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Preparation and characterization of UV-curable composite containing nano silica for glass-to-glass bonding

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Abstract

Glass is a transparent and geometrically ordered element in architecture. Glass bonding can be classified into adhesive, layered, and mechanical categories. One of the best bonding materials for glass applications is adhesive. The preparation and investigation of Ultraviolet (UV)-curable composites for glass-to-glass bonding is the goal of this research. The investigation focused on the effect of raw material type and weight percentage on mechanical properties. Epoxy acrylate resin was employed as an oligomer in this study. Trimethylolpropane triacrylate (TMPTA), tripropyleneglycol triacrylate (TPGDA), and pentaerythritol tetraacrylate (PETA) were used as monomers. As photoinitiators, were used bis (2,4,6 trimethylbenzoyl)-phenylphosphine oxide, hydroxycyclohexyl phenylketone, and 2,4,6-trimethylbenzoyl diphenyl phosphineoxide. Benzophenone, acrylic acid, nano silica, and trimethoxysilylpropyl methacrylate (TMSPMA) were used as the additives. The mechanical properties of the samples were studied using compressive and shear tests. The degree of conversion (DC) was obtained using Fourier transform infrared spectroscopy (FTIR). The mechanical behavior of the samples were measured as a function of temperature using dynamic mechanical thermal analysis (DMTA); and the distribution of nanoparticles was examined using scanning electron microscopy (SEM). The results showed that the sample containing 4 wt.% nano silica, which also contained 24 wt.% PETA and 7 wt.% TMSPMA, has the best mechanical properties with 0.42 and 0.63 MPa compressive and shear strength, respectively. In the FTIR, was observed an increase of nano silica from the lowest (1 wt%) to the highest amount (5 wt.%), the DC current decreased by 11%. In the DMTA analysis, it was observed that an increase of nano silica from 1-5 wt.%, the storage modulus increased by 41%. The highest loss modulus and loss temperature were related to the samples containing 1 and 2 wt.% nano silica with 221 MPa and 113.8 °C. Finally, the best distribution of nanoparticles was observed in the sample containing 1 wt.% nano silica. In the SEM images, agglomeration of nanoparticles was seen in the sample containing 5 wt.% nano silica.

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Keywords: Glass, Adhesive, Ultraviolet, Epoxy acrylate, Degree of conversion, Nano silica.

1. Introduction

Glass is a transparent element in architecture and construction. Glass has a geometric order and an understanding of interior spaces [1]. Glass is a brittle material without plastic deformation. Glass joints are divided into the mechanical, layered and adhesive connections categories [2]. Ultraviolet (UV)-curable adhesives are among the best adhesive materials for bonding glass to glass. UV-curable technology is an environmentally friendly process. These adhesives have high resistance to aging, moisture and heat and can withstand temperatures from -20 to 80 $^{\circ}$C. In addition, they have low linear shrinkage and small thickness at the joint (less than 0.2 mm) [3-5]. The advantages of these adhesives containing acrylate groups include their low price, environmental friendliness due to the non-emission of volatile compounds, fast curing and absence of solvents [6-9]. The market of UV-curable adhesives will reach $1.2 billion per year in 2021, growing at a CAGR (compound annual growth rate) of 15.9%, with glass bonding adhesive accounting for the largest share of this market [10]. The applications of these adhesives include bonding of automobile headlights, glass containers, the art glass industry, the manufacture of shelves and glass display cases in the furniture industry, crystal sculptures, optical glass lenses and appliance panels [2].

Due to the application mentioned, UV-curable adhesives should have a high chemical and thermal resistance, as they are also used in the electronics and medical industries. Epoxy resins are oligomers with large amounts of polar groups and very reactive epoxy groups, which are often used to produce adhesives with high adhesion for bonding glass, plastic and metal materials [6]. Park et al. [11] prepared a dual UV and thermal curing adhesive by mixing an epoxy acrylate resin with an epoxy resin. After successive UV-curing and thermal processing, the epoxy resin and epoxy acrylate resin formed a cross-linked network structure. Finally, additives such as silanes, titanate couplers, fillers and stabilizers [7, 12, 13].

Silanes create a covalent bond with glass and oligomers through the condensation of silanol; the polymer is hardened and bond to the glass surface [14]. According to other silanes, it acts as a bridge betwwen cured adhesive and glass [10]. The most well-known silanes used are methacryloxy propyldiethoxy methylsilane (MPDEMS), 3-aminopropyl triethoxysilane

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(AMPTES), and vinyl triethoxysilane (VTES) [14]. Vacche et al. [10] used a highly fluorinated UV-curable resin characterized by perfluoropolyether (PFPE) chains as an adhesive for glass bonding. Due to the high fluorine content, adhesion could be achieved through the use of an acrylate silane that can form covalent bonds with both of the glass substrates and the methacrylic adhesive formulation. It has been found that the adhesion of the joints is improved to a large extent even when used in very petty amounts. Zhang et al. [15] showed that modification of nano silica with AMPTES results in a 100% increase in the adhesion of the epoxy group to the glass substrate compared to unmodified nano silica.

The combination of organic materials and mineral fillers as nanocomposites has attracted great attention in recent years [16-19]. The strengthening process of nanomaterials in UV-curable compounds is determined by crack deflection, crack pinning, and rupture [20-22]. In the crack deflection mechanism, the microcracks formed in the area of unreacted monomers are deflected and destroyed by nanoparticles during their growth and movement. The pinning mechanism, stops the growth of microcracks in contact with the nanoparticles and delays failure [20, 23, 24]. Peng [25], modified silica nanoparticles were dispersed with methcryloxypropyl trimethoxysilane in an acrylic copolymer matrix. The presence of nano silica improved the thermal stability of the nanocomposite in terms of degradation temperature and residual weight. Furthermore, the glass transition temperature (Tg) and storage modulus were improved by increasing the weight percentage of nano silica. In UV-curable nanocomposites, nano silica does not act as a UV absorber and does not interface with the curing performance [26]. The presence of nano silica was found to improve the degree of conversion (DC) and increase the Tg and elastic modulus [20]. Kim et al. [27] produced a UV-curable adhesive based on epoxy acrylate. The DC of their best sample was 69%. They also proved that increasing the weight percentage of nano silica reduces DC, because the presence of nanoparticles reduces the rate of polymerization (R_p). Li et al. [28] prepared a UV-curable coating based on epoxy acrylate reinforced with nano silica modified with AMPTES. The DC of their best sample was 70% and also showed that the presence of nano silica lowers the R_p.

