SYNTHESIS AND CHARACTERIZATIONS OF NOVEL MISWAK POWDER-BASED DENTAL COMPOSITES

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This study aimed to synthesize a novel miswak based dental composite by incorporating miswak powder, chlorhexidine (CHX), and silica fillers in different proportions into a resinous mixture and assess the influence of these fillers on the physical, mechanical, and biological properties of newly developed composites. The stock monomer solution was synthesized using urethane dimethacrylate, triethylene glycol dimethacrylate, and hydroxyethyl methacrylate. Four experimental groups (A, B, C, D) were prepared using 30% stock monomer with 70% filler formulations, while two commercial composites were used as control groups (E, F). The highest value of degree of conversion was shown by Group D, whereas the lowest – by Group A. Compressive strength evaluation showed Group E had the highest value, while the lowest value was recorded for Group B. Furthermore, cytotoxicity assessment showed that all the groups of composites had a biocompatible nature, except Groups A and B, having slight cytotoxicity. Thus, the experimental groups can be used as restorative materials as they exhibited optimum properties.

Keywords: resin based dental composites, miswak, chlorhexidine, silica filler

INTRODUCTION

Tooth decay or dental caries is a chronic infectious disease of dental hard tissues. It is among the most serious and common oral diseases in the world. Over time, a complex interaction involving acid-producing bacteria, fermentable carbohydrates, and host components, such as saliva and teeth, leads to the development of this condition.[1] It causes demineralization of dental hard tissues and results in cavitation in the tooth. Several dental restorative materials, such as amalgam, glass ionomer cement, resin-modified glass ionomer cement and composites, are being developed to combat the caries process.[2]

Resin based dental composites (RBCs) have become popular materials for restoring decayed as well as damaged teeth. Their increased demand is due to superior aesthetics, direct-restorative capability, and better clinical performance.[3] Extensive research has led to significant advancements in resin compositions, composite fillers, polymerization properties, and manipulative techniques. However, RBCs have various inherent problems, which include high coefficient of thermal expansion, polymerization shrinkage, and low mechanical properties.[4] Besides this, failure from bulk fractures and recurrent caries also poses challenges to clinical durability.[5] Therefore, current studies are increasingly focused on the fabrication of RBCs with antimicrobial additives.

Various leachable antimicrobial additives, such as antibiotics, fluoride, silver, chitosan, and nanoparticles, have been incorporated into RBCs.[6] This approach works by releasing antimicrobial agents into the oral environment. Chlorhexidine diacetate (CHX) is used as an organic antimicrobial additive in dentistry due to its low toxicity and widespread activity against oral pathogens. It is also considered a gold standard for assessing antimicrobial additives.[7] The

incorporation of CHX into direct restorative materials can prevent microbial leakage at tooth-restoration interface, which may cause recurrent caries. However, the incorporation of CHX in RBCs has been reported to have a detrimental effect on mechanical and physical properties.[8]

The miswak stick is commonly used in Muslim countries for oral health care and is known as a natural toothbrush. It has also been endorsed by the World Health Organization for oral hygiene due to its antimicrobial properties.[9] Cellulose is one of the major constituents of miswak stick.[10] It has various applications in the field of biomaterials, filtration membranes and regenerative medicine due to its superior mechanical properties. Therefore, it can be considered as a potential reinforcing agent in the field of dentistry.[11] Studies have shown that the use of mouth washes containing miswak extract had a significant reduction in both gingival bleeding and gingival inflammation, and has a protective action against bacterial species like *Streptococcus mutants* and some periodontal pathogens like *Bacteroides* species.[12] Moreover, its incorporation into restorative materials, such as heat-cure acrylic resins[13] and glass ionomers cement,[14] has been evaluated for enhanced mechanical and antimicrobial properties.

Therefore, the aim of this study was to design a novel miswak based dental composite and investigate its mechanical, chemical and biological properties.

EXPERIMENTAL

Materials

This was an *in vitro* experimental study. The novel dimethacrylate composites were based on fillers, including silica, chlorhexidine (CHX) and miswak (*Salvadora perscia*). The silica and CHX were obtained from Sigma Aldrich, USA, while miswak was bought from a local market. The experimental monomers [urethane dimethacrylate (UDMA), triethylene glycol dimethacrylate (TEGDMA), 2-hydroxyl ethyl methacrylate (HEMA)], initiators [camphoroquinone (CQ)], co-initiators [N, N; dimethyl p-toluidine (DMPT)] were also purchased from Sigma Aldrich, USA. The samples complying with ISO standards *i.e*., ISO 4049 and ISO 10933, were included into the tests, whereas samples with broken and wear surfaces were excluded from the study. For control groups, commercially available RBCs [Filtek ™ Z250 (3M ESPE, USA) and NEXCOMP (META BIOMED)] were used.

Preparation of miswak powder and salinization of fillers

Miswak sticks were cut into small pieces with the help of a sharp knife. Then, these small pieces were put into an electrical grinder. The large sized particles obtained from the electrical grinder were sieved through different types of sieves (sizes: 0.1-5 mm) in a sieve shaker, at a voltage of 230 V and frequency of 50 rpm, for about 15-20 minutes. The small sized particles obtained from sieve shaker were placed into a ball milling machine for 3-4 h at a frequency of 300 rpm to get fine grade miswak powder. This powder was then salinized according to the protocol described in the literature.[15] The silica powder was also salinized in a similar manner.

