



Effect of Argon laser Power Density and Filler content on Physico-mechanical properties of Dental nanocomposites

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ABSTRACT

The effect of the 472 nm wavelength of argon laser at different power densities and irradiation times and different filler contents on the degree of conversion (DC), flexural strength (FS) and flexural modulus (FM) of two dental nanocomposites was studied. The nanocomposites were cured applying the laser beam with power densities of 260 mW/cm² and 340 mW/cm² at different irradiation times. The DC% of the nanocomposites were measured using Fourier transform infrared spectroscopy. The flexural strength and flexural modulus were also measured and compared. The results indicated that the degree of conversion of composite containing 20% filler cured at power density of 340 mW/cm² was significantly higher than other composites. The minimum degree of conversion was achieved for the cured composite with 260 mW/cm², involving 25% filler. The composite with 25% filler cured at 340 mW/cm² showed the highest flexural strength and flexural modulus. The composite including 20% filler cured at 260 mW/cm² exhibited the lowest flexural strength and flexural modulus. The results confirmed that higher power density results in higher degree of conversion, higher mechanical properties and decreased curing time. In addition, higher filler loading improved the flexural properties.

Keywords: Dental nanocomposite, Argon laser, Degree of conversion, Flexural properties, filler

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INTRODUCTION

When the hard tissue of teeth is damaged by dental caries and cavity preparation, the use of visible light-cured dental composites is the best treatment due to easier to use, excellent aesthetic and positive physical properties. Light-cured dental composites consist of hard inorganic particles dispersed in a soft organic resin matrix. Properties of composites influenced by properties of their fillers and chemical structure of monomers used in matrix phase [1- 3]. The most widely used resin in dental composites is that based on the copolymer prepared from a combination of bisphenol A glycol dimethacrylate (Bis-GMA) and triethylene glycol dimethacrylate (TEGDMA). TEGDMA is usually added to Bis-GMA in order to achieve workable viscosity limits since the latter monomer possesses very high viscosity due to the intermolecular hydrogen bonding [4]. Silica is used in preparation of dental nanomaterials. Most of the fillers which are used to reinforce dental composites are silicate glasses. These materials with nanosilica fillers provide materials with high mechanical properties and low polymerization shrinkage [5] Camphorquinone (CQ) is extensively used as a photosensitizer for the visible light cured composite resin [6]. Since the introduction of dental restorative resins, several energy sources, such as UV, Halogen lamps and LED (light emitting diodes) curing units, have been used to initiate the photopolymerization process of the resins. UV curing units is no longer used in dental applications because of its side effects on the oral mucous [7- 9], but lasers such as argon laser with its inherent optical characteristics like low beam divergence, monochromacity, collimation, coherency, absorption selectivity because of wavelength tunability, and fiber delivery capability can all make it to be practically a better candidate, which effectively can reduce curing time, provide a higher degree of conversion (DC) of monomers, and enhance physical properties of cured composites [10-12]. On the other hand, the argon laser appeared as a

suitable alternative polymerization source of composite resins, particularly when the camphoroquinone, with broad peak activity in the 470 nm range, is used in dental composite as a photoinitiator [13]. Therefore, the aim of this study was to apply argon laser as a light source for photopolymerization of dental composite resins and investigate the effect of power density of the argon laser and filler content on the flexural properties and degree of conversion of dental nanocomposites.

MATERIALS AND METHODS

The resin mixture used in this investigation consisted of Bis-GMA and triethylene glycol dimethacrylate (Fig. 1) with a mass ratio of 65/35 were obtained from Rohm (Degussa Group, Germany). This composition is similar to that used in a variety of commercial dental resin formulations. 20 wt% and 25 wt% of nanosilica fillers (Waker), with primary particle size of 10 nm, aggregated particles size of 50nm and agglomerated particles in micrometer size were added to the resin. The nanocomposites were then placed in a sonication instrument (Bandelin, Sonopuls, HD2200, P150W/300W, AF Frey-35 kHz) for 10 min, to break up the agglomerates of SiO₂ particles. The initiator used in this experiment was the visible light-initiating system of camphorquinone (Fig. 2) (0.5 wt%) and N,N'-dimethyl aminoethyl methacrylate (DMAEMA, 0.5 wt%) were supplied by Fluka.[14] 0.5% CQ as a photoinitiator was used for the optimization of the DC. Finally, two types of nanocomposites with different filler contents were obtained. The specimens (N=10, n=5) were prepared by the molding method with different shapes and diameters. The samples were then cured in an *in vitro* situation by an argon laser (Mellos Griot, 543 series, USA) at 472 nm wavelength using different power densities and irradiation times.

