent

Acetic acid ester (ester) supplied by Biesterfeld, Germany was used as the solvent. It was a clear, colourless, and low viscosity organic solvent available in liquid form.

2.4. Coating procedure of glass roving

Coating experiments were carried out by using 'Coatema Basecoater BC32' laboratory coating machine in ITM, TU Dresden. Mentioned coating polymers were applied continuously by impregnating the GF roving in a padder. The most typical padder was equipped in the coating machine for this purpose. A schematic representation of coating machine used in this experiment and working flow chart is presented in Figure 2 [20].

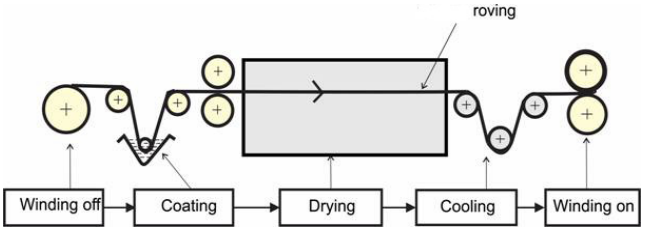

Fig. 2. Schematic representation of working principle of coating

Different parameters such as machine speed, drying and curing time, and curing temperature were maintained during the coating procedures which are represented in Table 2 below. The detail recipes used for coating of GF are shown in Table 3.

Table 2. Parameters of 'Coatema Basecoater BC32' machine

Parameters	Value
Machine speed	1 meter/min
Drying and curing time	1 min
Curing temperature	205 ºC

Table 3. Formulation of coating recipes

Recipe number	Chemicals	Amount (by weight %)	Substrate
1	Silikophen P80/MPA	15	AR GF (640tex)
	Acetic acid ester	85	
2	Silres K	20	AR GF (640tex
	Acetic acid ester	80	
3	Silres IC 836	15	AR GF (640tex)
	Acetic acid ester	85	
4	KiON® HTT 1800	15	AR GF (640tex)
	Acetic acid ester	85	

Finally, the roving was cured and dried in the drying unit at the required time and temperature. Polymers were cross-linked to each other and formed a siloxane (SiOSi) film on the top of the glass surface during the curing process. The mechanism of polysiloxane coating is shown in the Figure 3 and mechanism of adhesion of polysilazane is shown in the Figure 4.

2.5. Determination of coating percentage

The amount of coating percentage (%) of GF roving was determined by weighing the sample before and after coating. The following simple equation was used to calculate the coating percentage.

$$\text{Coating } [\%] = \frac{w_a - w_b}{w_b} \times 100\,, \qquad (1)$$

where w_a – is the weight of GF roving of five meters length after coating; w_b – is the weight of GF roving of five meters length before coating.

