Influence of process parameters in production of resin film on kevlar fabric prepreg

Biljana Kostadinoska[*], Svetlna Risteska[*], Blagoja Samakoski[*], Samoil Samak[+], Nikolce Romevski [+]

Institute for Advanced Composites and Robotics (IACR)
Krusevski pat bb - Prilep, R.N. Macedonia
biljanak@iacr.edu.mk; svetlanar@iacr.edu.mk; bsamakoski@ iacr.edu.mk;
[+]*MIKROSAM D.O. O.*
Krusevski pat bb Prilep, R. N. Macedonia
ssamak@mail.mikrosam.com;nikolcer@mail.mikrosam.com

***Abstract*— The optimization of parameters for the Impregnation process in order to produce high-quality prepregs with high impregnation quality is challenging. The purpose of the study is to access the applicability of full factorial experimental design 2^3 in predicting properties of polymer composite prepregs. This paper presents an experimental study of the impregnation of aramid cloth with thermosetting resin matrix. Experiments (DoE) method is used to create quality prepreg and to find optimized input parameters for this technology. The major benefits gained of the current study are that the thickness of film is an important parameter, which has a prominent effect on the final product. A model is developed to describe the resin content as functions from thickness film, temperature and impregnation velocity in horizontal impregnating process. Results are obtained methodically through the separation of variables.**

The methods and results presented in this paper offer a valuable insight of how to measure the quality and how to use this information for the optimization of Impregnation processes Experimental studies were performed with changing parameters – temperature, thickness of film and speed. Finally, it describes the content of resin inside of the composites. The resin content is controlled by a combination of the thickness resin film, the impregnation velocity and the oven temperature. These properties are good enough for aramid fabric prepreg to be used as engineering materials in many structural applications.

***Keywords*— DoE, impregnation velocity, thickness resin film, temperature, fabric prepreg**

I. INTRODUCTION

In many industries, e.g. in aerospace, prepreg has become the standard technology for making fibre-reinforced plastic (FRP) structures; in other industries, too, its use is continuously increasing. Rather than impregnating the reinforcement fibres with resin during the manufacture of the final component or structure, as is the case for the hand lamination technique, or for infusion and injection techniques, the fibres are resin-impregnated in a separate process. This pre-impregnation takes place prior to the actual component manufacture, in order to create a semi-finished product which can be transported and stored in rolls - the prepreg [1].

Prepregs allow for higher mechanical strength and stiffness properties, which is in part due to the more homogeneous and straighter fibre arrangement.

Over the last 70 years, prepregs have significantly influenced the technological development of high-performance fiber reinforced components. Today, these mate trials are globally prevalent and are used for the manufacture of composite parts in the aerospace industry, for high-speed trains, cars, boats, and many other applications. More than half of the global carbon fiber production is used to manufacture prepregs. The goal to develop a prepreg material that was easily manufactured and processed inspired numerous research projects and studies. To this day, research efforts are focused on the improvement and further development of prepreg materials [2], [3]. There are four methods of impregnation:

- solution dip;
- solution spray,
- direct hot-melt coat; and
- film calendering.

The resin formulation can be cast into a film from either hot-melt or solution, and then stored. Thereafter, in a separate process, the reinforcing fibre is sandwiched between two trims and calendered so that the film is worked into the fibre. The machines necessary to accomplish these procedures are many and varied [4]-[7]. In this paper, the third and fourth method are studied.

The proposed in situ method monitors the pattern of resin saturating the empty spaces between the fibers and the tows within the fabric over time, which allows for characterization of such prepregs. The experimental technique compresses a resin film into the fabric at a known temperature and pressure [8]-[12]. After the production of prepreg, the quality of the produced prepreg is determined [13]. The quality of the prepreg is checked by several standards [14]-[19].

In order to optimize the quality of fabric prepregs and its manufacturing process, the influence of temperature of ovens, thickness of resin film and speed of process on the content of resin was determined. The content of resin in prepregs is increases or decreases. A reason for this behaviour, change of process parameters. Experiments from these changes are given

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in following paper. In order to make these experiments is used DOE analysis [20]-[23].

They were made 8 combinations and each combination with 5 samples was tested as shown in Table III.

II. Experimental

A. *Materials and equipment*

For the manufacture of fabric prepreg were used following materials:

- Aramid fabric - Kevlar fabric Style 258 Heatest, 410gsm, wide 1000mm,
- KS 1200 natural color paper, silicone coating double side with wide 1100 mm and 120 gsm,
- Phenolic resin. The applicable resin system is liquid phenolic resin. The dynamic viscosity of the resin mix, including solvent at the room temperature is in the range of 500-900 mPas.