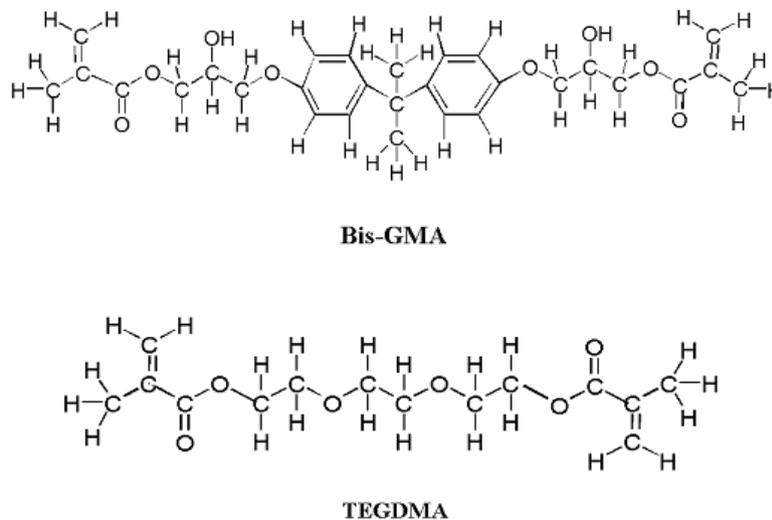


Fig. 1. Chemical structural formula of monomers used in dental resin

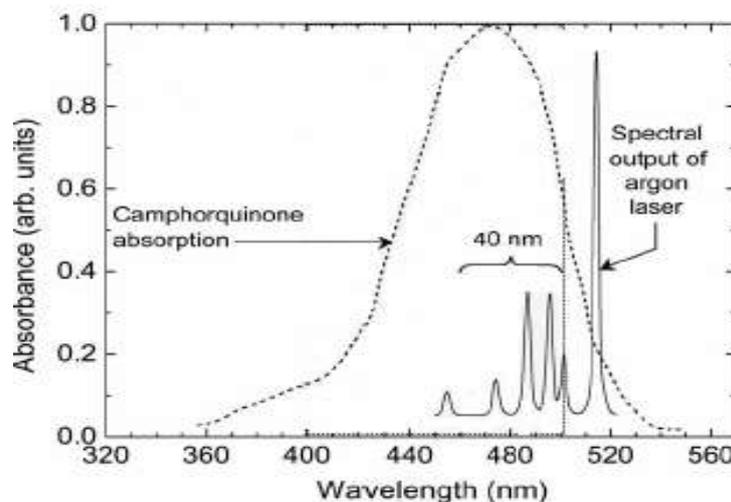


Fig. 2. Absorption spectrum of camphorquinone [14]

Nanocomposites characterization

Degree of conversion (DC%)

In this work, to measure the DC%, the uncured paste of each nanocomposite was placed between two polyethylene films, pressed to form a 100- μm -thick film and mid-infrared spectra of absorbance peaks were obtained by transmission mode of Fourier Transform infrared (FTIR; Bruker, EQUINOX 55, Germany). The samples were then cured by applying the argon laser beam at wavelength of 472 nm and power densities of 260mW/cm² and 340mW/cm² for successive short periods of irradiation times of 5, 10, 20, 40 and 60 s and the absorbance peaks were recorded for the cured samples. For the reactions at different power densities of laser beam, we selected two densities. The wave number mentioned led to a 340 mW/cm² output. Meanwhile, we used a filter to obtain another power density (260 mW/cm²). The power densities were obtained with Coltolux lightmeter (Coltene, Whaledent Inc, USA). The percentage of unreacted carbon-carbon double bonds was determined from the ratio of absorbance intensities of aliphatic C=C (peak at 1638 cm⁻¹) / aromatic C...C (peak at 1608 cm⁻¹) before and after curing of the specimen. The degree of conversion was then obtained as follows [15]:

$$DC\% = \left(1 - \frac{(1638 \text{ cm}^{-1}/1608 \text{ cm}^{-1}) \text{ peak area after curing}}{(1638 \text{ cm}^{-1}/1608 \text{ cm}^{-1}) \text{ peak area before curing}} \right) \times 100 \quad (1)$$

Flexural strength (FS) and flexural modulus (FM)

Bar-shaped test specimens 2.0 mm \times 2.0 mm \times 25.0 mm in size were prepared using stainless steel moulds. The specimens were polymerized with the argon laser at the wavelength of 472 nm and power densities of 260 and 340 mW/cm² for 45 and 35 s, respectively. After that, the cured specimens were stored in distilled water for 24h at 37°C before the test. The flexural strength and flexural modulus were measured using three-point bending fixture of SMT-20 universal testing machine (Santam, Iran) with a span of 20 mm between supports and loaded to failure point at a crosshead speed of 0.5 mm/min using a 60N load cell. Five specimens for each group were tested and their mean value was determined. The values of flexural strength and flexural modulus were calculated using the following formulae [16, 17]:

$$FS = 3Fl/2bh^2 \quad \text{and} \quad FM = l^3F/4bh^3d \quad (2)$$

where f is the maximum force (N), l the distance between the supports (mm), b the specimen width, h the height of the bar (mm) and d the deflection under load (mm). The experimental results were examined by one-way analysis of variance (ANOVA), based on quintuplicate specimens for degree of conversion, flexural modulus and, flexural strength testing. Multiple pairwise comparisons were further conducted, using Tukey's test with a significance level of 0.05.

