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EXPERIMENTAL STUDY ON MECHANICAL PROPERTIES OF PE/CNT COMPOSITES

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High density polyethylene/carbon nanotube composites have been produced for investigations. Four CNT volume fractions and two injection pressures were considered. According to the Taguchi approach, eight experiments were designed. The effect of the carbon nanotubes weight fraction and injection pressure on hardness and impact strength of nanocomposite samples were investigated. The results showed that the effect of the carbon nanotube weight fraction on hardness and impact strength of nanocomposite samples was much higher than the effect of injection pressure. By adding 1%wt (weight) carbon nanotube into the polymer, the impact strength and hardness of the samples improved by 74% and 47%, respectively.

Keywords: nanocomposite, carbon nanotube, polyethylene, injection molding, mechanical properties

1. Introduction

Carbon nanotube composites hold a great potential as the materials to be used in the future. Extraordinary properties of these fantastic materials lead to an exhaustive research. One of the first investigations about polymer/carbon nanotube composites was carried out by Ajayan *et al.* (1994). In their study, they dispersed randomly multi-walled carbon nanotubes in a liquid epoxy resin by mechanical mixing. After that, a large number of theoretical and experimental researches using carbon nanotubes as reinforcing fibers have been performed to address the exceptional mechanical and electrical properties of nanotube-based composites (Schadler *et al.*, 1998; Jin *et al.*, 1998; Haggenmueller *et al.*, 2000; Azizi *et al.*, 2015a,b; Najipour and Fattahi, 2016; Safaei and Fattahi, 2016; Sahmani and Fattahi, 2016, 2017; Fattahi and Safaei, 2017). In the last few years, nanocomposites have significantly advanced compared to composite materials in convention dimensions due to changes in the composition and structure of the materials in nanoscale and presenting unique and special properties. Improvement of mechanical and other properties of such composites strongly depends on the particle content, particle shape and size, surface characteristics and dispersion degree (Komarneni, 1992; Jordan *et al.*, 2005). Consequently, toughening of these composites could be caused by a number of mechanisms such as crack-tip pinning, crack-surface bridging, debonding/microcracking and crack deflection. According to previous reports, mechanical and thermomechanical properties of composites filled with micron-sized filler particles are inferior to those filled with nanoparticles of the same filler. Carbon nanotubes exhibit exceptional mechanical (modulus= 1 TPa, strength= 10 times that of steel), thermal and electrical properties. Accordingly, it can be suggested that development of these nano-level particles can offer tailorability of desired properties in a material. Increasing economic need for fuel in different areas has increased the demand for using lighter new materials such as polymers. On the other hand, because of lower strength of polymers compared to metals, their reinforcement seems inevitable. Nylon 6 was the first polymer which was used to produce nanocomposite by Toyota Company in 1990; but today thermosetting polymers such as epoxy and polyimide, polypropylene, polyethylene, and polystyrene are being used as matrix materials in composites. Among the nanocomposites, most attention has been paid to polymer based nanocomposites. One reason for the progress of polymer nanocomposites is their unique mechanical, chemical and physical properties. Polymer nanocomposites generally have high strength, low weight, high thermal stability, high electrical conductivity and high chemical resistance. The second reason for the progress of polymer based nanocomposites and increased research in this area is the discovery of carbon nanotubes in 1991. The strength and electrical properties of carbon nanotubes are significantly different from those of graphite nanolayers and other filling materials. Supreme mechanical and physical properties of carbon nanotubes along with their low density have made carbon a perfect candidate for strengthening the composites. The diameter of these nanotubes can range from 1 to 100 nm and their aspect ratio is bigger than 100 or even 1000. Numerous works have been done on polymer based nanocomposites. Among them the most significant works by Navidfar (2014) can be mentioned. He produced polymethyl methacrylate/carbon nanotube composites using the injection method. Effects of CNT weight fraction, injection temperature, and maintenance pressure on the mechanical properties of the samples were investigated. According to his results, the increasing of the concentration of carbon nanotubes in the nanocomposites slightly increased their hardness and the impact strength. Among other works done in this area Shishavan *et al.* (2014) can be mentioned. They mixed acrylonitrile butadiene styrene polymer with 0, 2, and 4%wt nanoclay, and the effects of nanoclay and process conditions on the mechanical properties of nanocomposite were investigated. They found that by adding 4%wt nanoclay, the hardness of the samples was slightly increased.