In this study, samples were obtained by using an equipment for manufacture prepreg (direct film coat from resin), manufactured with machine from Mikrosam, Macedonia. Coating (Impregnation) machine is designed as single step process for manufacturing of aramid fabric prepreg materials as primary process. The coating concept is with utilization of the knife coating on the release paper to prepare resin film and later to perform lamination step on the fabric.

The basic diagram of the process is presented on the following Fig 1.

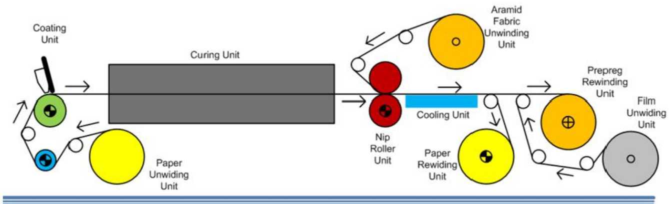

Fig. 1 Prepreg making system

B. *Prepreg making equipment*

The Station is Equipped with single-shaft un-winder for unwinding the release paper tape. The coating unit is designed for knife coating method of liquid resins. The unit includes coating squizzing element with resin applicator and knife with adjustable gap. The coating unit is working on room temperature. The Curing Unit is consisted of single horizontal drying channel with Heating Zone and temperature range from 60-150°C. The total length of the Curing Oven is 4m. The Cooling Unit is equipped with cooling table that is connected to the chilled water supply. The chilled water temperature range is 12-16°C. Prepreg Rewinding Station unit is equipped with single - shaft rewinder.

The drive system of the machine is consisted of AC servo motors equipped with gearboxes. This special selection of the servo system gives possibility of very fine controlling and regulation of the line speed into the required range of 0-3 m/min.

The whole process for producing this prepreg is given by the images 2-6.

During the experiments, the following parameters were changed: temperature, gap of knife and speed.

