Study upon water sorption, solubility and mechanical properties of orthodontic adhesives

A. MESAROS[a*], S. BOBOIA[b], C. ALB[a], M. MESAROS[c], S. SAVA[d], M. BADEA[e]
[a]*Department of Dental Propaedeutics and Esthetics, Iuliu Hatieganu University of Medicine and Pharmacy, Cluj-Napoca, Romania*
[b]*Department of polymeric composites, Babes-Bolyai University, Cluj-Napoca, Romania*
[c]*Department of Pediatric Dentistry, Iuliu Hatieganu University of Medicine and Pharmacy, Cluj-Napoca, Romania*
[d]*Department of Dental Materials and Ergonomics, Iuliu Hatieganu University of Medicine and Pharmacy, Cluj-Napoca, Romania*
[e]*Department of Preventive Dentistry, Iuliu Hatieganu University of Medicine and Pharmacy, Cluj-Napoca, Romania*

Orthodontic adhesives are used to glue attachments directly to the surface of the tooth. The study investigates the performance of four orthodontic adhesives. Sample specimens of each adhesive were obtained and submitted to water absorption, water solubility and compression strength testing using standard protocols. Water absorption values for all materials studied were in the range of other literature studies. Statistical results show between all four of the orthodontic adhesives used there are differences between both in terms of absorption and solubility. Mechanical strength tests showed that the adhesives tested are rigid materials with compressive strength comparable dental tissues.

(Received May 03, 2014; accepted May 15, 2014)

Keywords: Orthodontic adhesives, Water absorption, Solubility.

1. Introduction

Orthodontic adhesives are used to glue attachments directly to the surface of the tooth. There are two types of adhesive material applied in orthodontic attachment bonding: resin base and resin hybrid glass ionomer base adhesives, but there is a large variety of commercial products with different physical properties.

The mechanical properties of orthodontic adhesives are critical to their long-term performance in the mouth. Traditionally, the mechanical properties of dental materials have been investigated using static tests. However these methods are not always well suited for measuring the complete material deformation and stiffness properties. This is particularly true for materials under load [1]. Since dental materials are subjected to dynamic loading rather than static loading in the mouth, dynamic tests have become increasingly relevant [2] and are often preferred [3] as they better mimic the cyclic masticatory loading to which these materials are subjected clinically.

Water present in the mouth is a major interfering factor when bonding adhesives and/or composites to the tooth [4]. Under *in vivo* conditions, there is little measurable control over the amount of water left on the tooth during bonding. In addition to being subject to masticatory stresses, dental materials absorb water in the oral cavity, compromising their physical and mechanical properties, accelerating the degradation of the material, softening of dental resins through plasticization, and facilitating the release of unreacted monomers and degradation products [2, 4, 6, 7]. The study of water sorption and solubility of dental materials is important to understand their long-term performance, since water may promote a variety of chemical and physical processes that create biological concerns as well as producing deleterious effects on the structure and function of the polymer matrix itself.

The selection of the materials used as orthodontic adhesives has a considerable influence on the reactivity, degree of conversion, mechanical properties and water uptake.Alternative research techniques are essential to prove the effectiveness of these materials and their viability, especially if we study experimental materials with commercial materials already studied. It is important to determine the effects of these techniques because they can demonstrate the viability and credibility of the research on its properties.

The mechanical properties of dental materials depend on a number of factors that make up the features of the two phases of composite material (the organic and inorganic), namely: the mechanical strength of the filler material, the state of the dispersed phase material, dispersed phase geometry, the orientation of the dispersed phase, the composition of the dispersed phase, the ratio of the two phases, and the links between them.

The studies presented here investigate the performance of four orthodontic adhesives, with emphasis on the water sorption, solubility and dynamic mechanical properties.

2. Experimental

The adhesives used in the present study were:

- the resin base adhesive Light Bond (Reliance Orthodontic Products, Itasca, Il)
- the resin base adhesive Opal Bond MV(Opal Orthodontics, Ultradent)
- the resin hybrid glass ionomer adhesive Fuji Ortho LC (GC)
- an experimental resin hybrid glass ionomer material developed at the Raluca Ripan Institute of Chemistry (UBB)

The experimental resin hybrid glass-ionomer is a new chemical formula developed by the researchers from the Raluca Ripan Institute of Chemistry, for which the present testing was the first performance test.

