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Improvement mechanical properties of flowable dental composites by modified Cloisite

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Abstract:

Objective: The aim of the study was to modify Cloisite Na+ by glycidyl methacrylate and its use as a reinforcement filler in the flowable dental composites and also investigation of their physical and mechanical properties.

Methods: The hydroxyl groups of Cloisite was reacted with glycidyl methacrylate to bond with flowable dental composites (Tetric Flow, Invoclar, Vivadent, USA) and then mixed with flowable dental composites by high-speed mixer at room temperature in different concentration of 0.2, 0.5, 1, 2.5, 5 and 7.5 Wt% and one control group with no additive. Morphology of composite layers was confirmed with different techniques, such as transmission electron microscopy (TEM) and X-ray diffraction (XRD). The physical and mechanical properties of modified composites such as flexural strength, flexural modulus, compressive strength, water sorption and depth of cure for dental use were measured.

Results: The results showed that glycidyl methacrylate attached to the surface of Cloisite along with grafting in flowable dental composites on chemical attachments which improved the strength and flexural modulus as well as the tensile strength of the flowable dental composites.

Conclusion: The use of 0.2Wt% modified nanocloisite with glycidyl methacrylate in flowable dental composites can improve their mechanical properties.

Key Words: Clay, Composite resin, Mechanical tests, Calcium aluminosilicate clay.

Introduction

Dental caries and its clinical management is still a public concern in dentistry in the world and many articles have been published at this background which lead to increase the demand of dental materials [1]. Amalgam has known as a traditional dental material that has good mechanical properties and acceptable longevity without remarkable beauty, in addition, the release of mercury has caused global concern. For this reason, the tendency to use composites for dental restoration has increased [2]. Nowadays dentists prefer using composites for dental restorative due to the beauty. Dental composites involve a methacrylate base matrix resin with an optical primer and the high percentage of mineral fillers that give rise to optical activity and improve their mechanical properties [3]. The use of composites in dentistry has begun for more than 50 years and many of their features have improved such as filler size to improve their mechanical properties.

Recently, the flowable dental composites have been marketed and the purpose of producing these compounds is to easily work with them and to better penetrate in the cavity wall than previous products. The biggest problem of these composites is the weakness of their mechanical strength, especially when are used in places with high bearing loading [3, 4]. Nowadays flowable dental composites are used as sealants liners, fissure and class V restorative materials that their viscosity has reduced with decreasing filler content and by adding modifying agents [5]. It has been reported that these composites have a lower filler content of 20%-30% than conventional composites which has released to less mechanical strength [6, 7]. In order to solve this problem, researcher have tried to increase their mechanical properties while maintaining the viscosity of flowable dental composites systems [8-11].

Nowadays, added clay nanoparticles to polymer matrix has been extensively investigated, but its use is limited to the plastic industry [12-13]. The nanoclays involve nanometric lamellar structure which these thin layers filled by alkaline cations such as Al^{+3}, Mg^{+2}, Fe^{+2} and Li^{+} with a weak bond [14].

Recently, nanoclay fillers have been added to engineering composites as "phase reinforce" to increase mechanical properties [15-17], but their effects on flowable dental composites have been not studied yet [18]. The amazing properties of nanoclay are due to their intrinsic structure including a stacked thickness of 1nm and a width of a crystalline sheet with about 100 nanomaterials of silicon alumina. The dispersion of these plates by polymer matrix can improve the mechanical properties of nanocomposites [15, 16]. Cloisite is one of the most common types of clay which is used to create nanocomposites with general formula ($(Al_2Mg_3)Si_4O_{10}(OH)_2M^+$) that M^+ is a single valence cation. Few studies have been done about clay's effect on flowable dental materials [19]. In continuing our interest to improve dental composites [20-23], we wish to report in this paper the improvement mechanical flowable dental composites. To protect the filler surface against fracture and improve the distribution and transmission stress in resin [24], we used the glycidyl methacrylate to react with hydroxyl groups on the surface of the clay sheets to create an effective coupling between the clay and polymer matrix. The structure of modified Cloisite was determined using the standard methods including IR, TEM, TGA and XRD and eventually investigated its effect on mechanical properties, depth of cure and water sorption on flowable dental composites.