Fig.2. Knife – gap for thickness of resin film

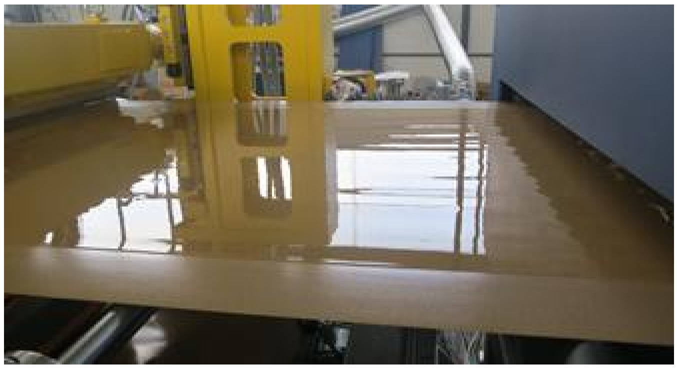

Fig.3. Resin film before oven

Fig.4. The oven - Resin film after oven

Fig.5. Resin film after oven and Kevlar fabric

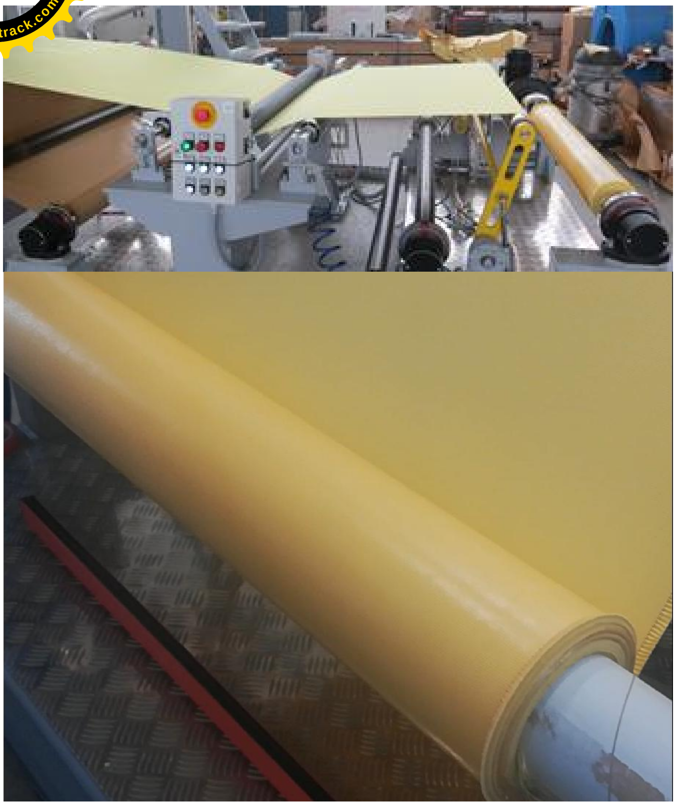

Fig.6. Kevlar/ phenol fabric prepreg coating only one side

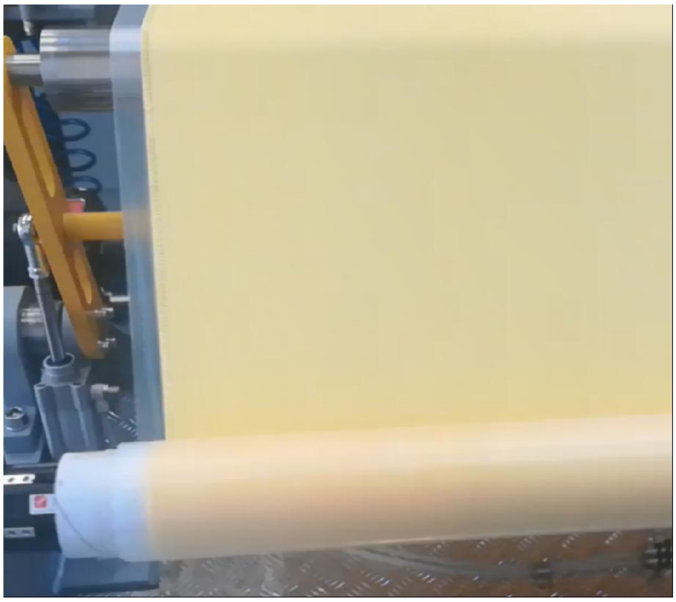

Fig.7. Kevlar/ phenol fabric prepreg coating from both side

The eight rolls with different parameters were produced as given in the table I. Five samples were taken from each roll that were prepared according standard for the determination of the resin content. Preparation of samples, execution and calculation are carried out by standard [14]-[16]. The results are given in a table IV in the next chapter. The purpose of this paper is to prepare prepreg with content resin about 15-20%.

TABLE I
Process parameters for manufacturing of aramid fabric prepreg

No	Constants		Process control			final
	Viscosity of resin mPas	Kevlar fabric gr/m^2	temperature of oven ^{0}C	gap mm	speed m/min	Resin in prepreg W t%
1	800	410	100	0.6	3	11.77
2	800	410	75	0.6	3	13.66
3	800	410	100	0.4	3	6.15
4	800	410	75	0.4	3	6.85
5	800	410	100	0.6	2	17.18
6	800	410	75	0.6	2	19.41
7	800	410	100	0.4	2	5.68
8	800	410	75	0.4	2	7.82

C. *Thermogravimetric analysis of resin and fabric prepreg (TGA)*

Thermogravimetric analysis (TGA) is conducted on an instrument referred to as a thermogravimetric analyzer type NETZSCH – TG 209 F1 Libra TM. A thermogravimetric analyzer continuously measures mass while the temperature of a sample is changed over time. Mass, temperature, and time are considered base measurements in thermogravimetric analysis while many additional measures may be derived from these three base measurements.

For to make control of content of resin in final products (fabric prepreg) are used TGA instrument for pure resin before impregnation in order for to calculate % volatile of phenolic resin.

D. *Design of experiment (DoE)*

More and more factors have an influence on effectiveness and efficiency in the industrial processes and systems. To find the optimum in control of the processes there are often a lot of experiments to realize – practical and theoretical ones. Design of Experiments involves designing a set of experiments, in which all relevant factors are varied systematically (Fig. 8). When the results of these experiments are analysed, they help to identify optimal conditions.

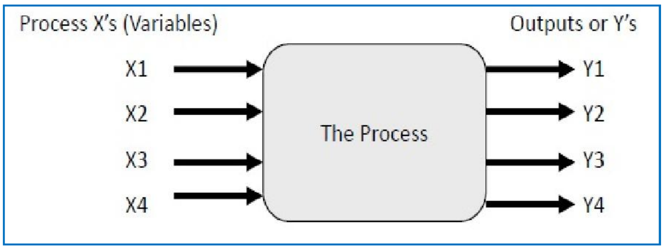

Figure 8. Model of design of experiments

There are lot of factors that influence the process, but only three we have used in the experimental design:

- temperature of oven (factor 1),
- gap – defines film resin (factor 2) and
- speed of process (factor 3).