For each test we used 10 sample specimens from each material.

Determination of water absorption the absorption is expressed as the weight gain of cylindrical specimens, after 7, 14 and 21 days of maintenance in distilled water at 37 ° C. In order to assess the absorption of water, samples were prepared from the all commercial and experimental materials used according to the method described below.

To determine the water absorption was used a Teflon mold that produced the test specimens with the following dimensions: d = 15 ± 1mm diameter and thickness h = 1 mm in accordance with the ISO 4049/2000. Each specimen was kept in the mold with a transparent foil irradiated with visible light portions, ensuring that each portion of the test specimen is exposed for 9×20 seconds. Translux Energy ® / Heraeus - Kulzer halogen lamp was used, which has a light intensity of 900 mv/cm^2. After exposing the entire surface of the sample, which was extracted from the mold surfaces must be smooth and made flat using sandpaper.

The discs removed from the mold were dried in the desiccator in the presence of calcium chloride at 37°C for 24 hours. Before being weight, the specimens were kept in the desiccator at 23 ° C to obtain constant weight. Weighed discs were immersed in distilled water, at 37 ± 1 ° C where were maintained for 7, 14 and 21 days, during which weight was determined daily. After the right amount of time are removed from the water with tweezers, wiped with cellulose paper and air-dried for 15 seconds. After 1 minute from the removal of water the samples were weighed. The extent of absorption in water for each disc was calculated as follows:

$$W_{sp} = (m_2 – m_3)/V$$

where: m2 - mass of the sample after immersion in water for 24 hours (μg)
m3 - mass of the sample kept in desiccator until constant mass (in μg)
V - volume of the sample (in mm3)

Solubility in water

Solubility is the weight loss of specimens of materials used in dentistry, by dissolving the material in water. The testing was done by immersing the test pieces (size of test pieces is the same as the absorption of water) in distilled water at 37 ° C for 7 , 14, 21 days. Solubilization is carried out by: diffusion of the solvent into the polymer matrix; dissolve itself; macromolecules dispersion in the solvent. A high solubility can be a great deficiency for cementing materials. Insolubility of all components of a composite resin is a prerequisite for clinical success. Inorganic fillers are virtually insoluble, but the softening and dissolution of the resin surface, they remain exposed and are easily removed by external agents. Solubility increases the separation between the bracket and enamel favoring and color changes due to fluid entering the separation zone between enamel and bracket. Cementing materials containing a higher proportion of low molecular weight components (TEGDMA, HEMA, etc.) have a higher solubility. The protocol map to evaluate the solubility in distilled water, artificial saliva and the cementing material used in the orthodontic is similar to that described above, the determination of water absorption . The difference is the formula by which the experimental values obtained , expressed in μg/mm3 were calculated using the formula:

$$SL = (m1 - m3) / V$$

where: m1 - mass constant sample before immersion in water (in mg)
m3 - mass of the sample kept in desiccator until constant mass (in mg)
V - volume of the sample (in mm3). Solubility values were recorded at 1,2,3,4,5,6,7,14 and 21 days

Compressive strength

Among mechanical stress from the oral cavity the predominant one is the compression force during which there are developed the greatest forces to which are subjected hard tooth tissues. During masticatory act, muscle contraction develops considerable force; 70 kg in the molars and 45kg in premolars to 25kg in anterior teeth. In addition, however, appear short dynamic loads acting significant increase in local compression of the material. Factors which influence the resistance to compression are:

- chemical nature of the inorganic phase
- temperature (compressive strength decreases with temperature increase)
- interfacial adhesion of the composite resin / filler

Dental materials which have a high content of fillers and poor interfacial adhesion crack at the interface resin / filler due to compression. The resistance to compression is a relative indicator of the resistance to abrasion. To the extent that is abrasion and removal of the layer of filler material, high levels of resistance to compression provides good resistance to abrasion. Abrasion resistance assesses the loss of substance with time due to wear during mastication and act under the action of tooth surfaces due to friction with food, toothpaste, brushing, etc.