According to previous studies, neither silane with an acrylate group nor hydrophobic nano silica on epoxy acrylate were used for UV-curable adhesives for bonding glass to glass. The aim of this study is to prepare UV-curable composites for glass-to-glass bonding. Oligomers,

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monomers, photoinitiators, silane coupling agents, adhesion promoters and silica nanoparticles were used in these composites and the effect of their presence and weight percentage on the mechanical behavior was studied. The prepared composites were tested for mechanical compressive and shear properties, FTIR analysis to obtain DC, DMTA analysis to determine mechanical-thermal properties, and SEM to check the distribution of nanoparticles.

2. Materials and methods

2.1. Raw materials

In this research, an aliphatic epoxy acrylate oligomer (commercial grade of Troxy resin system, China) was used. Trimethylolpropane triacrylate (TMPTA), pentaerythritol tetraacrylate (PETA), and Tripropyleneglycol diacrylate (TPGDA) as monomer (commercial grade of Troxy resin Co., China) were supplied. Also, 1-hydroxycyclohexyl phenylketone (IRGACURE 184), bis(2,4,6-trimethyl benzoyl) phenyl phosphineoxide (IRGACURE 819), and 2,4,6-trimethyl benzoyldiphenyl phosphine oxide (LUCIRIN TPO) (commercial grade of Apuvcoaing Co., China) as photoinitiator, and UV stabilizer benzophenone (laboratory grade of Merck, Germany), were used. Trimethoxysilylpropyl methacrylate (TMSPMA) as a silane coupling agent (laboratory grade of Sigma-Aldrich, USA), acrylic acid (laboratory grade of Merck, Germany) as an adhesion improver and nano silica (AEROSIL R972 Hydrophobic from Aeonik, Germany) were supplied.

2.2. Preparation method

The preparation of the UV-curable composite in this study involved two general steps. In the first step, epoxy acrylate and monomers according to Table 1 were combined at 70 °C for 30 minutes in a 250 ml two-neck flask equipped with nitrogen gas and a mechanical stirrer (Heidolf, Germany) at 200 rpm on a hot plate (MR Hei, Heidolf, Germany). In the second step, the photoinitiator and benzophenone were added and combined at 40 °C for 30 minutes. The product obtained was placed in a black container without UV light and allowed to be cured. According to Table 1, TMSPMA and acrylic acid were added to the sample in the stages where TMSPMA and acrylic acid were present, each after the completion of the second stage at 40 °C for 15 minutes. For the samples containing nano silica, after completing the first step, nano

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silica was added to the sample and probed in an ultrasonic device (Homogenous 4000R, Nasir Research Technology Co., Iran) for 30 minutes. For all samples, the amount was 2 wt.%.

Table 1. The raw material of the studied samples (wt.%)

Sample No.	Epoxy Acrylate	TMPTA	TPGDA	PETA	IRGA CURE 184	IRGA CURE 819	LUCIRIN TPO	TMSPMA	Acrylic Acid	Nano Silica
1	65	0	30	0	0	3	0	0	0	0
2	65	0	0	30	0	3	0	0	0	0
3	65	30	0	0	0	3	0	0	0	0
4	65	15	0	15	0	3	0	0	0	0
5	65	10	0	20	0	3	0	0	0	0
6	63	0	0	27	0	3	0	5	0	0
7	63	0	0	26	0	3	0	6	0	0
8	62	0	0	26	0	3	0	7	0	0
9	62	0	0	25	0	3	0	8	0	0
10	63	0	0	25	3	0	0	7	0	0
11	63	0	0	25	1.5	1.5	0	7	0	0
12	63	0	0	25	0	0	3	7	0	0
13	62	0	0	25	0	4	0	7	0	0
14	63	0	0	26	0	2	0	7	0	0
15	63	0	0	25	0	3	0	7	1	0
16	62	0	0	25	0	3	0	7	0	1
17	62	0	0	24	0	3	0	7	0	2
18	61	0	0	24	0	3	0	7	0	3
19	60	0	0	24	0	3	0	7	0	4
20	59	0	0	24	0	3	0	7	0	5

2.3. Characterization techniques

In this study, five tests such as compressive strength, shear strength, Fourier transform infrared spectroscopy (FTIR), dynamic mechanical thermal analysis (DMTA), and scanning electron microscopy (SEM) were carried out. For the pressure test, 10 x 10 cm glass with a thickness of 1 cm was prepared as shown in Fig. 1. Before pressure is applied, the edge of the glass is polished until the contact surface reaches the highest level. Then, 100 µl of the adhesive sample was poured onto the glass surface and another glass was placed on it. Finally, the glass was irradiated with UV light for 15 seconds at a distance of 5 cm from the glass. The sample was subjected to a compression test so that the machine applied a force at a speed of 0.5 mm/min

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at a distance of 5 cm from the joint (at the center of mass). This test was carried out using a universal device (model STM20, Santam, Iran).

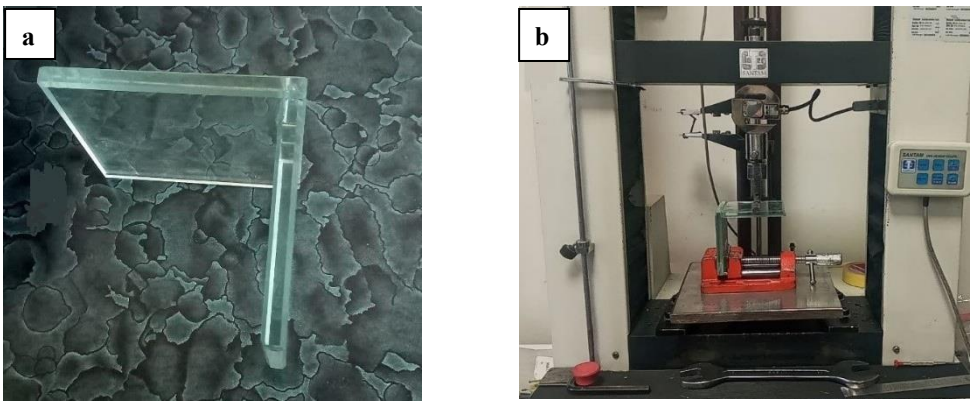

Fig. 1. Preparation of samples, a) connection of two glasses after applying UV light, b) subjecting the sample to the pressure test.