2. Material and methods

All chemicals used in this study were obtained from Sigma, Merck chemical companies and so Cloisite compound was purchased from southern clay products, Inc. United States.

A Shimadzu FT-IR 8300 spectrophotometer was used to prepare Fourier transform infrared (FTIR) spectra. The characterization of nanostructures was carried out by means of a Holland Philips Xpert X-ray powder diffraction (XRD) diffractometer (Cu Kα, radiation, $\lambda = 0.154056$ nm). A Philips CM10 analyzer operating at 100 KV was used for transmission electron microscopy (TEM) measurements. Thermal gravimetric analysis (TGA) was performed by means of a Shimadzu-50 system at a heating rate of 10◦C/min. Weight changes were followed as a function of temperature increase from 30 ◦C to 650 ◦C at a heating rate of 10 ◦C/min in N2 atmosphere. Scanning electron microscopy (SEM) Au spin coating for SEM sample preparation.

2.1. Modification of Cloisite

Cloisite (3 gr) was transferred to dry tetrahydrofuran (50 ml) and stirred for 10 hours at ambient temperature. The glycidyl methacrylate (24 ml) was dissolved in tetrahydrofuran (30 ml) separately and then added dropwise to the mixture of stirring solution of Cloisite for 6 h, and stirring was continued for an additional 24 h. Finally modified Cloisite was washed several times using distilled water and dried at ambient temperature, characterized by general techniques such as IR spectroscopy, TGA and X-ray Diffraction

2.2. Preparation and characterization of modified Cloisite composites

Modified Cloisite was mixed by flowable dental composite in seven groups with weight percentages of 0, 0.2, 0.5, 1, 2.5, 5, 7.5 using a high speed mixer at room temperature and conventional conditions. The mechanical tests were performed for all of groups the results which were obtained, revealed that the samples containing 0.2% modified Cloisite exhibit the most acceptable result and characterized by TEM and SEM techniques [25].

2.3. Flexural strength test

The three-point flexural strength was measured to prepare samples of modified Cloisite composites using the 4049 standard [26] with a testometric device (M350-10 CT Rochdale, Lancashire, England). These samples were cured (LED, DEML, SDS Kerr, USA, with an intensity of circa 800 mW cm-2) in 25mm * 2 mm molds between two sheets of polyester and flexural strength values were obtained from the following equation:

$$BS=3PL/2BD2$$

In which P is the load at the breaking point, L is the bearing spacing, B is the width and D is the sample thickness. In the bending test, the speed of the jaw was 0.5 mm/min and the maximum load was 1KN, and repeated for five times for each sample.

2.3. Diametric tensile strength test

The modified composites (according to ANSI/ADA specification n.27 for light cure resin) [27] were cured in cylindrical molds with 6 mm thickness and 4 mm diameter between two glass plates by the light cure device for 20 seconds, and the diametric tensile strength of samples was calculated by a testometric device and the equation below: $DTs=2p/\pi dl$

In which P is the load at the breaking point, D is the diameter of the sample and I related to its height. The speed of the jaw was 10mm/min with the maximum load 2KN [28].

2.3. Compressive strength test

The compressive strength test of modified composites was performed using the 4049 standard [26]. Samples in cylindrical molds with dimensions of 4 * 6 mm were cured and then subjected to compressive strength test with a testometric device and obtained in term of the peak and the diameter of samples [25].

2.3. Determine depth of cure

In order to determine the depth of cure, the samples were transferred to steel molds of 4 *10 mm and polymerized by the light cure device in over of molds for 40 seconds. The composites were removed from the molds and separated uncured composite by using a spatula. The height of each cured sample was measured and divided by two in order to obtain the depth of the cure [25].

2.3. Water sorption test

Samples were transferred to cylindrical molds with a diameter of 15 mm and a thickness of 1 mm which cured in accordance with ISO4049. The samples were transferred to an incubator in 37 ° C until their weight become fixed. Sample volume V (mm3) was measured and then placed in 60 ml of distilled water at 37 ° C for one week. After this time, the samples were brought out from the water and the excess water were removed. The samples were weighted separately (m2) and dried again, then reweighted (m3). Water sorption data were obtained from the following equation [29].