Preparation of samples

We prepared specimens from orthodontic adhesive material for said mechanical tests according to international standards, as follows: we inserted material

with a plastic spatula in a teflon mold standard of varying sizes depending on the type of determination made. Matrix sample was coated on both sides by a glass plate with thickness of 1 mm, and then polymerized 40 sec using Translux Energy ® / Heraeus - Kulzer halogen lamp. The specimen was removed from the mold and light cured identically on the opposite side is then finished with 140 grit sandpaper. After 3 minutes of curing, the whole assembly was fixed with a small vise and sat in a water bath at 37 ± 1 ^{o}C. After 15 minutes the assembly was removed from the water, was opened, the specimen was removed to deburring where appropriate, and the specimen thus obtained was immersed in distilled water at 37 ± 1^{o}C where it was maintained for 24 hours. After 24 hours, the specimen was placed in the second bath at a temperature of 23 ° C distilled water. After 50 hours, the specimen was removed, dried and sectional dimensions were measured with a micrometer. Then, it was introduced into the measuring compressive strength. Measurements were performed on a universal testing machine (Lloyd LR5K Plus). The records were viewed using Nexygen PC software . The device is provided with a transmission electronic system and can measure the strength and elongation as well as can adjust mechanically the speed of movement of the clamps. Measurements were carried out in the range of 0-400 kgf measuring the force at 23^{o}C. Measured diameter d of each specimen and the force F recorded when the specimen breakage occured. Compressive strength (in MPa) was calculated using the formula:

$$RC = 9.81 \times F / 0.785 \times d2$$

Compressive strength value was given by the average of at least 5 determinations of 10 . Specimens that were turned over 15% of the mean value were not taken into account. If more than 2 samples deviated by 15% from the average value the measurments for the entire series was repeated.

The results were statistically analyzed. We calculated the average values and standard deviations for water absorption and solubility of each composite for the 7, 14, and 21 days.

Data were statistically analyzed using ANOVA for analysis of the four groups materials and T-test for comparing two groups of materials. To assess the correlation between the water absorption and dissolution rate Pearson correlation test was used. Data obtained for the mechanical tests were processed statistically using ANOVA and Scheffe posthoc tests to compare mean values of the four orthodontic adhesive materials. The threshold of significance (p) was 0.05. For statistical analysis we used PASW Statistics 18.0 software.

3. Results

Determination of water absorption and solubility

The results of water absorption and solubility of materials studied are given in Figs. 1 and 2. In accordance with ISO 9000, composite resin materialsin order to be suitable for use as dental materials should have more water absorption values μg/mm3 than 50 and less than 5 μg/mm3 solubility [10]. Water absorption values for all materials studied are in the range of ISO. In tables 1 and 2 are the average values, standard deviation and statistical parameters for absorption and solubility in water (μg/mm3) after 21 days. Statistical results show that between all four of the orthodontic adhesive materials used there are statistically significant differences ($p < 0.002$) in the 21 days between the average values both in terms of absorption and solubility.

Table 1. Mean, standard deviation and statistical parameters for the absorption of water (μg/mm3) after 21 days.

	Adhesive	Average (μg/mm^3)	SD	SEM	p (t- test)				ANOVA	
					vs. Opal Bond MV	vs. Light Bond	vs. Fuji Ortho LC	vs. Experimental	p	F
Water absorbtion	Opal Bond MV	15.711	0.5420	0.2710	1.0000	0.0001	0.0042	0.0005	0.0000	77.7
	Light Bond	11.323	0.4622	0.2311	0.0001	1.0000	0.0029	0.0004		
	Fuji OrthoLC	42.604	7.3000	3.6500	0.0042	0.0029	1.0000	0.0153		
	Experimental	1.5097	0.3268	0.1887	0.0005	0.0004	0.0153	1.0000		

Table 2. Mean, standard deviation and statistical parameters for water solubility (µg/mm3) after 21 days.

	Adhesive	Average (µg/mm^3)	SD	SEM	p (t –test)				ANOVA	
					vs. Opal Bond Mv	vs. Light Bond	vs. Fuji Ortho LC	vs. Experimental	p	F
Water solubility	Opal Bond MV	-21.798	3.2524	1.6262	1.0000	0.7996	0.0203	0.1628	0.000	41.91
	Light Bond	-15.287	1.4979	0.8648	0.7996	1.0000	0.0005	0.3628		
	Fuji OrthoLC	-135.126	25.144	14.517	0.0203	0.0005	1.0000	0.0115		
	Experimental	-5.473	1.3075	0.7549	0.1628	0.3628	0.0115	1.0000		