For the shear test [29] according to the ASTM D1002 standard, 20 cm x 2.54 cm glasses with a thickness of 6 mm were prepared. Then 100 µl of the adhesive sample was poured onto the glass surface and another glass surface was placed on it. Finally, the glass was irradiated with UV light at a distance of 5 cm for 15 seconds (Fig. 2). This shear strength test was carried out using the same compressive strength testing machine.

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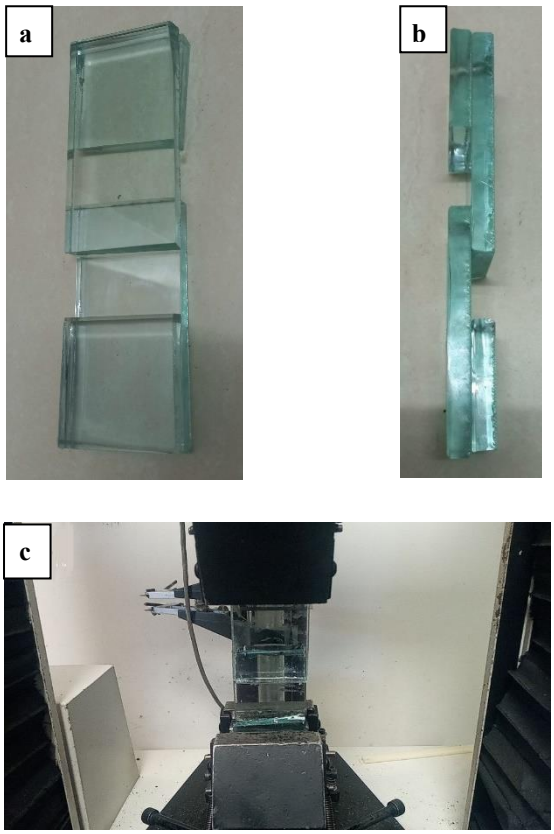

Fig. 2. Preparation of samples for the shear test, a) side view of the bonding of two glasses, b) front view of the connection of two glasses, c) placement of the sample under the machine.

One of the most important properties of UV-curable materials is their DC parameter, which was determined from the FTIR test. This evaluation was carried out by comparing the ratio of vibrational bands of unpolymerized aliphatic (C=C) acrylates at 1635 cm^{-1} with the aromatic (C=C) bands at 1610 cm^{-1} according to equation 1 [30]. First, the samples were cured with a UV lamp for 30 seconds and then prepared for testing. To measure DC, at the first, five samples numbered 16 to 20 of uncured glue were placed on the diamond of the device, and using the special pit-like row, the IR spectrum completely covered the uncured glue, which was done for each five samples. The absorption bands were recorded in the transmission mode of the FTIR instrument (model 1720-X, Perkin Elemer, USA). Subsequently, the cured samples of the same five samples were placed in the FTIR device and absorption bands were obtained.

$$DC=\frac{\left(\frac{\text{absorption in 1610}}{\text{absorption in 1635}}\right)\text{cured}}{\left(\frac{\text{absorption in 1610}}{\text{absorption abs in 1635}}\right)\text{uncured}} \quad (1)$$

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The study of the mechanical deformation of the network at low strain was measured as a function of temperature using DMTA (model DMA 242C, Netzsch, Germany) in tensile mode. In the DMTA test, cured samples measuring 5 mm x 25 mm and 0.5 mm thick were prepared. The temperature range used in this analysis was -20 to 150 °C. The morphologies of the cured nanocomposite films were investigated by SEM (model VEGA3, TESCAN, Czech). For SEM analysis, the cured samples 16-20 should be covered with a thin layer of gold.

3. Results and discussion

The results of this study come from the preparation of UV-curable composites for glass to glass bonding. In this study, mechanical property tests including compressive and shear strength tests, DC determination from FTIR analysis, DMTA results and SEM analysis were performed on the cured samples to check the distribution and agglomeration of nanoparticles.

3.1. Results of mechanical properties

3.1.1. Effect of monomers

In the mechanical test, according to Table 1, in samples 1-5, the effects of different types and weight percentages of monomers were investigated under the same conditions. The compressive and shear strength results are shown in Figs. 3-a and 3-b, respectively. Sample 2 showed the best results in term of both strength and failure strain. That is, the sample containing 30 wt.% PETA has a compressive strength of 0.3 MPa, and its failure strain is 113%. The shear strength was 0.45 MPa, and the failure strain was 120%. The sample containing 30 wt.% PETA (sample 2) showed an improvement in compressive and shear strength of 150% and 120%, respectively, compared to the weakest sample (sample containing 30 wt.% TMPTA). It can be argued that PETA brings higher strength than TPGDA and TMPTA due to its higher functionality (more cross-linking compounds) [31].

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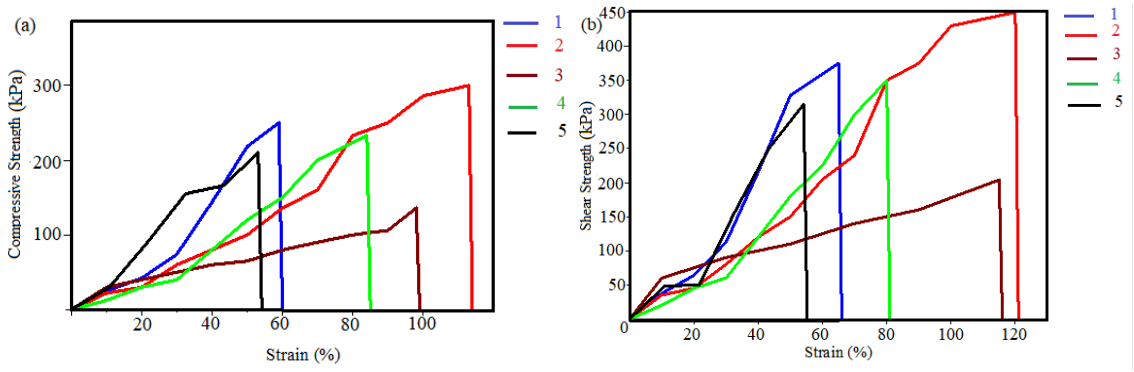

Fig. 3. Comparison of the mechanical properties of samples of UV-curable composites containing TMPTA, TPGDA and PETA (samples 1-5) in, a) compressive strength, b) shear strength.