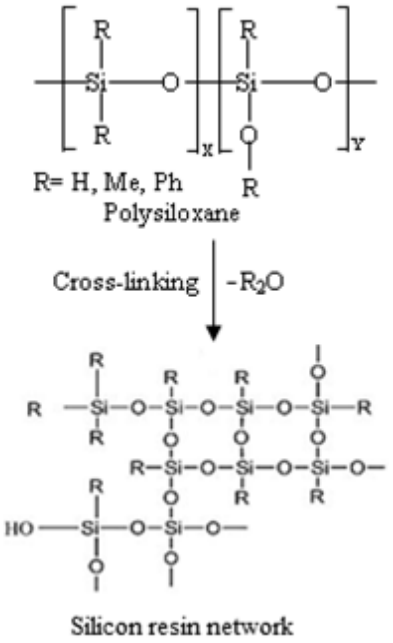

Fig. 3. Chemistry of polysiloxane polymer coating

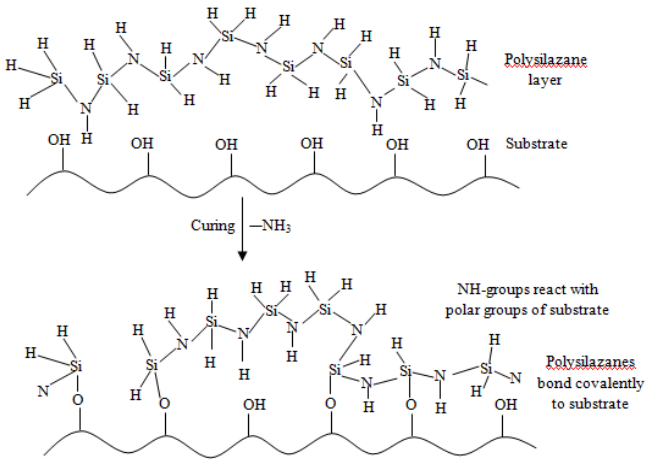

Fig. 4. Mechanism of adhesion of polysilazane

Before coating, weight of five meters GF roving was measured. Then coating material was applied on sample according to recipe and parameters. The sample was dried at 205 °C temperature. After drying, the weight of the sample was measured again. The difference between the two weights was calculated. The coating percentage of GF roving was calculated by the above mentioned equation. To get optimum result, 3 samples of each item were measured and average value was calculated.

2.6. Investigation of stress-strain behaviour

The tensile tests of polymer coated GFs roving's were carried out based on the international standard ISO 3341 at room temperature by using Zwick-Z100 material testing machine to determine the effect polymer coating on the mechanical properties of the fibers presented in Figure 5. The tensile strength, stress-strain behaviour as well as E-module of the original, polymer coated GF were investigated. To compare different samples, values of stress (Force per unit area) and strain (deformation %) were used. The tensile stress (MPa) versus elongation (%) was recorded during testing and 10 measurements for each sample were taken to average value of the results.

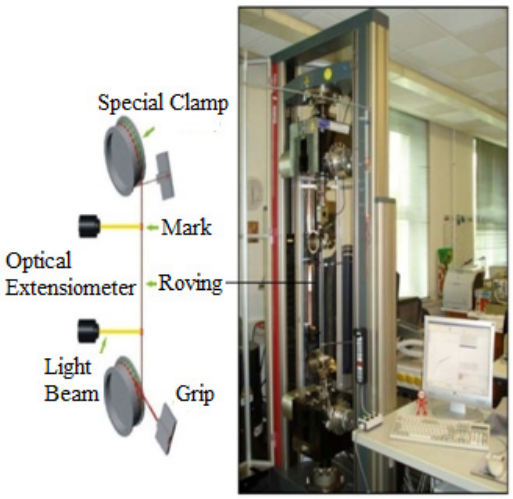

Fig. 5. Actual set up of Zwick material testing machine at ITM, TU Dresden

The following two equations were used to calculate stress and strain values of sample.

$$\text{Stress [MPa]} = \frac{F_{\max} \times \rho \times 1000}{\text{Tex}}, \quad (2)$$

where $F_{\max}$ – is the breaking force (N), ρ – is the density of GF, 'Tex ' – is the fineness of AF roving (g/1000 m).

$$\text{Strain [\%]} = \frac{\Delta}{l} \times 100, \quad (3)$$

where, Δ – is the change of length of roving due to stress (mm), l – initial length of AF roving (mm).

3. RESULTS AND DISCUSION

The amount of coating percentage on GF roving was determined with the help of Equation 1. The coating percentages according to sequence of sample number (R1, R2, R3 and R4) are summarized in Table 4.

Glass fiber is coated with a size containing silane coupling agent, which is hydrolyzed to silanols. Silanol groups form a chemical bond through intermolecular reaction while polymeric silane deposit takes place on the GF surface due to the intra-molecular reaction within the hydrolyzed coupling agents. Figure 6 below depicts the coating percentages and % of polymer by weight on different samples of GF. The highest percentage of polymer is observed in sample 2. On the other hand coating percentage of the same sample reached the lowest value almost 6 %. Other three samples contains same amount of polymer approximately 15 percentages. Different coating percentage is found in all samples. Coating percentage on sample 4 is highest, which is 12 %. Coating percentage on sample 1 and 2 is differing by almost 2 %. Although the overall polymer percentage of four samples is same except sample number 2, the coating percentage fluctuates randomly in case all samples. It may

be occurred due to lower solid content as well as lower viscosity of this polymer. Beside this, surface smoothness plays a major role in adhesion of polymers. If the surface is modified by radiation then adhesion percentage of polymers will be increased. Radiation can increase markedly the surface energy of synthetic fibers, improving the mechanical characteristics of the final products. In addition, functional groups present in coating materials accelerate the adhesion percentage.