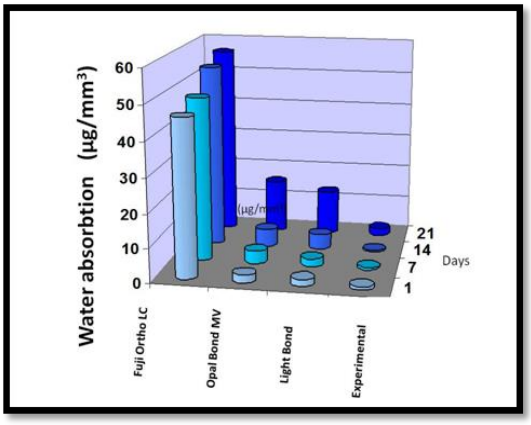

Fig. 1. The graphical representation of the values of water absorption of samples immersed in water.

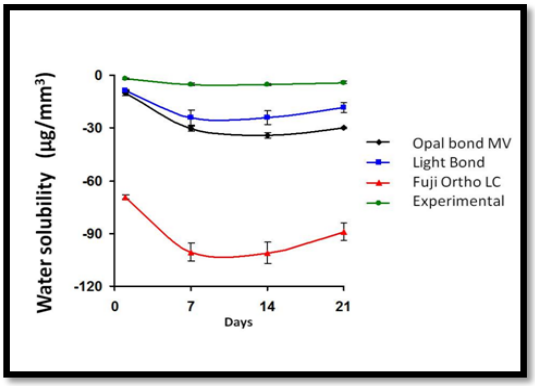

Fig. 2. Graphical representation of solubility values for samples immersed in water.

ANOVA results revealed that between all four adhesive materials used are statistically significant differences ($p < 0.002$) between the mean values for the three test periods, both in terms of absorption and solubility. Posthoc Scheffe test shows that the differences between the periods between mean values are statistically significant.

Regarding the water uptake, the differences are statistically significant for composite cements OPAL BOND MV, LIGHT BOND and Fuji Ortho LC glass ionomer between 7 and 14 days, but for the range of 7-21 days and 14-21 days the differences are not statistically significant regarding the Fuji Ortho LC ionomer. Between 7 to 21 days the differences are statistically significant for the adhesives Experimental and OPAL BOND MV. Also LIGHT BOND and OPAL BOND MVcomposite cement have statistically significant differences in the range 14 to 21 days.

In terms of solubility in water, significant differences appear between the average for the period 7-14 days. For OPAL BOND MV composite adhesive in the range of 7-21 days and 14-21 days we observed statistical signifficant differences in comparison with Light Bond.

Table 3. Comparative statistical analysis for differences between periods during water absorption.

Days	Opal Bond MV			Light Bond			Fuji Ortho LC			Experimental		
	Average	SD	p	Average	SD	p	Average	SD	p	Average	SD	p
7	3.82	0.54	**0.0154**	2.40	0.28	**0.0006**	48.26	2.82	**0.0021**	0.71	0.28	0.6376
14	5.23	1.07		4.53	0.46		54.21	2.86		0.57	0.46	
7	3.82	0.54	**0.0049**	2.41	0.28	**0.0000**	48.26	2.82	0.1935	0.71	0.28	**0.0105**
21	15.9	3.42		13.30	0.32		56.47	8.41		2.265	0.46	
14	5.23	1.07	**0.0051**	4.52	0.46	**0.0001**	56.47	8.41	0.6993	0.56	0.46	**0.0000**
21	15.9	3.42		13.3	0.32		54.21	2.86		2.26	0.46	

Table 4. The comparative statistical analysis of the differences between the periods during water solubility.

Days	Opal Bond MV			Light Bond			Fuji Ortho LC			Experimental		
	Average	SD	p	Average	SD	p	Average	SD	p	Average	SD	p
7	-30.29	2.79	**0.0167**	-23.92	8.61	0.6042	-100.6	9.98	0.7608	-4.90	1.72	0.4226
14	-34.39	2.86		-24.20	7.69		-101.0	12.1		-5.28	1.30	
7	-30.29	2.79	0.7276	-23.92	8.61	**0.0334**	-100.6	9.98	0.0668	-4.15	1.30	0.1835
21	-29.72	0.98		-18.25	6.01		-89.03	10.12		-4.90	1.72	
14	-34.39	2.86	0.0743	-24.20	7.69	**0.0146**	-89.03	10.12	0.1054	-5.28	1.30	**0.0000**
21	-29.72	0.98		-18.25	6.01		-101.1	12.12		-4.15	1.30	