...ext step was determination of the factor levels as shown in the Table II. Each factor has two factor levels, a low one and a high one. The low one has the value of (-1), the high one has a value of (+1). There are three factors with two levels and eight combinations (N= 8) shown in the Table III. For the statistical analysis five tests of each combination were realized so the number of replications is 8 x 5= 40 samples. With that assumption, we have taken the first-order linear model with interactions to predict the response function, i.e. RAW (gr/m^2)

TABLE III
Level of process parameters.

Symbol	Factor	Level	
		-1	+1
X_1	Temperature of oven ^0C	75	100
X_2	gap -film (mm)	0.4	0.6
X_3	Speed (m/min)	2	3

The full factorial experimental design allows making mathematical modelling of the investigated process in a study domain in the vicinity of a chosen experimental point. In the frame of this paper full factorial experimental design was applied to predict the content of resin in Kevlar fabric prepreg. The important entries that influence on the improved solution were defined.

TABLE IIIII
Factorial design 2^3.

Nº of exp.	Factor		
	X_1	X_2	X_3
1	100	0.6	3
2	75	0.6	3
3	100	0.4	3
4	75	0.4	3
5	100	0.6	2
6	75	0.6	2
7	100	0.4	2
8	75	0.4	2

For the range of the temperature of oven, gap and speed of process the experimental measurements of the content of resin of were carried out by implementing the 2^3 full factorial experimental design.

III. RESULTS AND DISCUSSION

A. TGA Analysis

The results for % volatile of phenolic resin are shown in Fig 9. From diagram shows the % of evaporation of resin is 50-55%, it is important for calculate content of resin in prepreg.

The results for % content of resin in prepreg are shown in Table I and Table IV. The content of resin in prepreg is in range from 5.68 Wt% (24.7gr/m^2) up to 19.41 Wt%. (98.8 gr/m^2).

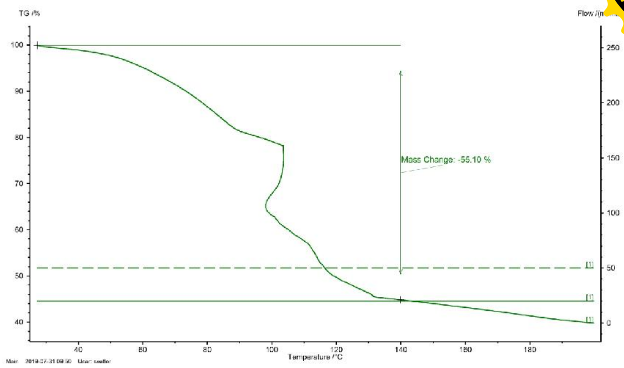

Figure 9. TGA of phenolic resin

B. DOE and Impregnation process

The results for RAW *tests* (content of resin), dispersion and minimal value of parameter's final coefficients for factorial design 2^3 in this research (Impregnation plant process for manufacture fabric prepreg) are shown in table III. Parameter's function and their interaction with 5% mistake are represented with equation 1.

TABLE IVV
Results from design of experiment (DOE)

y exp	y cal	Sy^2	$S_y{}^2{}_{sum}$	$S_y{}^2{}_{sum\ mid}$	S^2b_i	Δb_i
54.74	55.13	3.028	12.85	1.61	0.04	0.39
64.84	64.45	1.303				
26.88	26.49	0.567				
30.14	30.53	0.523				
85.05	84.66	0.81				
98.80	99.19	2.82				
24.70	25.09	0.52				
34.76	34.36	3.28				

$$y=52.624-4.16X_1+23.4X_2-8.374X_3-1.18X_1X_2+1.17X_1X_3-7.866X_2X_3 \quad (1)$$

From design 2^3 were calculated Cochran criteria (Gcal) with value 0.26 and Fisher criteria (f cal) with value 0.77, which fulfill the rule Gcal < Gtab and Fcal < Ftab [23]. According to this, the hypothesis for model 2^3 is acceptable with 5% mistake.

A correlation equations were established for content of resin in fabric prepreg as a function of the temperature of oven, gap of resin and speed of process of the interaction between them. Namely, it was created the regression equitation which the best describes the process. It was observed that if the study domain is precisely established (narrow enough), the factorial experimental design can be employed in order to give good approximation of the response. It was made verification of the model i.e. the adequacy of the regression equation and it was found that the model is adequate and can be accepted and further used.