In the present study, the effect of the addition of multi walled carbon nanotubes and the process conditions on the hardness and impact strength of high density polyethylene-carbon nanotube nanocomposite samples with different carbon nanotube weight fractions is investigated.

2. Experimental research

2.1. Materials and equipment

In this study, high density polyethylene (HDPE) polymer produced in Ilam Petrochemical Company was used as the matrix phase. Also multi-walled carbon nanotubes with 5-10 nm inner diameter, 10-30 nm outer diameter and 90% purity, obtained from US Research Nanomaterials, USA, were used as the reinforcement phase. Following primary manual mixing with certain weight fractions, mixing was completed by fusion in a twin screw extruder (ZSK-25, Coperion Werner & Pfleiderer, Germany) device at Iranian Petrochemical and Polymer Research Center, and the nanocomposites were obtained as granules. Then the samples were prepared using a plastic injection device NBM HXF-128 obtained from Neckou Behine Mashin Company and were subject to mechanical hardness and impact tests. In order to conduct Rockwell hardness tests on the samples, Zwick/Roell device (UK) which is shown in Fig. 1a, was used. To obtain the impact strength of the samples, a Charpy impact tester device with pendulum weight of 2.036 kg and arm length of 39.48 cm, which is shown in Fig. 1b, was employed.

In this study, Taguchi approach (Montgomery, 2013) has been used to evaluate the tests and draw final conclusions. Taguchi's method is one of the most popular methods of statistical analysis. The weight of carbon nanotubes in four levels and the pressure injection in two levels were considered, and according to Taguchi's approach, eight experiments were designed. The parameters and levels are shown in Table 1. Also, Table 2 shows the tests designed in accordance with the Taguchi method. In the experiments designed according to Taguchi's method, there is

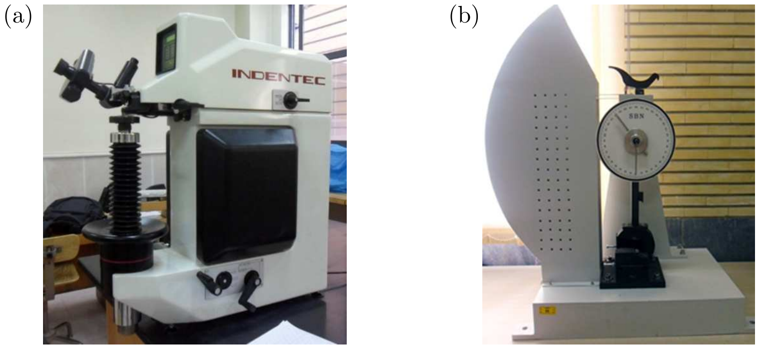

Fig. 1. (a) Hardness test device, (b) impact test device

a loss function which is ultimately introduced as the signal to noise ratio S/N (Montgomery, 2013)

$$S/N = -10 \log \left(\frac{1}{n} \sum_{i=1}^{n} \frac{1}{y_i^2} \right) \tag{2.1}$$

where S/N is the signal to noise ratio, n is the number of observations on the particular product and y is the respective characteristic. According the Taguchi method, the parameter with the biggest S/N has the biggest effect on the experiment.

Table 1. Experimental parameters and their levels

Level	1	2	3	4
Carbon nanotube weight fraction [wt%]	0	0.5	1.0	1.5
Injection pressure [MPa]	60	80	–	–

Table 2. Experiments designed according to Taguchi method

Sample number	CNT weight fraction [wt%]	Injection pressure [MPa]
1	0	60
2	0	80
3	0.5	60
4	0.5	80
5	1.0	60
6	1.0	80
7	1.5	60
8	1.5	80

In order to produce nanocomposite samples, first polyethylene and carbon nanotube with different weight fractions, which are presented in Table 2, were mixed in a two screw extruder device using the fusion mixing method at 170°C, and the nanocomposites were produced as granules. Then the granules were dehydrated in the feeding funnel of the injection device at 60°C for 20 h and the samples were produced based on the designed experiments according to ASTM D638 standard. Once the nanocomposite samples were prepared, hardness and impact mechanical tests were conducted on the samples. Three samples were made, then hardness and impact tests according to Rockwell and Charpy methods were done (see Fig. 2). The hardness test according to the Rockwell M method with three replications in each experiment and the Charpy impact test with three replications in each experiment were conducted for every sample.