The evaluation of the correlation between solubility and absorption rate was carried out by Pearson test. Pearson index values 0.9711 0.9177 respectively, shows a statistically insignificant correlation between the values of absorption and solubility

Compressive strength

Assuming that the compressive strength estimates the resistance of the adhesion material or enamel to compression results obtained regarding the adhesive materials studied by us confirms that the connection between them is strong. The alues of compressive strength of the two commercial resin-based materials OPAL BOND MVand LIGHT BOND are compared with corresponding values for the resin-modified glass-ionomer materials Experimental and Fuji Ortho LC. The mean and standard deviation of four tests carried out are shown in Table 5.

Table 5. Average and standard deviation of compressive strength (CS).

Adhesive	Average (MPa)	SD	N	SEM	p (t test)				ANOVA	
					vs. Opal Bond MV	vs. Light Bond	vs. Fuji Ortho LC	vs. Experimental	p	F
Opal Bond MV	106.760	3.404	5	1.522	1.0000	**0.0000**	0.1133	**0.0028**	**0.0000**	55.184
Light Bond	185.901	6.073	5	2.716	**0.0000**	1.0000	**0.0024**	0.1410		
Fuji Ortho LC	120.981	16.85	5	7.538	0.1133	**0.0024**	1.0000	**0.0013**		
Experimental	218.080	26.60	4	13.299	**0.0028**	0.1410	**0.0013**	1.0000		

For compressive strength (MPa), ANOVA results show that between the average values of the four adhesives tested there are statistically significant differences ($p < 0.0001$). Posthoc Scheffe test shows which are the pairs of adhesives between which mean values are statistically different. Mechanical strength tests showed that the adhesives tested are rigid materials with compressive strength comparable to that of enamel and dentin. The differences between the values obtained for compressive strength, between the materials we investigated are due among other things to a larger particle size distribution of hybrid materials, where very small particles of powder are inserted between the larger, reducing the interstitial spaces between them. The very small particles inserted, take effort in reducing the incidence of compression fracture. The results of compressive strength measurements, highlight the fact that a mixture of composite fillings gives good mechanical properties , and this is in agreement with literature studies [9,10], which shows an improvement of mechanical properties using hybrid fillers in general for applications in dentistry. The highest value of compressive strength had the Experimental (218 MPa), and was statistically significant ($p < 0.05$) higher than the commercial comoposit OPAL BOND MV(106 760 MPa) and Fuji Ortho LC resin-modified glass ionomer (120.981 MPa). The value of compressive strength for composite OPAL BOND MV(106 760 MPa) was significantly lower than for the LIGHT BOND (185.901MPa) but did not differ significantly from Experimental.

4. Discussion

Compressive strength is necessary in the mouth because during mastication act dental tissues undergo high-intensity forces (70 kg in the molar and premolar 45 kg to 25 kg in anterior teeth). Literature mentions a variety of values for the mechanical properties of dental restorative composites. Because of different methods for obtaining and testing of dental composites, there are contradictions between the values reported. These values reported in studies usual range of 250-400 Mpa. Recently, by micromanipulation techniques, Curtis et al. (2009) tested the compressive strength of nano-particle aggregates in certain nanocomposite structure, noting that nano-aggregates tend to present multiple fractures compared with conventional inorganic fillers, which could alter the overall mechanical strength of the nanocomposite material [10].

Recently, fracture mechanics concepts have been applied in the study of dental nanocomposites to characterize the behavior of these materials. Chan et al. (2007) shows that the main mechanisms of fracture in nanocomposites are crack deflection by the nanoparticle, that the evolution of the fracture along the interface particle / organic matrix The factors influencing the hardness of a composite material are the nature of the proportion of filler and interfacial adhesion of the composite / inorganic filler, the curing conversion or surface processing [11,12]. Microhardness of resin materialsis significantly less than that of amalgam or dental enamel [9,10].