3.1.2. Effect of TMSPMA

In the mechanical test, the effects of TMSPMA were investigated in samples 6-9. The compressive and shear strength results are shown in Figs. 4-a and 4-b, respectively. Sample 8 showed the best results in term of strength and failure strains. In other words, the sample with 7 wt.% TMSPMA showed a compressive strength of about 0.34 MPa and a failure strain of 100%. In addition, its shear strength was 0.51 MPa and its failure strain was 110%. The sample containing 7 wt.% TMSPMA (sample 8) had a 13% improvement in compressive and shear strength compared to the sample without silane content (sample 2).

In the strengthening mechanism of TMSPMA, alkoxysilanes release methanol when attached to the surface. In coatings where one surface is affected and the other surface is exposed, this methanol is vapored. However, in this application, where the adhesive is between two surfaces, it is not easy to vapor methanol. The methanol released has a destructive effect on the strength. Therefore, the presence of TMSPMA above a value is harmful to the system [32].

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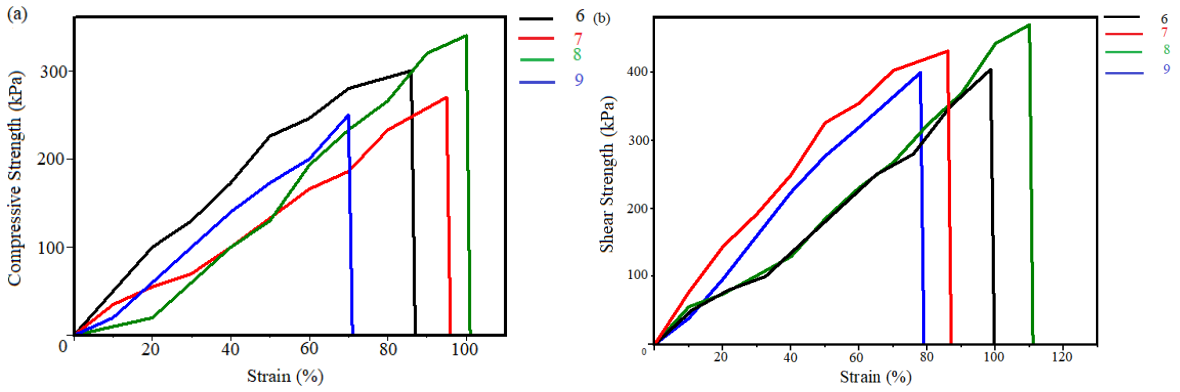

Fig. 4. Comparison of the mechanical properties of samples of UV-curable composites containing TMSPMA (samples 6-9) in, a) compressive strength, b) shear strength.

3.1.3. Effect of photoinitiators

In the mechanical test, the effects of various photoinitiators (IRGACURE 184 and 819 and LUCIRIN TPO) were investigated in samples 10-14. The compressive and shear strength tests are shown in Figs. 5-a and 5-b, respectively. As can be seen, none of the samples from 10 to 14 were better than sample 8 (sample 8 has the best mechanical properties among samples 1-8). That is, replacing IRGACURE 819 (sample 8) with IRGACURE 184 (sample 10) has the opposite effect on strength and reducing it (26% and 31% reduction in compressive and shear strength, respectively). Furthermore, replacing IRGACURE 819 (sample 8) with LUCIRIN TPO (sample 12) did not change strength and failure strain results in both shear and compression modes, and LUCIRIN TPO strongly discolors the sample. Finally, increasing IRGACURE 819 from 3 to 4 wt.% (sample 13) reduced both of compressive and shear strength by 35%, and even its decrease from 3 to 2 wt.% (sample 14). Also, reduced the strength 41 and 29% in compressive and shear strength, respectively. It can be concluded that 3 wt.% IRGACURE 819 (sample 8) has the best strength and strain while maintaining transparency and appearance properties. It is attainable. It can be explained that the reason for the superiority of IRGACURE 819 over IRGACURE 184 and LUCIRIN TPO is its stronger UV absorption than others. In addition, a high amount of photoinitiator leads to a large increase in the curing speed, and it is possible that some areas do not polymerize completely [31].

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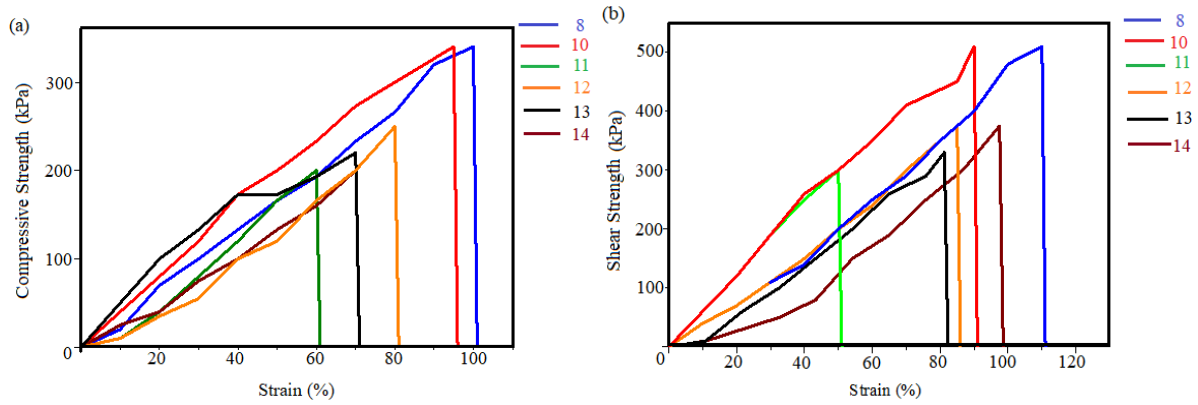

Fig. 5. Comparison of the mechanical properties of samples of UV-curable composites containing IRGACURE 184 and 819 and LUCIRIN TPO (samples 10-14) in, a) compressive strength, b) shear strength.