Water sorption = (m2-m3)/V

These values were compared with the control composite samples and the water sorption test repeated five times for each group.

2.3 Statistical data analysis:

Data were analyzed by one-way ANOVA, and the Tukey post hoc HSD multiple comparison test. The level of significance was determined as $p = 0.05$.

3. Results

3.1. FTIR:

The results of the FTIR for Cloisite and modified Cloisite are shown in Fig.1, as it can be seen in the IR spectra of modified Cloisite, two peaks have been appeared in 1713 Cm-1 and 2975 Cm-1.

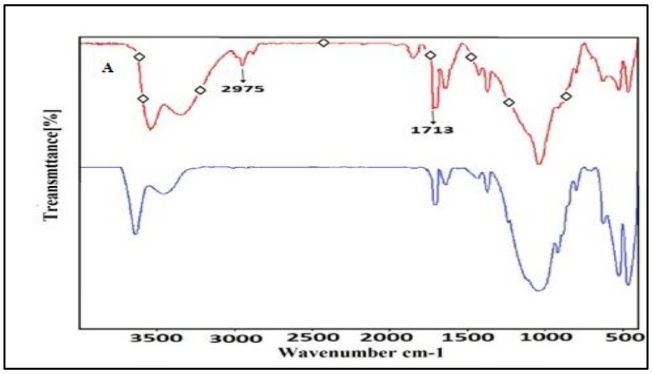


Fig.1. IR spectroscopy Cloisite (-) and modified Cloisite ( )

3.2. TGA:

The results of TGA were shown in fig. 2, elucidate the change in the thermal behavior of the modified Cloisite compared to pure Cloisite, as another factor to confirm the presence of glycidyl methacrylate in the Cloisite structure.

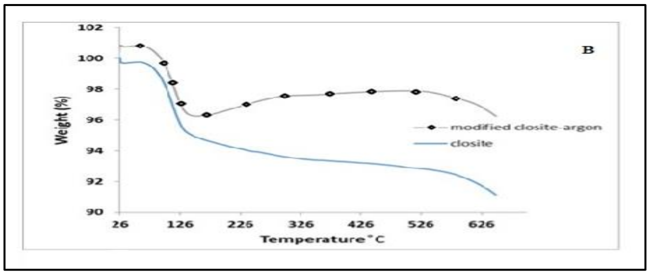
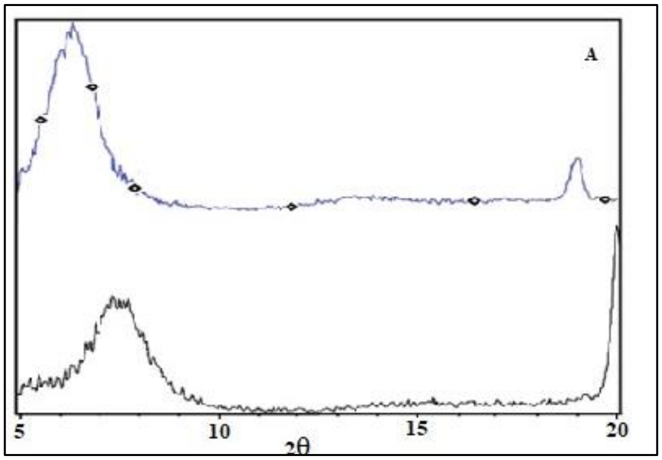


Fig. 2. TGA of Cloisite (-) and modified Cloisite ( )



3.3. XRD:

X-ray Diffraction (XRD) patterns of Cloisite and modified Cloisite has been shown in fig. 3. The peak has been shifted toward lower angles due to increase distance between clay plates.

Fig. 3: XRD of Cloisite (-) and modified Cloisite (-)

3.4. TEM:

The distance between the plates of Cloisite and modified Cloisite were confirmed by the passage of light from the general plates in the TEM which is shown in fig. 4.