Published data regarding the absorption and solubility of composite cements are difficult to correlate while using different test periods, the use of different units, different sizes of specimens tested [7]. Another drawback is the assumption that the specimen is increasing in weight due to water , when in fact this increase is the difference between the weight gain due to water and the weight lost by the dissolution of organic compounds of low molecular weight ; Thus, the water absorption values would in fact be larger than those reported [14]. In this study, the water absorption was different for the four materials tested, at all three time periods. The highest values were presented by Fuji Ortho LC followed by OPAL BOND MV, LIGHT BOND and Experimental. Composite solubility is influenced by: the residual monomers , the type , proportion and the average particle size of the inorganic filler, the surface area thereof, silane coupling agents , the presence of air pockets within the composite [12,15] . Solubility values for four to seven days materials were in the range of -0.47 to 1.50 μg/mm3 , and at 14 and 28 days all values became negative , increasing specimen weight (m3 > m1) regardless of immersion. A possible explanation for this increase in weight could be water into the chemical reactions of the adhesive [16]. The values obtained for all materials show a negative value, increasing the value of immersion from the first day to the last day of measurement of the tests (21 days), without going to a constant value. It is noted that materials exhibiting high absorption, have high solubility values and the average of the same strip and for measuring the same range. Literature studies state that solubility registered in the artificial saliva immersion has lower values in comparison with samples immersed in distilled water solubility.

Glass ionomers are susceptible to erosion in water [17] depending on the clinical success of these materials, moisture protection and dehydration, is weakened by early exposure to moisture while on the other hand, drying causes shrinkage and cracks [18,19].

5. Conclusions

Water sorption and dynamic mechanical properties have been studied for orthodontic adhesives from both resin-based and resin-modified glass-ionomer based material categories and the results of our study revealed differences in some aspects between materials and similarities in the same class of adhesives. Experimental adhesive showed good chemical and mechanical properties, but stills needs to undergo other mechanical and biocompatibility tests.

Acknowledgements

The research is part o the PN-II-PT-PCCA-2011-3-2-1275 Contract nr 165/2012 research grant.

References

[1] R. Whiting, PH Jacobsen. J Den Res. **59**, 55 (1980).
[2] J. M. Yang, H. M. Li, M. C. Yang, C. H. Shih, J Biomed Mater Res Part B-Appl Biomater. **48**, 58 (1999).
[3] K. Saber-Sheikh, R. L. Clarke, M. Braden, Biomaterials. **20**, 817 (1999).
[4] J. L. Ferracane, J. L. Dent. Mater, **22**, 211 (2006).
[5] S. Ito, M. Hashimoto, B. Wadgaonkar, N. Svizero, R. M. Carvalho, C. Yiu, Biomaterials, **26**, 6449 (2005).
[6] I SideridouI, DS Achilias,C Spyroudi, M Karabela Biomaterials. **25**, 367 (2004).
[7] A. D. Wison, J. Dent. Res. **55**, 721 (1976).
[8] A. Yap, C. M. Lee, J Oral Rehabil; **24**, 310 (1997).
[9] J. C. Macorra, G. Praides. Clin Oral Invest; **6**, 198 (1976).
[10] P. A. Calgaro, A. Y. Furuse, G. M. Correr, B. P. Ornaghi, C. C. Gonzaga. Braz Oral Res; **27**, 403 (2013).
[11] M. A. Cattani-Lorente, V. Dupuis, J. Payan, F. Moya, J. M. Meyer. Dent Mater; **15**, 71 (1999).
[12] M. Fukasawa, S. Matsuya, M. Yamane, J. Dent Res. **69**, 1175 (1990).
[13] J. W. McLean, Oper Dent **17**, 184 (1992).
[14] P. J. Knibbs, A. W. G. Walls, J Oral Rehabil. **16**, 467 (1989).
[15] G. C. R. Nascimento, C. A. de Miranda, S. M. M. Machado, G. A. M. Brandao, H. A. de Almeida, C. M. Silva, Korean J. Orthod. **43**, 242 (2013).
[16] H. C. Margolis, E. C. Moreno, J. Dent Res. **69**, 606 (1990).
[17] J. Årtun, S. Bergland, Am. J. Orthod. **85**, 333 (1984).
[18] R. Ozoe, T. Endo, R. Abe, K. Shinkai, Y. Katoh, Quintessence Int. **43**, e60 (2012).
[19] L. Breschi, A. Mazzoni, E. S. Dorigo, M. Ferrari, J Adhes Sci Tech. **23**, 1053 (2003).

*Corresponding author: mesaros.anca@umfcluj.ro