Table 4. Amount of coating percentages of GF roving

Sample	Weight before coating (gram)	Weight after coating (gram)	Coating %	Average coating %
R1	3.32	3.63	9.337	10.24
	3.32	3.68	10.843	
	3.32	3.67	10.542	
R2	3.32	3.53	6.325	6.82
	3.32	3.54	6.626	
	3.32	3.57	7.531	
R3	3.32	3.64	9.638	9.27
	3.32	3.65	9.739	
	3.32	3.60	8.433	
R4	3.32	3.69	11.14	11.85
	3.32	3.70	11.44	
	3.32	3.75	12.95	

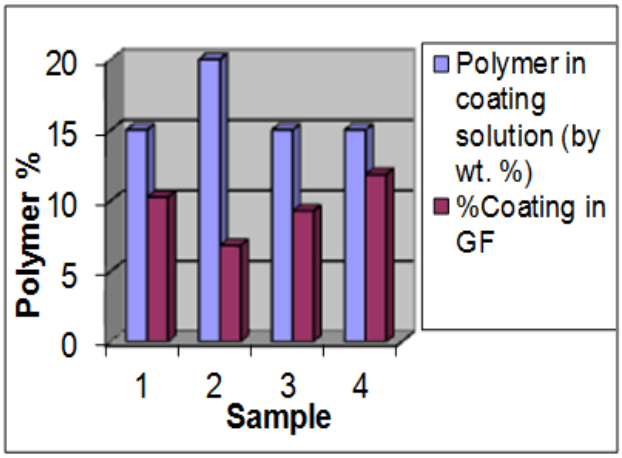

Fig. 6. Comparison of coating % on GF roving

The results of tensile test of various coated GFs are summarized and presented below at Table 5.

Table 5. Tensile properties of GF Roving

Sample	Breaking force (N)	E-Module (GPa)	Elongation at break (%)
Original AR GF	281 ±11.50	44.3 ±7.15	2.01 ±0.11
R1	470 ±14.50	64.4 ±4.14	2.84 ±0.10
R2	434 ±27.20	67.8 ±3.70	2.66 ±0.17
R3	396 ±18.40	69.6 ±2.97	2.45 ±0.11
R4	405 ±53.00	67.7 ±3.39	2.43 ±0.33

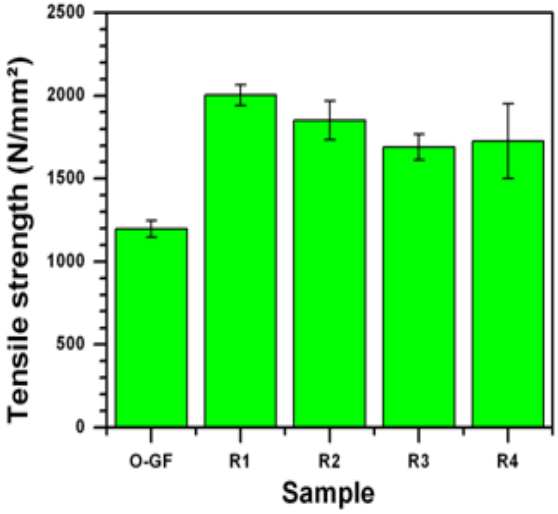

Fig. 7. Comparison of tensile strength of coated samples

From the Figure 7, it is clearly seen that coated GF roving has better tensile properties compare to original GF roving. Tensile properties decrease gradually from coated sample 1 to sample 3 and then it rise slightly in case of sample 4. A peek in tensile strength is observed in first sample. Conversely, sample 3 has the lowest tensile strength. Recipe R1 increased 67 % tensile strength compare to original GF. Hoferek et. al showed that composites fabricated from plasma coated glass fibers showed an increase of adhesion by 40 % with respect to unsized reinforcements [21]. Gao and Mäder found the same results for the sized glass fibers [22]. They also showed that variation of sizing properties strongly affects the fibre fracture behaviour because the critical flaws which limit the strength of fibres are located at the surface.