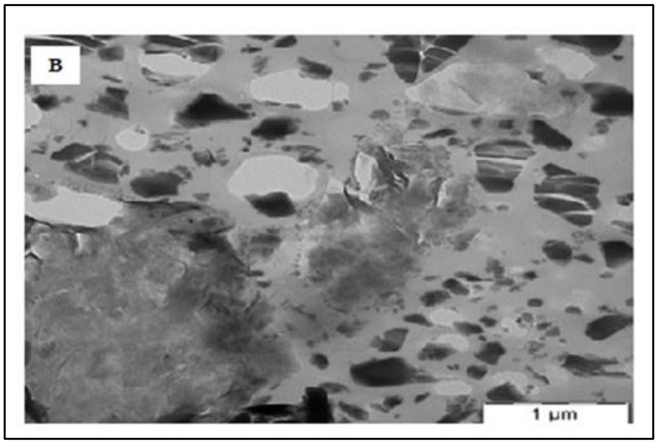


Fig. 4: TEM of composite contaning modified Cloisite

3.5. Flexural strength:

The average flexural strength results are presented in Fig. 5. As shown, with increasing the percentage of modified clay in the composite, flexural strength is changed and with increasing 0.2%, the greatest increase is observed in flexural strength. Which has a significant difference with the control group (composites with 0% of modified clay). ($p < 0.05$). There was no significant difference between the other groups.

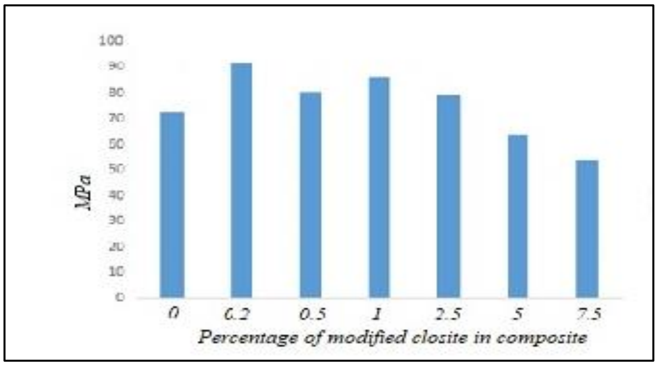


Fig. 5: Flexural strength

C3.6. Flexural modulus:

The average results of the flexural modulus are reported in Fig. 6, show a significant increase in the composite modulus containing 0.2 and 0.5% modified clay. ($p < 0.05$) there was no significant difference between the other groups

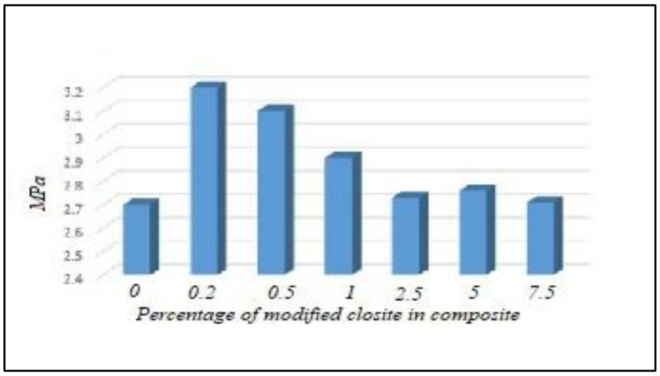


Fig. 6: Flexural modulus

3.7. Compressive strength:

The mean of compressive strength results are shown in fig 7, as shown, there is no significant difference between groups ($p > 0.05$).