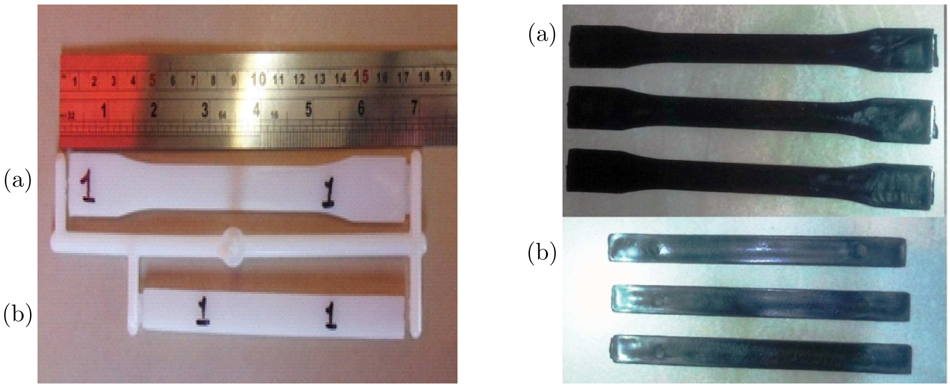

Fig. 2. Left – pure polyethylene samples and right – the produced samples (1%wt CNT): (a) hardness test sample, (b) impact test sample

3. Results

Once the mechanical tests were done, the results obtained for the hardness and impact tests of the samples were obtained and then were analyzed using Minitab software. Taguchi's method was used to evaluate the tests.

3.1. Impact test

In order to prepare the samples for the impact test, a crotch with 45° angle and 2 mm depth was created on the sample and then the sample was subject to the Charpy impact test. Some of the tested samples are shown in Fig. 3. Once the impact test was done, impact strength results were obtained according to Table 3.

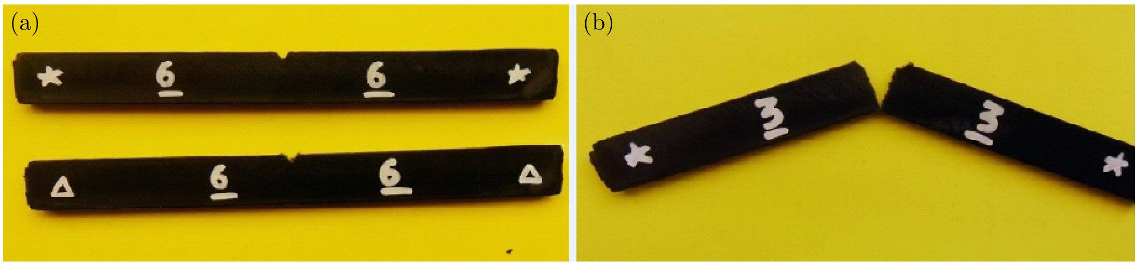

Fig. 3. (a) Selected samples prepared for impact tests, (b) nanocomposite sample after the impact test

Table 3. Impact strength test results

Sample number	Impact strength [kJ/m^2]			Average impact strength [kJ/m^2]
	Test 1	Test 2	Test 3	
1	105.455	111.766	133.128	116.783
2	199.720	172.644	154.208	175.524
3	232.168	178.364	200.776	203.776
4	175.385	170.112	181.075	175.524
5	232.168	302.462	228.168	254.266
6	276.364	221.486	264.950	254.267
7	201.385	238.350	198.866	212.867
8	199.720	252.665	2228.720	225.035

Table 4. Signal to noise ratio S/N for impact strength data

Effect of CNT [wt%]			Effect of pressure		
Level	Impact strength [kJ/m^2]	$S/N = -10\log\left(\frac{1}{2}\sum_{i=1}^{2}\frac{1}{y_i^2}\right)$	Level	Impact strength [kJ/m^2]	$S/N = -10\log\left(\frac{1}{4}\sum_{i=1}^{4}\frac{1}{y_i^2}\right)$
1	$y_1 = 116.783$ $y_2 = 175.524$	43.12	1	$y_1 = 116.783$ $y_2 = 203.776$ $y_3 = 254.266$ $y_4 = 212.867$	45.55
2	$y_1 = 203.776$ $y_2 = 175.385$	45.53			
3	$y_1 = 254.266$ $y_2 = 254.222$	48.11	2	$y_1 = 175.524$ $y_2 = 175.385$ $y_3 = 254.266$ $y_4 = 225.035$	46.23
4	$y_1 = 212.867$ $y_2 = 225.035$	46.80			
Δ	$48.11 - 43.12 = 4.99$			$46.23 - 45.55 = 0.68$	
Rank	1			2	