3.1.4. Effect acrylic acid

In the mechanical test, in sample 15, the effect of acrylic acid (1 wt.%) with the optimal conditions such as 25 wt.% PETA, 7 wt.% TMSPMA and 3 wt.% IRGACURE 819 was investigated. The compressive and shear strength tests are shown in Figs. 6-a and 6-b, respectively. The presence of acrylic acid not only improved the mechanical properties, but also reduced the compressive and shear strength by 27% and 28%, respectively, compared to sample 8 (sample 8 had the best mechanical properties among samples 1-14 and did not contain acrylic acid). The compressive strength and failure strain of sample 15 were 0.245 MPa, and 78%, respectively. Even in the shear condition, the strength and failure strain of sample 15 were 0.37 MPa, and 65%, respectively. The reason for the deterioration the properties can be seen in the increase in the curing speed, which led to incomplete polymerization in some places and reduced the mechanical properties [31].

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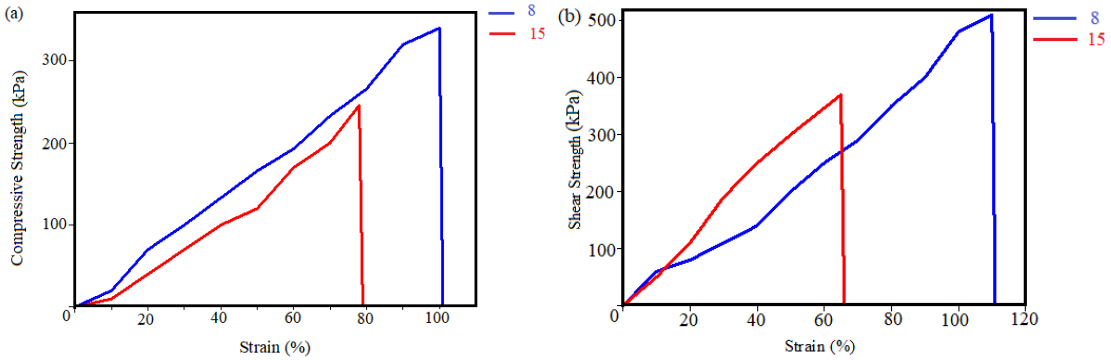

Fig. 6. Comparison of the mechanical properties of samples of UV-curable composites containing acrylic acid (sample 15) in, a) compressive strength, b) shear strength.

3.1.5. The effect of nano silica

In the mechanical test, the effect of the presence of nano silica (1-5 wt.%) with such as 25 wt.% PETA, 7 wt.% TMSPMA and 3 wt.% IRGACURE 819 were investigated in samples 16-20. The presence of nano silica from 1-4 wt.% increases the mechanical properties as shown in Figs. 7-a and 7-b. The presence of nanoparticles in the samples increases the strength after curing through pinning, cracks deflection and other mechanism [20]. Even with 5 wt.% nano silica (sample 20), the properties decrease due to the possibility of agglomeration of the nanoparticles. The best result between samples containing 1-5 wt.% nano silica refers to the sample containing 4 wt.% nano silica (sample 19), which has a strength of 0.42 MPa and 90% failure strain in the compressive state. For this sample, the shear strength and failure strain are 0.63 MPa and 100%, respectively. Compared to the best sample without nanoparticles (sample 8), sample 19 has 23% improved properties in both compressive and shear strength.

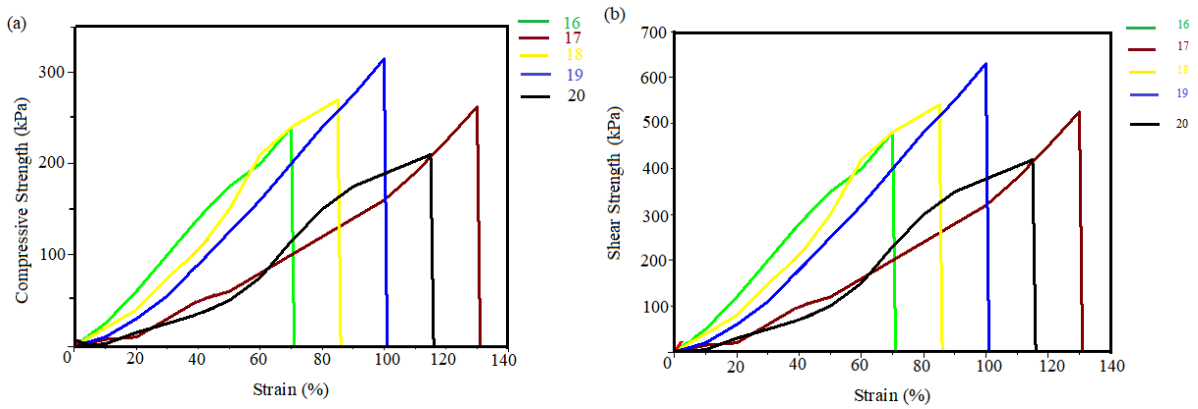

Fig. 7. Comparison of the mechanical properties of samples of UV-curable composites containing nano silica (samples 16-20) in, a) compressive strength, b) shear strength.

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3.2. DC results

The main purpose of the FTIR test was to obtain DC (equation 1) and determine the effect of weight percentage of nanoparticles on DC. This value was obtained directly from FTIR analysis and by comparing the two peaks at 1610 and 1635 cm^{-1} as shown in Fig. 8. FTIR graphs are obtained for samples containing 1-5 wt.% nano silica (samples 16-20) in cured and uncured states. Therefore, the DC can be determined in the form of the weight percentage of nano silica (according to Table 2). The composite containing 1 wt.% nano silica has the highest DC at 83%. By increasing the percentage of nano silica from the lowest (1 wt.%) to the highest (5 wt.%), the DC decreased by 11%. This result is due to the reduction in the depth of light penetration, the light does not reach to the base resin properly and the polymerization is not completed. The presence of nanoparticles during polymerization also causes disturbances in some places and leads to a decrease in DC [31].