RESULTS

Fig. 3 shows the FTIR spectra of the nanocomposite involving 20% and 25% filler, before and after curing with 260 and 340 mW/cm² and with a frequency range between 1690 and 1570 cm⁻¹. The quantity of the remaining double bonds was determined by a method described by Ruyter and Gyrosi [18]. The absorbance peak at 1638 cm⁻¹ refers to the aliphatic C=C stretching of the vinyl group, which in fact can be used to quantify the methacrylate double-bond conversion, and the peak at 1608 cm⁻¹ assigned to the aromatic C-C bond was used as an internal standard. Thus, the decrease in absorbance intensities of bonds and the area peak to peak under the curve effectively indicates the amount of double bond consumed during polymerization.

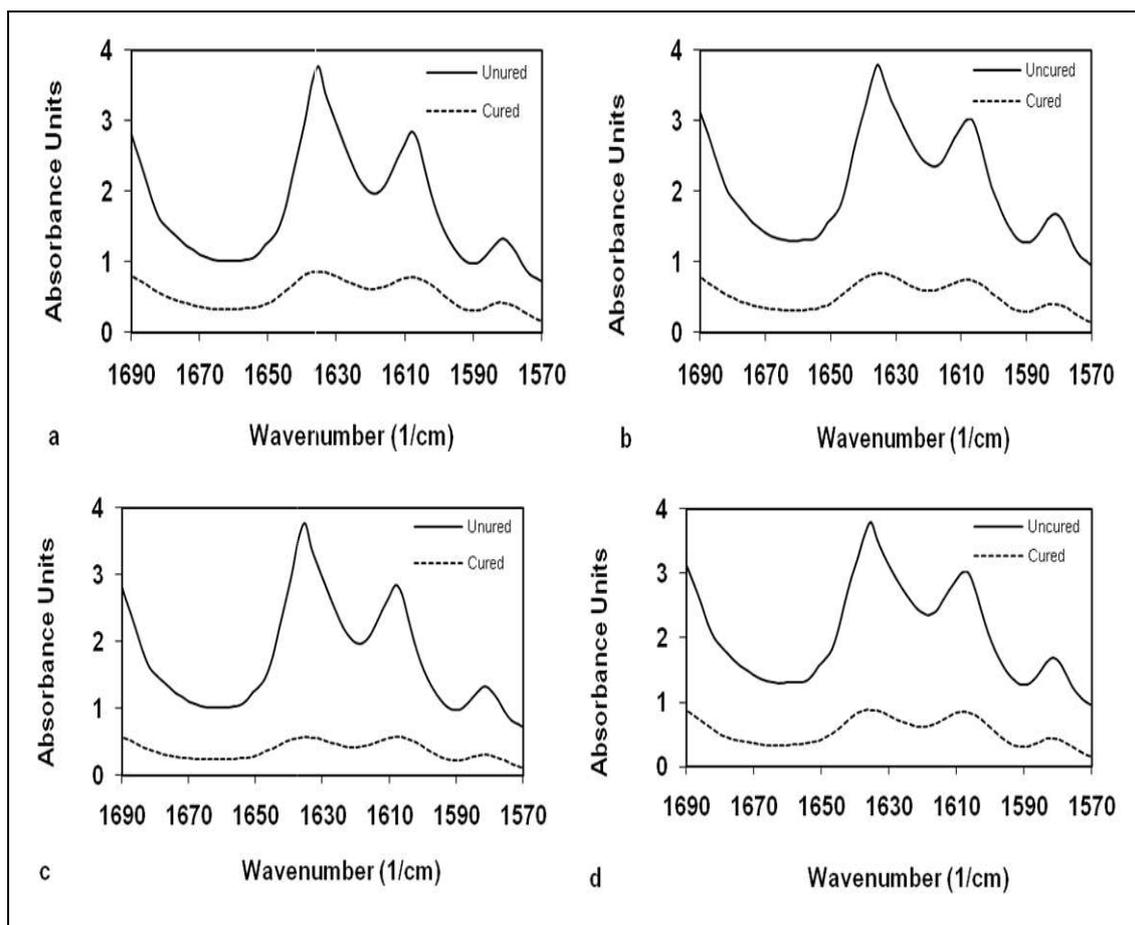
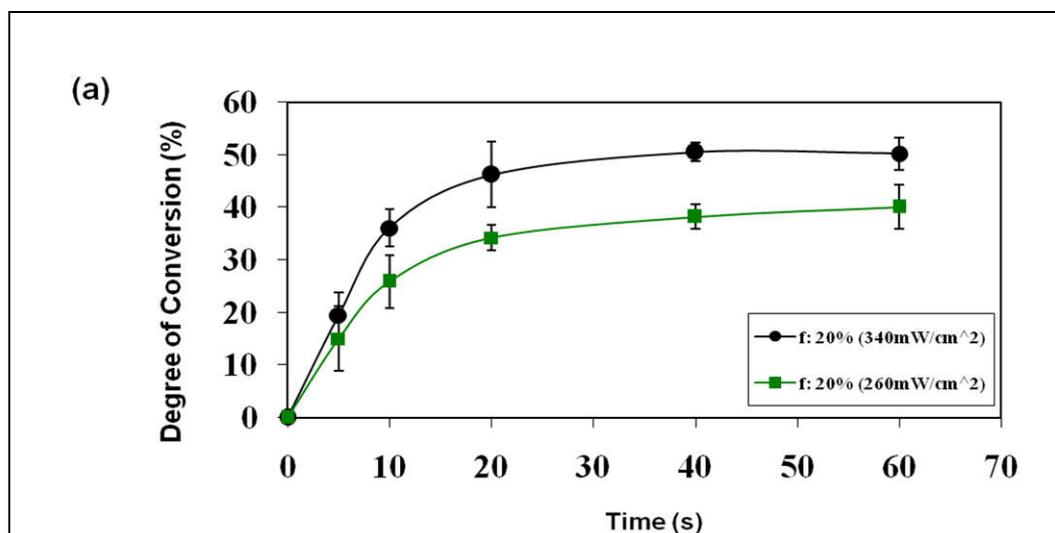