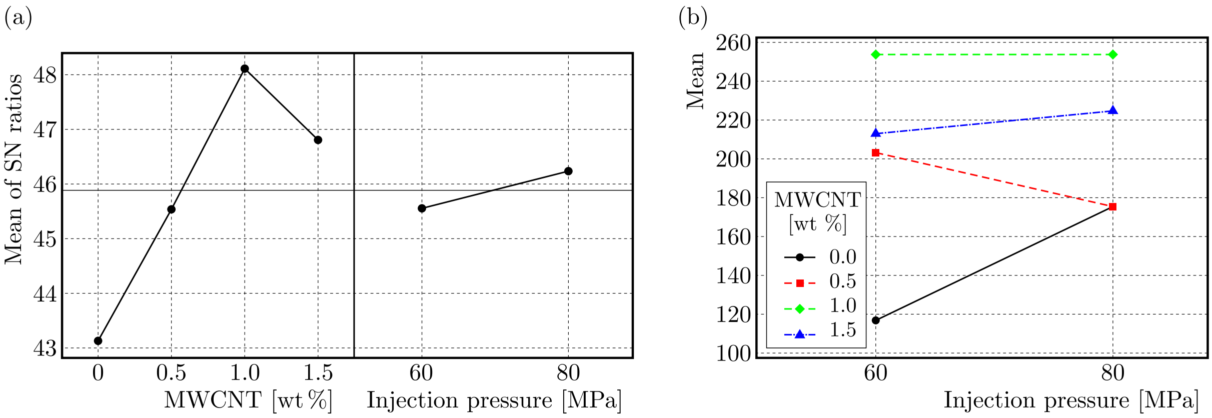

Fig. 4. (a) Signal to noise diagram for the impact strength data, (b) diagram of the counter effects of the parameters on the impact strength

By using these data as the input for Minitab software and analyzing them, the signal to noise ratio data were obtained according to Table 4 and Fig. 4a, where Δ is the difference between the largest and smallest data and Rank shows the impact of the parameter with respect to size of Δ. According to signal to noise results, carbon nanotube weight fraction had much more effect of the impact strength of the nanocomposite samples compared to the injection pressure. By increasing carbon nanotube weight fractions up to level 3 (1%wt carbon nanotubes), the impact strength of the samples were increased almost by 74% (Appendix A), and then it was decreased. This means that the optimum level for carbon nanotube weight fraction was level 3 with 1%wt carbon nanotube in which the highest value for the signal to noise ratio and impact strength was obtained. Increasing the carbon nanotube weight fraction to 1.5%, the signal to noise ratio and impact strength values were decreased, which could be attributed to the agglomeration of carbon nanotubes by increasing their weight fraction. Strong Van der Waals attractions along with their high contact surface (length to diameter ratio of 1000) generally results in special aggregation of carbon nanotubes and, therefore, prevents the transfer of their extraordinary properties to the matrix. The most optimum state for the impact strength among the experiments was when the carbon nanotube weight fraction was at level 3 and injection pressure was at level 2, which corresponded to test sample 6. Also Fig. 4b shows the counter effects of the parameters on the impact test. According to this diagram, when the injection pressure was at its lower level

(60 MPa), the highest impact strength was achieved when 1%wt carbon nanotube was chosen. Also when the injection pressure was at its higher level (80 MPa), the impact strengths of pure samples and samples with 0.5%wt carbon nanotube were similar. Another conclusion which was obtained from the diagram was that when the carbon nanotube weight fraction was at its third level being 1%wt, the impact strengths of the samples produced at 60 and 80 MPa were almost similar. Also according to the results of the signal to noise analyses, the injection pressure had less effect on the impact strength, which, through increasing the injection pressure from 60 to 80 MPa, ingreased the impact strength of samples by only 5%.

3.2. Hardness test

Hardness tests of the samples were conducted according to the Rockwell M method, and the obtained results are shown in Table 5.

Table 5. Hardness test results

Sample number	Hardness (Rockwell)			Average hardness (Rockwell)
	Test 1	Test 2	Test 3	
1	43.0	44.3	42.3	43.20
2	37.2	35.6	42.5	38.43
3	63.3	61.0	61.4	61.90
4	56.0	53.4	54.8	54.73
5	63.7	62.9	62.9	63.17
6	54.2	55.7	59.9	56.60
7	61.3	61.7	63.7	62.23
8	52.5	52.4	49.8	51.57

By analyzing the results obtained from the hardness test using Minitab software, the signal to noise analysis results were obtained and are presented in Table 6 and Fig. 5a.