Table 2. Absorption in FTIR peaks 1610 and 1635 cm^{-1} for cured and uncured samples of UV-curable composites containing 1-5 wt.% nano silica (samples 16-20) for DC calculation

Sample	**Peak on 1610 cm^{-1}**		**Peak on 1635 cm^{-1}**		**DC**
No.	**Uncured**	**cured**	**uncured**	**cured**	**(%)**
16	89.29	99.24	62.46	98.87	83
17	91.13	35.38	91.09	44.87	78
18	25.84	98.29	21.83	112.46	74
19	90.8	82.1	90.93	111.71	73
20	90.5	66.09	89.5	88.7	72

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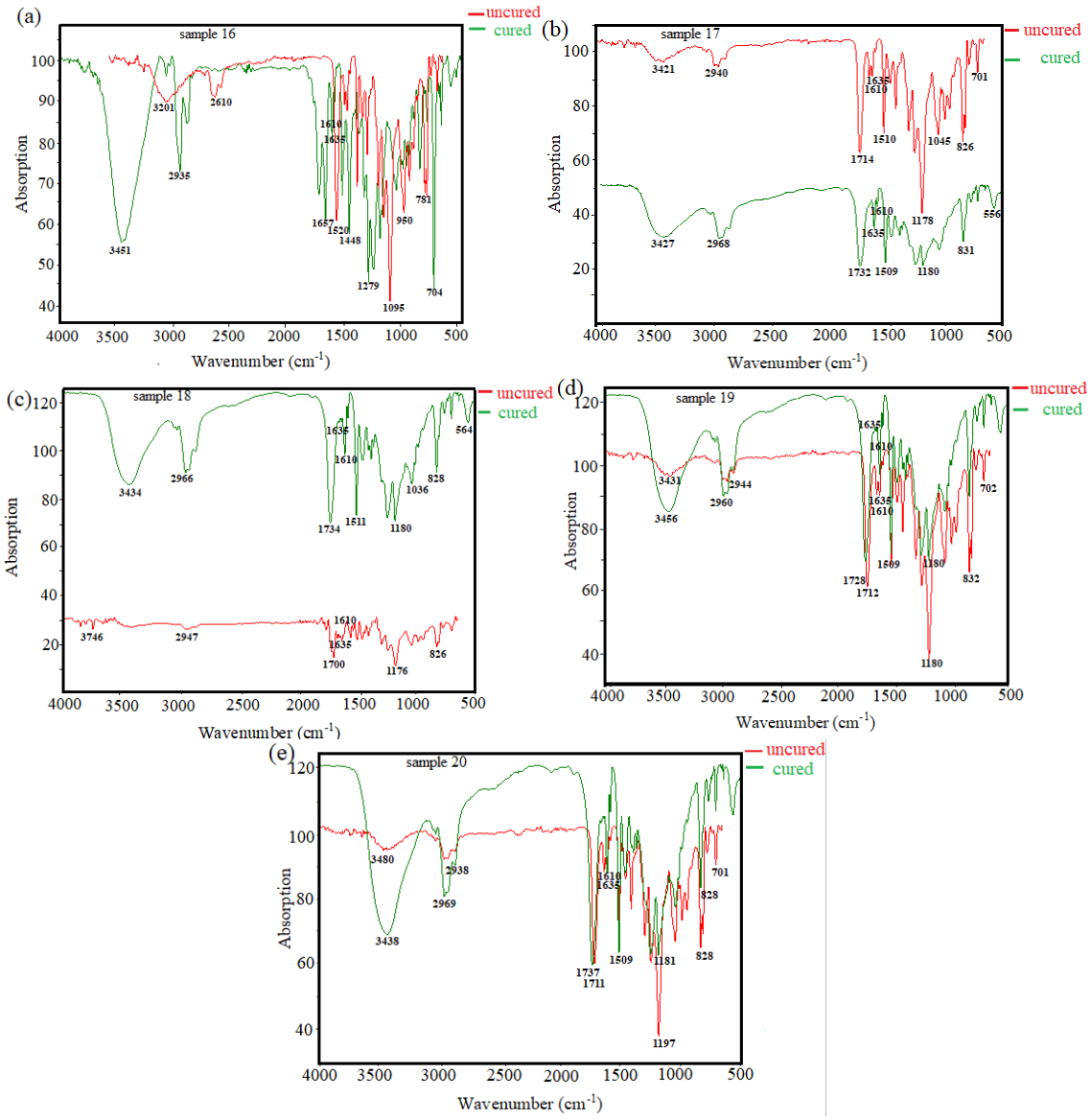

Fig. 8. FTIR graph for DC calculation for UV-curable composites samples, a) sample 16 (1 wt.% nano silica), b) sample 17 (2 wt.% nano silica), c) sample 18 (3 wt.% nano silica), d) sample 19 (4 wt.% nano silica and e) sample 20 (5 wt.% nano silica).

3.3. DMTA result

DMTA measures the deformation of a material in response to oscillating forces as a function of temperature. In principle, the DMTA technique captures the viscoelastic behavior of polymer materials. It provides quantitative results for the tensile storage modulus (E'), and the corresponding loss modulus (E''), the loss tangent (tan d), can then be expressed as the quotient of the loss and storage modulus. E' and E'' characterize the elastic and viscous components of a material under deformation, respectively, and E' is a measure of the mechanical power stored

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under load. E" quantifies the energy that is converted into heat when the material is deformed. As the flexibility of the composite materials increased, the Tg decreased [13, 33].

The storage and loss modulus, tan d and loss temperature of samples containing 1-5 wt.% nano silica are shown in Fig. 9. It can be seen that with an increase of nanoparticles from to 1-5 wt.% in the composition, the storage modulus has continuously increased (from 2444 to 3458 MPa), which may be caused by stronger bonding of silica nanoparticles to polymer chains [33]. Furthermore, a comparison of the loss modulus between samples containing 1 and 5 wt.% nano silica shows that sample containing 1 wt.% nano silica (sample 16) has the highest loss modulus at 221 MPa. Sample containing 5 wt.% nano silica (sample 20) has the lowest loss modulus at 169 MPa. An 18% decrease in loss modulus was also observed from to 3-5 wt.% of nano silica. Regarding tan d, it can be said that the highest value for a sample containing 3 wt.% nano silica (sample 18) is 1.03, and the lowest value is 0.39 for a sample containing 5 wt.% nano silica (sample 20). Furthermore, a 62% decrease in tan d was observed from to 3-5 wt.% nano silica. The temperature at the peak of the tan d diagram indicates the temperature at which the material reaches its maximum loss factor and energy loss. In other words, this point represents the best balance between flexibility and viscosity of the material. At this stage, the loss coefficient has highest value, indicating the highest energy loss in the material at this temperature. This point is commonly known as the "loss temperature peak" or the "tangent temperature peak". The temperature at the peak of the tan d chart helps identify the temperature at which the material exhibits the most significant loss and elasticity characteristics. Assuming that a sample containing 2 wt.% nano silica (sample 17) has the highest loss temperature (113.8 °C) and a sample containing 3 wt.% nano silica (sample 18) has the lowest loss temperature (92.5 °C). Also, an increase by 20.4 °C in loss temperature was observed from to 3-5 wt.% nano silica.