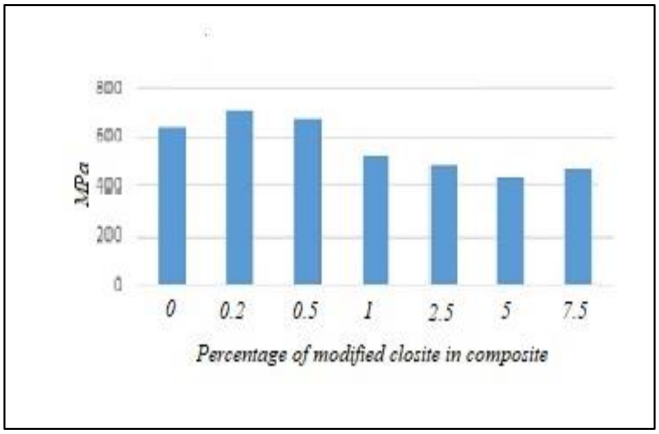


Fig. 7: Compressive strength

3.8. Diametral tensile strength:

The mean of diametral tensile strength (fig. 8) indicated a significant increase in composites containing 0.2% clay modified Cloisite with control group ($p < 0.05$). However, in 2.5 and 5% groups, showed a significant reduction with the control groups. There was no significant difference between the other samples

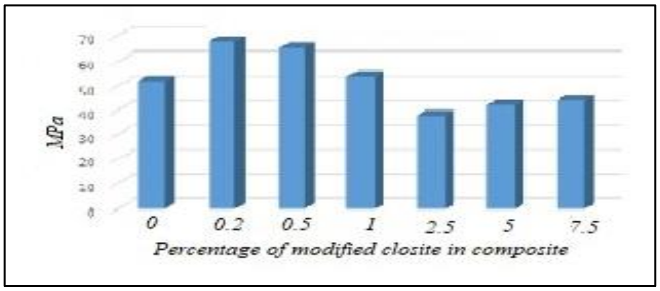


Fig. 8: Diametral tensile strength

3.9. Depth of Cure:

Depth of cure results, as shown in Fig. 9, did not show any significant difference between the control group and the groups containing 0.2 and 0.5% modified Cloisite ($P > 0.05$). There is a significant difference between other groups ($P < 0.05$).

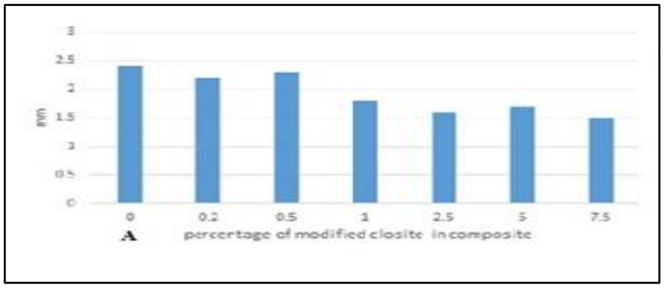


Fig. 9: Depth of cure

3.10. Water sorption:

As the modified composite increases, the absorption of water increases as well (Fig. 10). However, this difference was not significant between composites containing 0.2, 0.5, 1 and control group. ($P > 0.05$)