Table 6. Signal to noise ratio for hardness data

Effect of MWCNT [wt%]			Effect of pressure		
Level	Hardness (Rockwell)	$S/N = -10\log\left(\frac{1}{2}\sum_{i=1}^{2}\frac{1}{y_i^2}\right)$	Level	Hardness (Rockwell)	$S/N = -10\log\left(\frac{1}{4}\sum_{i=1}^{4}\frac{1}{y_i^2}\right)$
1	$y_1 = 43.2$ $y_2 = 38.4$	32.2	1	$y_1 = 43.2$ $y_2 = 61.9$ $y_3 = 63.2$ $y_4 = 62.2$	34.8
2	$y_1 = 61.9$ $y_2 = 54.7$	35.3			
3	$y_1 = 63.2$ $y_2 = 56.6$	35.53	2	$y_1 = 38.4$ $y_2 = 54.7$ $y_3 = 56.6$ $y_4 = 51.6$	33.9
4	$y_1 = 56.6$ $y_2 = 62.2$	35.06			
Δ	$33.53 - 32.2 = 3.33$			$34.8 - 33.9 = 0.9$	
Rank	1			2	

According to the results obtained from the signal to noise analysis for the hardness test, the carbon nanotube weight fraction had a more significant effect on the hardness of the samples compared to the injection pressure. The addition of up to 1%wt carbon nanotubes to polyethylene resulted in a significant increase in the hardness of the samples almost by 47% (Appendix A), and the highest values of the hardness and signal to noise ratio was obtained at level 3 (1%wt

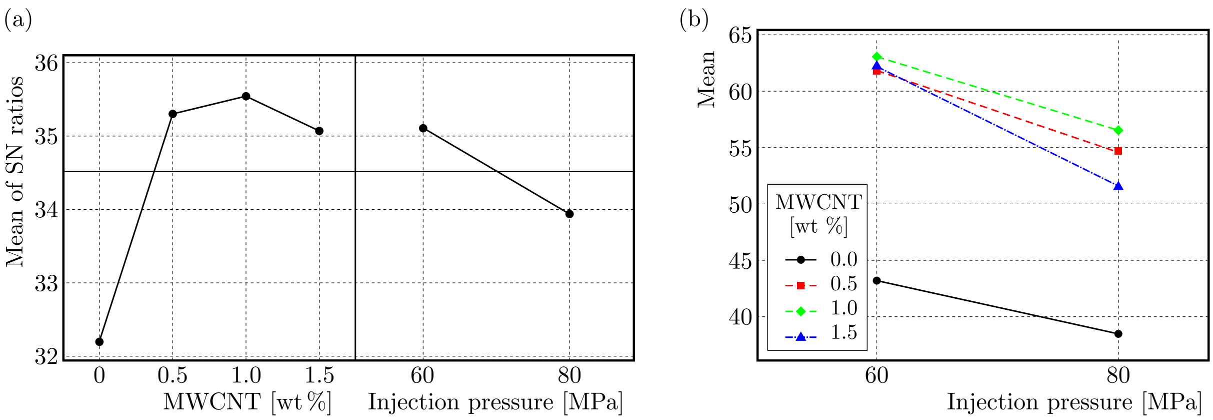

Fig. 5. (a) Signal to noise analysis diagram for hardness data, (b) diagram counter effects of the parameters on the hardness test

carbon nanotubes). A further increase in the carbon nanotube weight fraction up to 1.5% slightly decreased the hardness of the samples which was attributed to agglomeration of the carbon nanotubes as it was mentioned before. On the other hand, wt% by increasing the injection pressure from level 1 to level 2, the hardness and signal to noise ratio of the samples were decreased. The most optimum state regarding the hardness was achieved when the nanotube weight fraction and injection pressure were at levels 3 and 2, respectively, which corresponded to sample 5. Also Fig. 5b shows the counter effects of the parameters on the hardness test. According to this diagram, the hardness of the nanocomposites were significantly higher (almost 47%) than the pure samples and at either 60 or 80 MPa injection pressure. The hardness of the samples containing 1.5%wt showed the highest values. Also another conclusion from this diagram was that at 60 MPa injection pressure the hardness of the samples with 1.5%wt was higher than for the samples containing 0.5%wt while at 80 MPa injection pressure the order was opposite.