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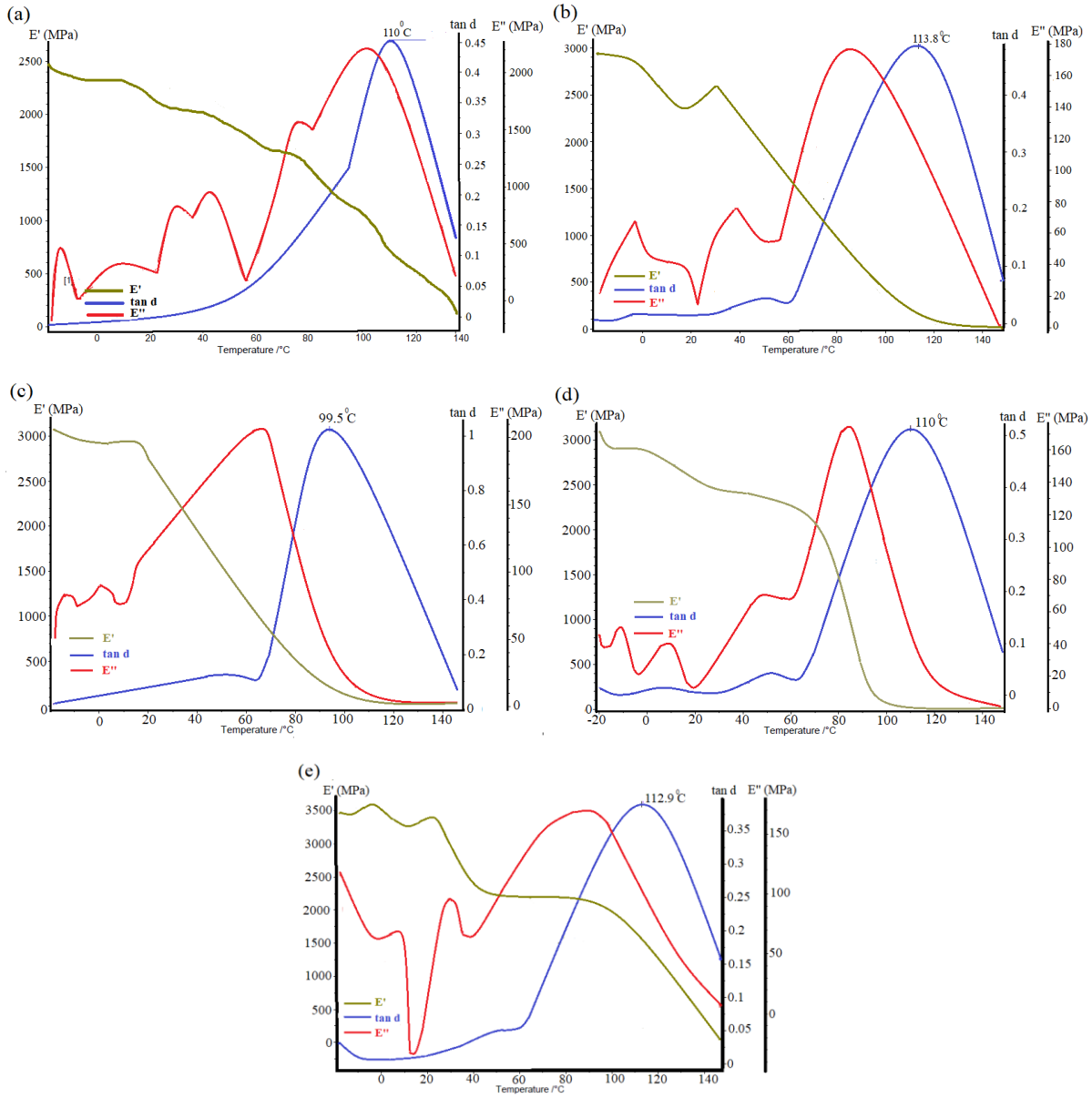

Fig. 9. DMTA curve for samples of UV-curable composite, a) sample 16 (1 wt.% nano silica), b) sample 17 (2 wt.% nano silica), c) sample 18 (3 wt.% nano silica), d) sample 19 (4 wt.% nano silica, e) sample 20 (5 wt.% nano silica).

3.4. SEM results

Cured samples containing of 1-5 wt.% nano silica (samples 16-20) were subjected to SEM analysis (Fig. 10). The main reason for selecting these samples was the presence of nanoparticles in their composition, the distribution of the nanoparticles, and their agglomeration. All samples confirmed proper distribution. The white dots in the images are nano silica particles as indicated by the red arrow and circle. In sample 16, which containing 1

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wt.% nano silica, can be seen, the nanoparticles are arranged at a suitable distance are on the nanoscale and do not show any signs of agglomeration as shown in Fig. 10-a. Another thing that can only be seen incidentally in this image indicates the existence of a molten pool, although this area can probably be considered an uncured part according to the explanation of the DC in FTIR section. Furthermore, with the increase of nano silica to 2 wt.% (sample 17), as shown in Fig. 10-b, the proper distribution of nanoparticles was confirmed and no effect of agglomeration was observed. In sample 17 wasn't seen effect of presence of the molten pool, nevertheless, the nanoscale particles were seen. As can be seen in sample 18 with 3 wt.% nano silica, the nanoparticles were not agglomerated and are still in a nanoscale as shown in Fig. 10-c. In sample contains 4 wt.% nano silica (sample 19), the particles are very close to each other and the nano silica particles are agglomerated in a small part of the points (Fig. 10-d,). Finally in Fig. 10-e, the agglomeration of nanoparticles can be seen and the loss of mechanical properties due to the addition of 5 wt.% nano silica (sample 20) to the base composition can be justified. Additionally, there were multiple SEM images for each sample containing nano silica in this study. The distance between the nanoparticles was almost the same in all of them, confirming the good dispersion. Since the ultrasonic duration was 30 minutes and this amount ensures a perfect distribution of the nanoparticles in the epoxy-acrylate base.