C. Optical images

Images from coating of the resin over fabric are shown in Fig 10. For these images are use Optical microscope.

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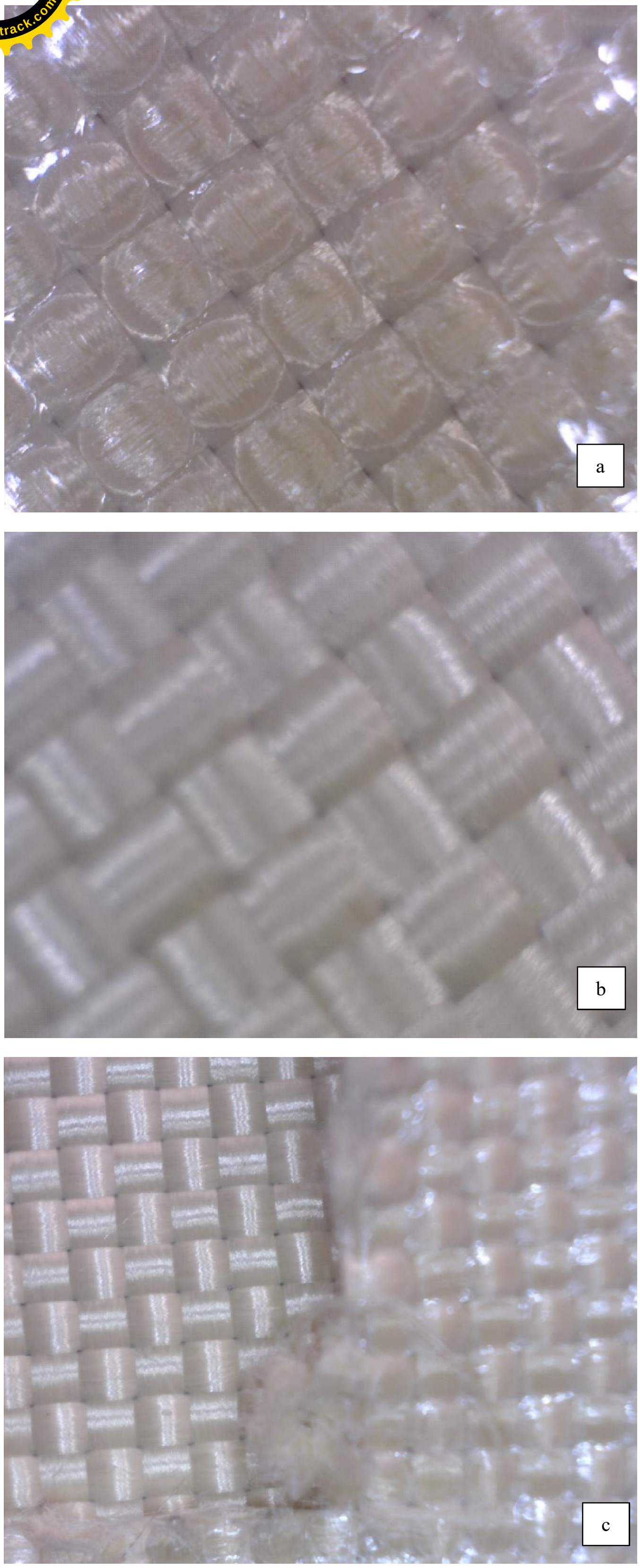

Figure 10. Image from optical microscope of kevlar/ phenolic prepreg with one side coating a) side with resin; b) side without resin and c) comparation of two side with or without resin at Kevlar fabric

IV. Conclusions

The experimental procedure described in the present work is suitable to study the influence of parameters on content of resin in final product - prepreg. The obtained function of the factorial design leads to the conclusion that the examined parameters and the interaction between them must be taken into account in this process for manufacture of prepreg. The parameter X2 (gap -film) is very important in this analysis.

It is assumed that the best prepreg with the required % of resin should use the following parameters in the process:

-temperature in I zone in oven: 75-80°C

- temperature in II zone in oven: 100°C
- speed of process:3 m/min
- gap of knife:0.6

These parameters in the future can serve as the initial way of working with such a coating machine when it is necessary to produce a film of this kind of resin and which should be further transferred to the fabric for manufacture of prepreg.