Fig. 3. FTIR spectra of uncured and cured nanocomposite with 260 mW/cm², involving 20% filler (a) 260 mW/cm², involving 25% filler (b) 340 mW/cm², with 20% filler (c) and 340 mW/cm², including 25% filler (d), with a frequency range between 1690 and 1570 cm⁻¹.

In Fig. 4 the degree of conversion is shown as a function of irradiation time for cured specimens with 260 and 340 mW/cm², which composed of 20 filler. As it is seen, the DC% reaches its maximum after about 20 s, which for the composite with 20% filler cured at power density of 340 mW/cm² and 260 mW/cm², correspond to about 50% and 40% (a) and for the composite cured at mentioned power densities involving 25% filler obtained about 45% and 35% (b) respectively. These values remained almost constant over a further irradiation time of 60 s. The results show that for the same filler loading, higher power density result in the higher degree of conversion.



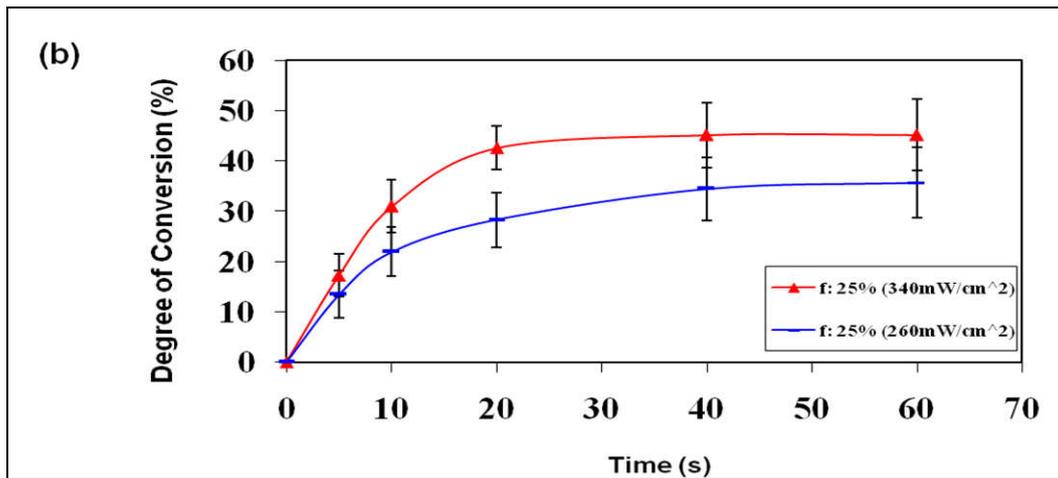


Fig. 4. The effect of power density on Percent degree of conversion for cured nanocomposites

Fig. 5 shows the influence of filler loading on degree of conversion for different power densities in separate diagrams. As can be seen, the degree of conversion decreased as the filler loading of nanocomposites increased. Therefore, for each power density, the maximum DC refers to specimen with lower filler content.