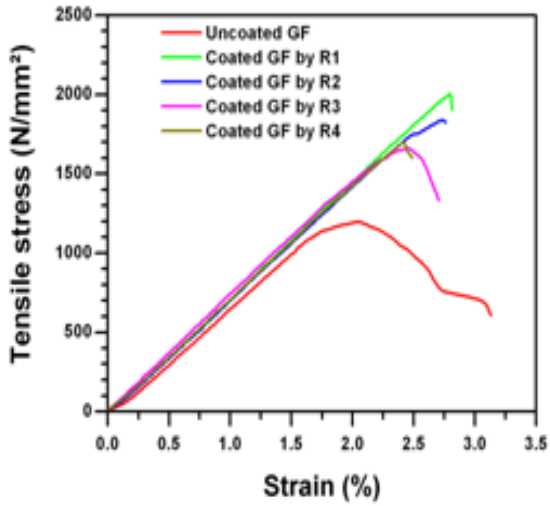

Fig. 8. Comparison of stress-strain curve of different coated glass roving

This increase of tensile strength may be caused due to the bonding of individual fibers each other with polymer matrix, where every single fibers acted as active fiber in roving and reduce the fiber defects. Besides, different polymer coating increased the filament-filament micro friction/adhesion within the roving. Thomason showed that chemical bonding between the fiber surface and polymers improves when the glass fiber surface is modified by

coupling agent [23]. As a result, interfacial strength and hydrothermal resistance to the surface improves. The fibers in the coating can induce stress relaxation, eliminate stress propagation and residual stress and reduce any internal stress shown by Mehan and Schadler [24].

From the stress-strain curves shown in Figure 8 above, it is noticed that the different polymer coated GF roving showed better result than the untreated (original) GF roving. The curves of the polymer coated GF roving were more linear than uncoated GF rovings. Fibers coated with recipe R1 showed highest stress-strain property comapre to other samples. Almost there is no difference in stree-strain property in sample R2 and R4. Third sample shows a slight decrease in tensile modulus when compared to other samples after coating with polymer.

4. CONCLUSION

In this study, the mechanical properties of GF coated with various polymers were investigated and obtained satisfactory result for tensile strength and stress-strain properties. The variation in coating recipes plays significant roles on tensile properties of GF. The experimental results of this study proved that polymer coating can improve the mechanical properties of GF. Tensile strength of GF roving coated with Phenylmethyl polysiloxane resin increased 67 % compare to original GF roving. The curves of stress-strain behavior of coated glass roving were more linear compare to uncoated GF roving. Finally, it can be said that these polymers would be a promising materials for coating GF roving. It would be advantageous to study further the results of different blends ratio of silanes and do more analysis like SEM, TGA for better understanding the results. Environmental impacts on the materials could be analyzed in order to improve environmental durability which is challenging for coated GF.