V. Acknowledgment

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VI. References

[1] J. BAIS. Ed., *Advanced fibre-reinforced polymer (FRP)composites for structural applications: Prepreg processing of advanced fibre-reinforced polymer (FRP) composites*," Industrial Services GmbH, Germany: Woodhead Publishing Series in Civil and Structural Engineering: Number 46, 2013, Part II pp. 125-154. DOI: 10.1533/9780857098641.2.125

[2] F. Wolff-Fabris, H. Lengsfeld, J. Krämer, "*Composite Technology Prepregs and Monolithic Part Fabrication Technologies: Prepregs and Their Precursors* Composite Technology, Publisher: Carl Hanser Verlag GmbH & Co. KG, 2015, pp 11-25. https://doi.org/10.3139/9781569906002

[3] H. Lengsfeld and M.Turner "*Composite Technology Prepregs and Monolithic Part Fabrication Technologies: Prepreg Technology*, Publisher: Carl Hanser Verlag GmbH & Co. KG, 2015, pp 27-45. https://doi.org/10.3139/9781569906002

[4] M. M. Yneux, "Prepreg, tape and fabric technology for advanced composites," Composites. vol 14. No.2. pp.87-91, April 1983.

[5] M. Mehdikhani, L. Gorbatikh, I. Verpoest and S.V Lomov, "Voids in fiber-reinforced polymer composites: A review on their formation, characteristics, and effects on mechanical performance," Journal of Composite Materials, pp1-91, 2018. DOI: 10.1177/0021998318772152

[6] Gruit, "Epoxy prepreg Processing," 23 Nov. 2018, Available: https://www.gurit.com/-/media/Gurit/Datasheets/Prepreg-Processing-Guide.pdf

[7] T. A. Cender, P. Simacek, S. G. Advani, "Resin Film Impregnation in Fabric Prepregs with Dual Length Scale Permeability**,"** J. Comp.: Part A Vol. 53, 2013, pp.118-128. http://dx.doi.org/10.1016/j.compositesa.2013.05.013

[8] C. D. Nguyen, C. Krombholz, "Influence of process parameters and material aging on the adhesion of prepreg in AFP processes" in ECCM17, 2016 paper pp1-5.

[9] Y. Pekbey, K. Aslantaş and N. Yumak, "Ballistic impact response of Kevlar Composites with filled epoxy matrix", Steel and Composite Structures, Vol. 24, No. 2, pp.191-200, December 2017. DOI: https://doi.org/10.12989/scs.2017.24.2.191
[10] K. K. Chawla, *Composite Materials,* Materials Research and Engineering Third edition Springer, 2013, pp 541.
[11] A. B. Strong, *Fundamentals of Composites Manufacturing*, Society of Manufacturing Engineers, 2008, pp620.
[12] HexPly Prepreg Technology. Publication No. FGU 017. Hexcel Registered Trademark, Hexcel Corporation, 2013. http://www.hexcel.com
[13] Mark E. Tuttle, *Structural Analysis of Polymeric composites materials*, second editition, CRC Press, Taylor Francis Group, 2013, pp650.
[14] *Standard Test Method for Volatiles Content of Composite Material Prepreg,* An American National Std. ASTM D3530, 2003.
[15] *Standard Test Method for Matrix Solids Content and Matrix Content of Composite Prepreg,* An American National Std. ASTM D 3529, 2003.
[16] *Standard Test Methods for Constituent Content of Composite Materials,* An American National Std. ASTM D3171, 2003.
[17] *Standard Test Method for Resin Flow of Carbon Fiber-Epoxy Prepreg,* An American National Std. ASTM D3531, 2003.
[18] *Standard Test Method for Gel Time of Carbon Fiber-Epoxy Prepreg*, An American National Std. ASTM D3532, 2003.
[19] http://www.nptel.ac.in/courses/101106038/mod02lec01.pdf
[20] J. Antony, *Design of Experiments (DOE)*, Elsevier Science & Technology Books October, 2003.
[21] A. Dean, D. Voss, *Design and Analysis of Experiments*, Springer Verlag, 1999.
[22] S. A. Risteska, J. K. Mickovski, "Experimental Modeling of Recrystallization Diagram of cold-deformed steel C 1531", 3rd Conference of Macedonian, Metallurgists Union, 2000;
[23] Ф. С. Новик, Ј. Б. Арсов, *Оптимизацијa процесов технологии металов методами Планиррованиjа експериментов*, Машиностроение, 1980.