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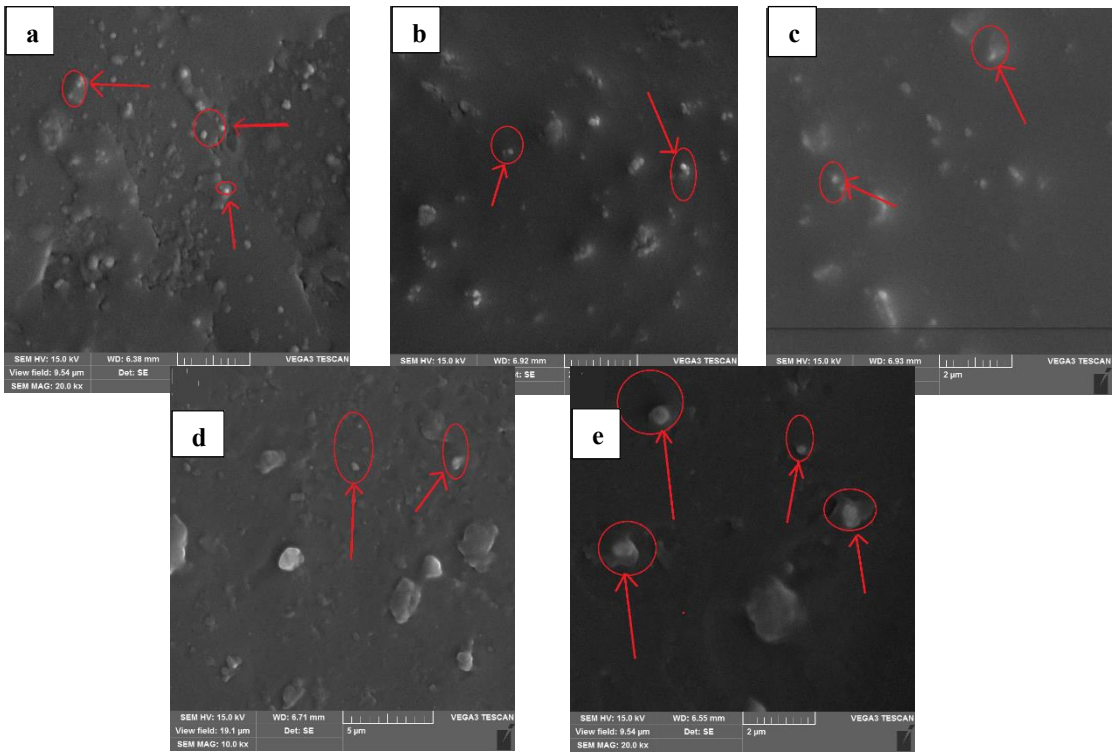

Fig. 10. SEM images for samples of UV-curable composites, a) sample 16 (1 wt.% nano silica), b) sample 17 (2 wt.% nano silica), c) sample 18 (3 wt.% nano silica), d) sample 19 (4 wt.% nano silica, e) sample 20 (5 wt.% nano silica).

In comparison results of prepared UV-curable composite for bonding glass to glass with related previous studies, briefly, it can say that there are advantages. In the present study, epoxy acrylate as an oligomer and TMPSMA as a silane coupling agent were used in the composition of a UV-curable composite for glass-to-glass bonding applications. But the other researchers weren't used that composition in their works [3, 30, 33-37]. Regarding the mechanical properties in current research, it was found that the presence of TMPSMA improved the shear and compression strength of this composite for glass to glass bonding. The curing process (DC parameter) in current study was better than related previous works. The best DC of samples was 83%, that the comparison with relevant works like as 40.94% [30], 78% [36] and 58% [26] was better. In the DMTA test, Shirkavand et al. [33] have the same storage modulus with this current research, but their loss temperature and loss modulus were lower. In the SEM study, samples containing nano silica in base of epoxy acrylate showed good dispersion in their all

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SEM images. This SEM results in comparison with pervious works with the same condition like as Xue et al. [26] and Hadavand et al. [33] were better.

4. Conclusion

In this research, a UV-curable composite was prepared for bonding glass to glass. The effect of various monomers (TMPTA, TPGDA and PETA), TMSPMA as a coupling agent, IRGACURE 184, IRGACURE 819, and LUCIRIN TPO as photoinitiator, acrylic acid and nano silica as adhesion improver were investigated in different weight percentages. The results of this research were obtained from tests of mechanical compressive and shear properties, FTIR, DMTA and SEM.

Due to the mechanical properties, PETA achieved the best result among three monomers at 30 wt.%. This sample containing PETA had a compressive strength of 0.3 MPa and a shear strength of 0.45 MPa. The best properties for samples containing TMSPMA were at 7 wt.% with a compressive strength of 0.34 MPa and a shear strength of 0.51 MPa. The higher silane content in the composition, the more methanol is produced and the mechanical properties become worse. The properties obtained, showed the superiority of a sample containing 3 wt.% IRGACURE 819 by compressive strength of 0.34 MPa and shear strength of 0.51 MPa. The effect of acrylic acid at 1 wt.% resulted in deterioration of properties with compressive strength of 0.245 MPa and shear strength of 0.37 MPa. The best result for samples containing 1-5 wt.% nano silica and the best sample overall was achieved for the sample containing 4 wt.% nano silica, 24 wt.% PETA, 7 wt.% TMSPMA and 3 wt.% IRGACURE 819 which gave a compressive strength of 0.42 MPa and a shear strength of 0.63 MPa. The sample containing 5 wt.% nano silica deteriorated the mechanical properties, because nanoparticles agglomerated or prevented UV radiation from achieving complete polymerization.

The samples containing 1-5 wt.% nano silica were subjected to FTIR test to obtain DC. By increasing the amount of of nanoparticles from 1-5 wt.%, the DC ultimately decreased, indicating the growth of unreacted monomers and polymerization defects in some places. In the DMTA test, have to measure the mechanical properties of samples as a function of temperature in samples containing 1-5 wt.% nano silica. The storage modulus increased by 41% from lowest content of nano silica to highest one. The highest loss modulus and loss

temperature refer to the sample containing 1 and 3 wt.% nano silica at 221 MPa and 113.8 °C, respectively. In the SEM study, samples containing 1-4 wt.% nano silica exhibited proper distribution of nanoparticles. All SEM images of all samples confirm these results, and the sample containing 5 wt.% nanoparticles showed agglomeration in all SEM images.

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