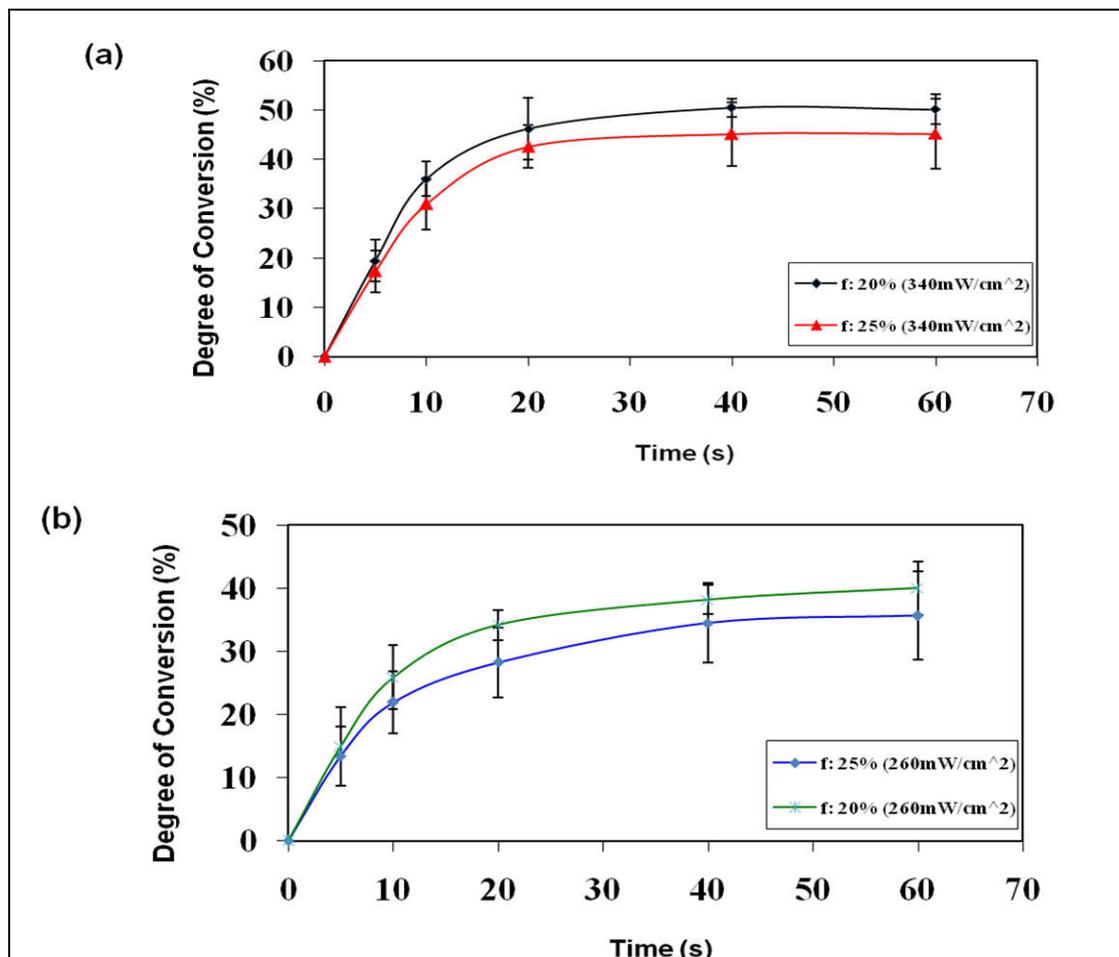


Fig. 5. The effect of filler content on Percent degree of conversion for cured nanocomposites

Other important factors that should be taken into account when characterizing a cured composite behavior are flexural strength and flexural modulus. Fig. 6 indicates the stress-strain curves of specimens cured with different power densities and filler contents. As this curves show, a given stress is required for each composite is broken down. The comparison of the stress-strain curves shows that by increasing the

intensity of the light source and filler loading of the nanocomposite the required stress for each given strain is increased. It should be noted that the shape of the stress-strain curve for such composites is often not unique, as the value of the strain produced depends upon the rate at which the stress is applied.