REFERENCES

1. **Shayed, M. A., Cherif, Ch., Hund, R. D., Cheng, T.** Carbon and Glass Fibers Modified by Polysilazane Based Thermal Resistant Coating *Textile Research Journal* 80 (11) 2010: pp. 1118 – 1128.
2. **Hearle, J. W. S.** High-performance Fibers. UK: Woodhead Publishing Ltd. ISBN 1-85573-539-3. 2000: pp. 213 – 219.
3. **Zhao, F. M., Takeda, N.** Effect of Interfacial Adhesion and Statistical Fiber Strength on Tensile Strength of Unidirectional Glass Fiber/Epoxy Composites. Part I: Experiment Results *Journal of Composites A* 31 2000: pp. 1203 – 1214. http://dx.doi.org/10.1016/S1359-835X(00)00085-3
4. **Wallenberger, F. T., Bingham, P. A.** Fiberglass and Glass Technology: Energy-Friendly Compositions and Applications. Springer. ISBN 978-1-4419-0735-6. 2010: pp. 124 – 135. http://dx.doi.org/10.1007/978-1-4419-0736-3
5. **Alvarez, V. A., Valdez, M. E., Vázquez, A.** Dynamic Mechanical Properties and Interphase Fiber/Matrix Evaluation of Unidirectional Glass Fiber/Epoxy Composites *Polymer Testing* 22 2003: pp. 611 – 615. http://dx.doi.org/10.1016/S0142-9418(02)00164-2
6. **Thomason, J. L.** Structure-Property Relationships in Glass-reinforced Polyamide. Part 1: The Effects of Fiber Content *Polymer Composites* 27 2006: pp. 487 – 498.
7. **Thomason, J. L.** Structure-property Relationship in Glass-reinforced Polyamide, Part 3: Effects of Hydrolysis Ageing on the Dimensional Stability and Performance of Short Glass-fiber-reinforced Polyamide 6.6 *Polymer Composites* 28 2007: pp. 344 – 354. http://dx.doi.org/10.1002/pc.20312
8. **Zaretsky, E., Botton, G. D., Perl, M.** The Response of Glass Fibers Reinforced Epoxy Composite to an Impact Loading *International Journal of Solids and Structures* 41 2004: pp. 569 – 584.
9. **Stevens, J. M., Keough, A.** The Application OF UV Coatings to Glass Optical Fiber *The Association for Finishing Processes of SME* FC78-551. 1978.
10. **Ellyin, F., Maser, R.** Environmental Effects on the Mechanical Properties of Glass-fiber Epoxy Composite Tubular Specimens *Composites Science and Technology* 64 2004: pp. 1863 – 1874.
11. **Roy, R., Sarkar, B. K., Bose, N. R.** Effects of Moisture on the Mechanical Properties of Glass Fiber Reinforced Vinyl Ester Resin Composites *Bulletin of Material Science* 24 2001: pp. 87 – 94.
12. **Abbasi, A., Hogg, P. J.** Temperature and Environmental Effects on Glass Fiber Rebar: Modulus, Strength and Interfacial Bond Strength with Concrete *Composites B* 36 2005: pp. 394 – 404.
13. **Curtis, P. T., Bader, M. G., Bailey, J. E.** The Stiffness and Strength of a Polyamide Thermoplastic Reinforced with Glass and Carbon Fibers *Journal of Material Science* 13 1978: pp. 377 – 90.
14. **Sato, N., Kurauchi, T., Sato, S., Kamigaito, O.** Microfailure Behaviour of Randomly Dispersed Short Fiber Reinforced Thermoplastic Composites Obtained by Direct SEM Observation *Journal of Material Science* 26 1991: pp. 3891 – 3898. http://dx.doi.org/10.1007/BF01184987
15. **Sato, N., Kurauchi, T., Sato, S., Kamigaito, O.** Mechanism of Fracture of Short Glass Fiber-reinforced Polyamide Thermoplastic *Journal of Material Science* 19 1984: pp. 1145 – 52.
16. **Takahashi, K., Choi, N. S.** Influence of Fiber Weight Fraction on the Failure Mechanisms of Poly(ethylene terephthalate) Reinforced by Short-glass Fibers *Journal of Material Science* 26 1991: pp. 4648 – 4656.
17. **Skutnik, B. J., Munsey, B. D., Brucker, C. T.** Coating Adhesion Effects on Fiber Strength and Fatigue Properties *Materials Research Symposia Proceedings* 88 1986: pp. 27 – 34.
18. **Gardelle, B., Duquesne, S., Vu, C., Bourbigot, S.** Thermal Degradation and Fire Performance of Polysilazane-based Coatings *Thermochimica Acta* 519 2011: pp. 28 – 37.
19. **Lee, S.** A Correlation between the Optical and Mechanical Properties of Novel Polysilane/Polysiloxane Nanocomposites *Polymer Bulletin* 63 2009: pp. 385 – 396. http://dx.doi.org/10.1007/s00289-009-0093-4
20. **Rana, M.** Coating of Technical Textiles. *Master Thesis* ITM, TU Dresden. Vol. 2962. 2010: 27 p.
21. **Hoferek, L., Janecek, P., Cech, V.** Plasma Coating of Glass Fibers Used for Polymer Composites *Proceedings of the 19th International Symposium on Plasma Chemistry* Poster Session 1, ID: P1.8.35 2009.
22. **Gao, S.L., Mäder, E.** Chapter 36: Advances in Nanocomposites - Synthesis, Characterization and Industrial Applications. Edited by Reddy, B. *InTech Publisher* 2011: 842 p.
23. **Thomason, J. L.** The Interface Region in Glass Fiber-reinforced Epoxy Resin Composites: 1. Sample Preparation, Void Content and Interfacial Strength *Composites* 26 1995: pp. 467 – 475.
24. **Mehan, M. L., Schadler, L. S.** Micromechanical Behavior of Short-fiber Polymer Composites *Composites Science and Technology* 60 2000: pp. 1013 – 1026.