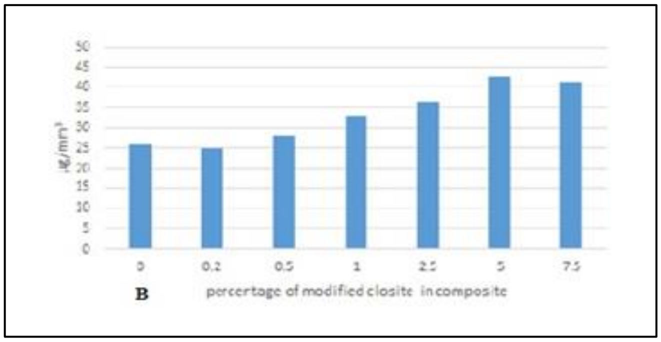


Fig. 10: Water sorption

3. Discussion

In this study, Cloisite was modified with glycidyl methacrylate and used to increase the mechanical properties of flowable dental composites. An important aspect of this study is the increasing strong bond between dental resin and Cloisite. Results of FTIR for Cloisite and modified Cloisite are shown in Fig.1. In this spectrum two peaks were appeared in the areas 1713 cm-1 and 2975 cm-1 which were assigned to the stretching vibrations of carbonyl and C-H group in modified Cloisite structure, respectively which indicate the presence of glycidyl methacrylate in the Cloisite structure. The results of TGA were shown in fig 2, elucidate the change in the thermal behavior of the modified Cloisite compared to pure Cloisite, as another factor to confirm the presence of glycidyl methacrylate in the Cloisite structure. X-ray Diffraction (XRD) patterns of Cloisite and modified Cloisite has been shown in fig 2. The peak has been shifted toward lower angles due to increase distance between clay plates. By using of Bragg equation the distance between the plates of Cloisite and modified Cloisite were calculated and obtained 12.12A° and 14.12A° for Cloisite and modified Cloisite respectively, and was confirmed by the passage of light from the general plates in the TEM which is shown in fig 2. In addition this increase is due to the steric effect of glycidyl methacrylate at the edge of the plates and covalent bonding with hydroxyl groups in Cloisite. The results of average flexural strength are presented in Fig 3. As it can be seen that the composite flexural strength was changed with increasing the percentage of modified clay and therefore the largest increase observed in flexural strength with increasing 0.2%, of modified clay which has a significant difference with the control group (composites with 0% of modified clay). ($p < 0.05$). There was no significant difference between the other groups. The average results of flexural modulus were reported in Fig 3 which were shown a significant increase in composites containing 0.2 and 0.5% modified clay ($p < 0.05$) and there was no significant difference between the other groups. In fig 3 was shown the mean of compressive strength results with any significant difference between groups ($p > 0.05$). The mean of diametral tensile strength indicated a significant increase in composites containing 0.2% of modified Cloisite with control group ($p < 0.05$) (Fig. 3). However, in the groups of 2.5 and 5%, modified Cloisite were shown a significant reduction with the control groups.

Depth of Cure was calculated based on the results which are shown in Figure 4, without any significant difference between the control group and the groups of 0.2 and 0.5% modified Cloisite. ($P > 0.05$). As by increasing amount of modified Cloisite in the composites, water sorption increased (Fig. 4). However, this difference was not significant between composites with 0.2, 0.5 and 1 weight percentage of modified Cloisite and control group. ($P > 0.05$)

Recent studies show that natural clays contains hydrophilic groups and are naturally prone to absorb water. In the superabsorbent field, much attention has been paid to clay for the preparation of superabsorbent materials [30-32]. This hydrophilic behavior of composites belong on chemical characteristics of individual components, but is mainly addressed to the organic matrix and/or to clay presence. In this study, the presence of clay caused higher water sorption levels but according to ISO 4049 standard water sorption below 40 μg/mm3 is acceptable for dental restorative materials [33]. Several studies regarding experimental resin composites with montmorillonite clay [34] reveal similar results. Water uptake in dental resin composites may cause hydrolytic degradation of matrix and filler/matrix junction and finally cause a decrease in mechanical properties [34].

The presence of 0.2% modified Cloisite in flowable dental composites along with increasing flexural strength and modulus and so diametral tensile strength. These results are consisted with the results which have been reported by LR Menezes and D. Parra [35, 36]. The mechanical and physical properties of this modified composite (0.2%) have the highest values[37]. Atai and et al [38] have studied the effect of nanoclay on poly methacrylic acid and they were observed 0.5 was the best nanoclay percentage for improving flexural properties. This difference may be due to the presence of glycidyl methacrylate in Cloisite structure and bonding with the resin. In this research, flexural and tensile strength of flowable dental composites were improved without any change at the compressive strength, so in our future research we will try to improve this effect on these composites. Attention to design, produce and characterize new resin-based composites that would apply better and are safer for use, we suggest experimental composites focused on clay addition. The aim of this study was to assess the mechanical and physical characteristics of this type of material encouraging further studies

3. Conclusion

The modification of Cloisite was successfully done by a simple method using glycidyl methacrylate. Addition of 0.2% of modified Cloisite to flowable dental composites cause to increase the flexural strength and modulus and so the tensile strength of flowable dental composites without significant effect in the depth of cure and their water sorption. Addition of this certain additive can be caused an effective strategy on the development of the flowable dental composites.

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Conflict of Interest

The authors declare no conflicts of interest.

Acknowledgments

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Data Availability

The data used to support the findings of this study can be made available upon request to the corresponding authors.

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