4. Conclusion

In this work, high density polyethylene-carbon nanotube nanocomposites were produced according to the injection molding method. The effect of addition of carbon nanotubes at four different levels of 0, 0.5, 1, and 1.5 and of two injection pressure levels of 60 and 80 MPa on hardness and impact strength of the nanocomposite samples was investigated. The results showed that the effect of the carbon nanotube weight fraction on the hardness and impact strength of nanocomposite samples was much higher than the effect of the injection pressure. Addition of 1%wt carbon nanotube into the polymer significantly increased hardness and impact strength of the samples; however addition of 1.5%wt carbon nanotube reduced the values, which was due to agglomeration of carbon nanotubes in the polyethylene matrix.

A. Appendix

From Table 4, the average amount of impact strength at 0% and 1%wt CNT are 146.2 and 254.266 kJ/m^2, then the impact strength increment is

$$\frac{254.266 - 146.2}{146.2} \cdot 100 \approx 74 \tag{A.1}$$

From Table 6, the average amount of hardness at 0% and 1%wt CNT are 40.8 and 59.9 Rockwell, then the hardness increment is

$$\frac{59.9 - 40.8}{40.8} \cdot 100 \approx 47 \tag{A.2}$$

References

1. Ajayan P.M., Stephan O., Colliex C., Trauth D., 1994, Aligned carbon nanotube arrays formed by cutting a polymer resin-nanotube composite, *Science*, **265**, 1212-1214
2. Azizi S., Fattahi A.M., Kahnamouei J.T., 2015a, Evaluating mechanical properties of nano--platelet reinforced composites under mechanical and thermal loads, *Journal of Computational and Theoretical Nanoscience*, **12**, 1, 4179-4185
3. Azizi S., Safaei B., Fattahi A.M., Tekere M., 2015b, Nonlinear vibrational analysis of nano--beams embedded in an elastic medium including surface stress effects, *Advances in Materials Science and Engineering*, **12**, 1, 1-7
4. Douglas C., 2013, *Statistical Quality Control*, Wiley, 7th Edition
5. Fattahi A.M., Safaei B., 2017, Buckling analysis of CNT-reinforced beams with arbitrary boundary conditions, *Microsystem Technologies*, **2017**, 3, 1-13
6. Haggenmueller R., Gommans H.H., Rinzler A.G., Fischer J.E., Winey K.I., 2000, Aligned single wall carbon nanotubes in composites by melt processing methods, *Chemical Physics Letters*, **330**, 219-225
7. Jin L., Bower C., Zhou O., 1998, Alignment of carbon nanotubes in a polymer matrix by mechanical stretching, *Applied Physics Letters*, **73**, 1197-1199
8. Jordan J., Jacob K.I., Tannenbaum R., Sharaf M.A., Jasiuk I., 2005, Experimental trends in polymer nanocomposites-a review, *Materials Science and Engineering A*, **393**, 1-11
9. Komarneni S., 1992, Nanocomposites, *Journal of Materials Chemistry*, **2**, 1219-1230
10. Navidfar A., 2014, Experimental study of mechanical properties of nano-composites containing carbon nanotubes produced by injection molding, M.D. Thesis, Ourmieh University, Ourmieh
11. Najipour A., Fattahi A.M., 2016, Experimental study on polyethylene/carbon nanotube injected composites, *Croizatia*, **93**, 2, 145-152
12. Safaei B., Fattahi A.M., 2016, Vibrational response analysis of polymer/CNT plates using molecular dynamics simulation, *Croizatia*, **93**, 2, 65-79
13. Sahmani S., Fattahi A.M., 2016, Size-dependent nonlinear instability of shear deformable cylindrical nanopanels subjected to axial compression in thermal environments, *Microsystem Technologies*, **2016**, 12, 1-15
14. Sahmani S., Fattahi A.M., 2017. Thermo-electro-mechanical size-dependent postbuckling response of axially loaded piezoelectric shear deformable nanoshells via nonlocal elasticity theory, *Microsystem Technologies*, **2017**, 2, 1-15
15. Schadler L.S., Giannaris S.C., Ajayan P.M., 1998, Load transfer in carbon nanotube epoxy composites, *Applied Physics Letters*, **73**, 3842-3844
16. Shishavan, Mamaghani S., Azdast T., Ahmadi S.R., 2014, Investigation of the effect of nanoclay and processing parameters on the tensile strength and hardness of injection molded Acrylonitrile Butadiene Styrene-organoclay nanocomposites, *Materials and Design*, **58**, 527-534

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