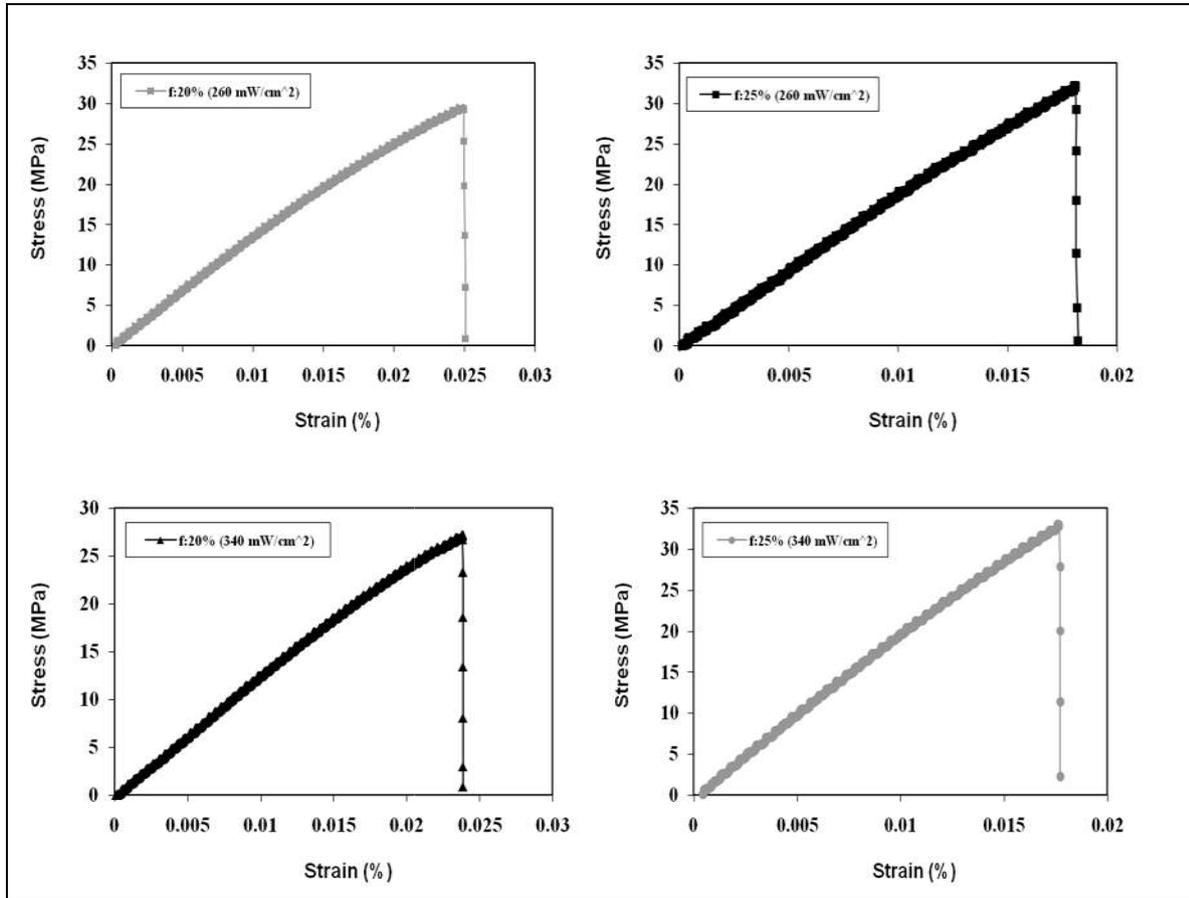
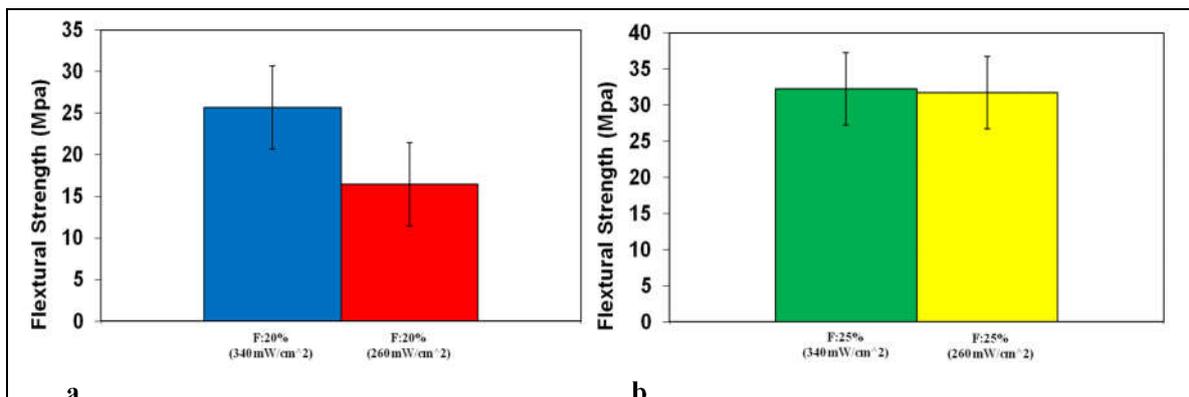


Fig. 6. The relationship between stress and strain of cured nanocomposites

The effect of power density on Flexural strength and Flexural modulus of cured nanocomposites including 20 and 25% filler is shown in Fig. 7. As this figure show, the FS and FM of the specimens cured at power density of 340mW/cm² are higher than the specimens cured at power density of 260mW/cm² for each filler content.



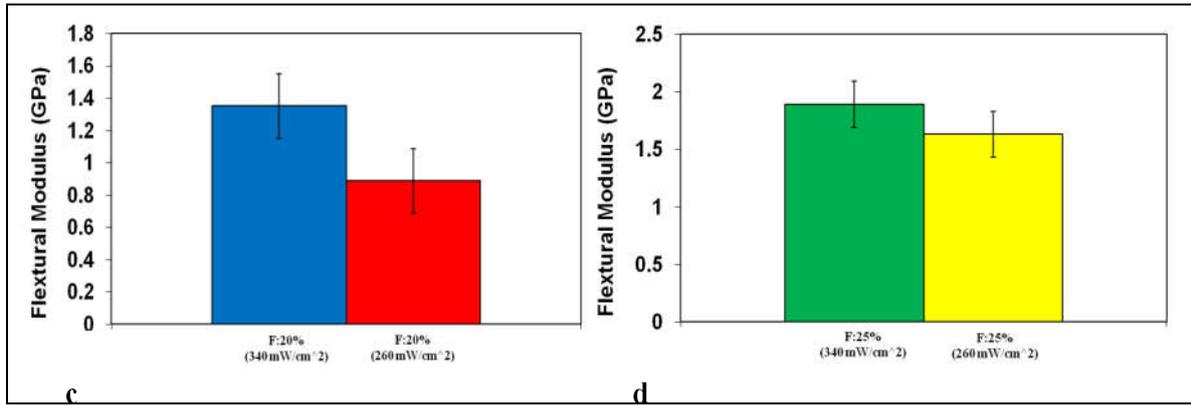


Fig. 7. The influence of power density on Flexural strength (a and b) and Flexural modulus (c and d) of cured nanocomposites.

Fig. 8 indicates the effect of nanocomposites-filler-percent on flexural strength and flexural modulus.

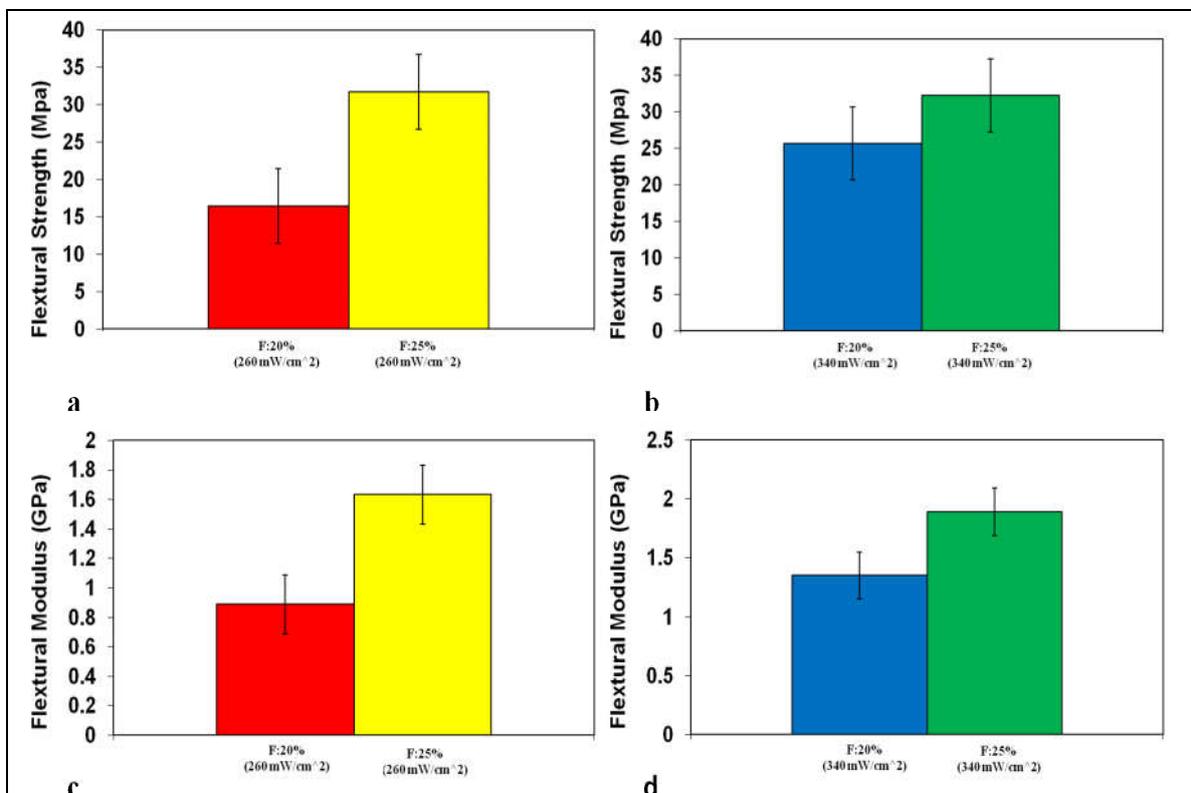


Fig. 8. The effect of filler loading on Flexural strength (a and b) and Flexural modulus (c and d) of cured nanocomposites.

According to obtained results from figure 7 and 8, we believe that increasing the intensity and filler loading were effective to maximize the flexural strength and flexural modulus of nanocomposites. Both of these factors can contribute to enhance mechanical properties of specimens.

DISCUSSION

Recently, argon laser is extensively used in dentistry in a variety of applications. For the argon laser curing the highest degree of conversion can be obtained using 472 nm emission line which is in the range of blue light. The 472 nm line is close to the camphorquinone absorption peak. The absorption peak of camphorquinone is about 470 nm. However, the spectral distribution of the output from the curing light source and the maximum absorption peak of a photoinitiator can affect the chemical and physical properties of dental nanocomposites. Furthermore, to our knowledge, not much is available in the literature about the effect of power density and percent of filler on degree of conversion and physico-mechanical properties of the (Bis-GMA+TEGDMA+SiO₂) mixture with the mass ratio as in our case being

treated by a 472 nm argon laser. Thus, with respect to this, we have studied and presented the results regarding the effects of laser intensity and filler loading on DC, FS and FM.

The photopolymerization of dimethacrylate at room temperature normally leads to a glassy polymer in which only a fraction of the available double bonds are reacted. The degree of conversion depends on the chemical structure of dimethacrylate monomers, the polymerization conditions, i.e., type of light source, temperature, intensity, wavelength, and photoinitiator concentration [20, 21]. The DC% can be increased at high resin content and as expected, increasing the intensity increased the DC% from 35 to about 50% to form a reacted polymer network (Fig. 4). Generally, a low DC% of composites at a lower light intensity can cause lower free radical formation than when higher light intensity is used. This effectively means that there will be a delayed interaction between adjacent chains and hence will take somewhat a longer time to reach the gel points. Knowing that the viscosity is a measure of the resistance of molecules to flow and a high viscosity value is indicative of the presence of intermolecular interactions, therefore, these interactions can cause a decreased mobility of monomer molecules during photopolymerization and also a decreased flexibility of the corresponding polymeric network. On the other hand, one must note that the higher DC% exposed to a higher intensity level is not really advisable because cross-linking occurs faster and localized microgels can be formed, which can act as filler particles and increase the viscosity of the material [22]. As a result, the pure viscous flow is disappeared, and stresses begin to develop as the polymerization proceeds. It is therefore best to operate under an optimized condition where a clinically acceptable mechanical strength of resin because of complete conversion is achieved without marginal shrinkage and residual thermal stresses because of fast network formation. The enhanced conversion is reflected in the resultant improvement in physical properties and flexural strength. The degree of conversion results obtained in this investigation indicated that the composite containing 80% resin cured at power density of 340mW/cm² gave maximum DC%. The minimum of DC% occurred when the content of resin was 75% and applied power density was 260 mW/cm².

The closest works we found in the literature to compare our results with were those performed by Khosroshahi *et al*. In the same research, they prepared a dental resin without any filler. The monomer mixture used in their investigation consisted of Bis GMA and TEGDMA with a mass ratio of 75/25. They also used camphorquinone (CQ, 0.5% wt) and N,N'-dimethyl aminoethylmethacrylate as a photoinitiator system and then cured the dental resin by our argon laser (Mellos Griot, 543 series, USA) at 488 nm using different power densities. As their results show, the specimen degree-of-conversion cured with 1075 mW/cm² is higher than the specimen cured with 700 mW/cm² (52.2% and 40.1%, respectively). [19]

Other important parameters affecting the physicomaterial properties of dental composite are flexural strength and flexural modulus. Changes in composition and chemistry of the constituent monomers and filler can alter the physical properties of the materials. Figure 6 shows that the value of the strain produced for each nanocomposite depends upon the rate at which the stress is applied. The values of flexural properties depend mainly on the intensity and percentage of filler. As it is observed in figures 7 and 8, high intensity enhances mechanical strength due to increasing of the degree of conversion and high filler loading can also improve the flexural properties due to hardness of this component. The results show that the nanocomposite with 25% nanosilica that cured with 340mW/cm² exhibited the maximum flexural strength and modulus.

It is worth mentioning that Chen *et al*. demonstrated that, when the content of inorganic fillers was increased, the nanocomposites exhibit high flexural strength and modulus. [1] Soares MA *et al* explained that high power density can improve flexural strength and flexural modulus and result in a high degree of conversion. [23].

CONCLUSIONS

Argon laser beam was applied to initiate the photopolymerization of experimental dental nanocomposites. The investigation of degree of conversion and flexural properties of visible light-cured dental nanocomposites by different filler content and argon laser power density showed that by using high intensity the degree of conversion was increased and the curing time was reduced to about 20 s. In addition, flexural strength and flexural modulus enhanced as the filler loading of nanocomposites increased. Furthermore, the developed cured nanocomposite with higher power density and percent of nanosilica is suitable for dental restorative material applications due to short curing time and high degree of conversion and flexural properties.

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