Preparation of stock monomer

The stock monomer was prepared with 68% UDMA, 25% TEGDMA, 5% HEMA, 1% CQ and 1% DMPT. All these chemicals were added one by one in a brown colored bottle and covered with aluminum foil to avoid premature polymerization. The mixture was stirred with a magnetic stirrer for 30 min at 70-80 rpm.

Preparation of experimental composites

The experimental composites were synthesized by adding various fillers in different proportions into the prepared stock monomer using 30% stock monomer with 70% filler formulations. Table 1 shows the groups of experimental composites along with their filler formulations.

Table 1
Experimental and control dental composites groups along with filler formulations

Experimental group	Filler formulation
A	95% silicate fillers + 5% chlorhexidine powder
B	95% silicate fillers + 5% miswak powder
C	90% silicate fillers + 5% chlorhexidine powder + 5% miswak powder
D	100% silicate fillers
E (Commercial control) (3M Z250)	Silane treated silica 1-10% + silane treated ceramics 65-90%
F (Commercial control) (Nexcomp)	Silane treated borosilicate particles 54%

Preparation of samples

For compressive strength testing, Teflon molds of 6 mm × 4 mm were used, whereas for SEM, FTIR and other tests, brass molds of 8 mm × 2 mm were used. Samples were prepared in clean molds placed on clean and smooth glass slabs. The composite material of each group was poured carefully inside the molds, covered with transparent cellulose acetate on both sides to prevent entry of oxygen. This method has shown promising results to prevent formation of an oxygen-inhibited layer on the surface of composites.[16] Then, the material was cured on both sides at a constant distance of 1 mm for 60 seconds using a light curing device (LED, Woodpecker, wavelength 470 nm). The samples were then removed with caution from the molds, polished with 800, 1000, 1200 and 2000 grit papers, and processed for further characterizations.

Characterizations

Fourier transform infrared spectroscopy (FTIR)

Two samples from each group of composites were subjected to FTIR both before and after curing. An Attenuated Total Reflectance (ATR) accessory was used for FTIR (Thermo Nicolet 6700, USA) to assess the degree of conversion and structural changes. A background scan was obtained prior to each set of tests. Each sample was placed on the clean diamond window of the FTIR equipment to collect spectra over the region 4000-400 cm^{-1} at a resolution of 8 cm^{-1}, averaging 256 scans. The data were interpreted using OMINIC software and peaks were matched based on the literature.[17]

Degree of conversion (DC)

The freshly made unpolymerized and polymerized samples of each group were subjected to FTIR to determine the DC. The samples were cured for 60 seconds on both sides with a light curing lamp. The DC of two samples from each composite group was calculated by using the following formula (1):

DC% = 100 × [1 - (R polymerized/R unpolymerized)]

where DC stands for degree of conversion and R denotes the ratio of peak height of polymerized aliphatic to polymerized aromatic and unpolymerized aliphatic to unpolymerized aromatic groups of samples. DC was determined by assessing changes in the ratio of the absorbance intensities of aliphatic C=C peak at 1638 cm^{-1} and that of an aromatic C=C at 1608 cm^{-1} of the uncured and cured samples.[18]

Scanning electron microscopy (SEM)

Firstly, composite discs from each respective group were polished with grit papers in ascending order to obtain a smooth and homogenous surface, devoid of any scratches. The samples were then gold coated in a sputter coater (Quorum) and images were taken in the range of 25X to 500X, with voltage kept at 15 kv. Then, surface morphology was analyzed using a scanning electron microscope (Tescan Vega-3 LMU, Czeck Republic).[19]

Compressive strength (CS)

Prepared cylindrical samples (6 mm x 4 mm) were placed in separate glass vials containing fresh distilled water. Then, the samples were removed from the respective vials and dried with blotting paper. After this, the samples were kept in a dry heat oven at 370 °C for 24 h. Each sample was then placed on the platens of a universal testing machine (UTM, Testometrics United Kingdom) for loading it to failure in compression at a crosshead speed of 1.0 mm/min. CS was calculated on the basis of peak load and diameter of the sample. For CS, the following formula was used:

$$ucs = 4f/\pi d^2 \quad (2)$$

where ucs is ultimate compressive strength (MPa), f is maximum load (N), and d is cylindrical specimen diameter (mm).[20]

Cytotoxicity

On composite samples, fibroblast cell lines (NIH/3T3 ATCC® CRL-1658TM) were utilized for cell cultures. The media were prepared using Dulbecco's Modified Eagle's Medium (DMEM; ThermoFisher Scientific, USA), to which 10% fetal bovine serum (Sigma Aldrich, USA) and 100 µg/mL Pencillin/Streptomycin (Sigma Aldrich, Life Sciences, USA) were added. Then, a T75 culture flask (Corning Biosystem) was used for expanding NIH3T3 cells, followed by keeping in a humidified incubator containing 5% CO_2 at a temperature of 37 °C. Cells were grown to 90% confluency, and the medium was replaced every two to three days. After this, the cell detachment was done with trypsin–EDTA (Sigma Aldrich, USA). [21] Cell counting was done with a microscope and a hematocytometer on the day of seeding. The samples were washed with 70% alcohol for 24 hours, and then washed three times with 1x Phosphate Buffer Saline at a 15-minute interval. Then, a total of 50,000 cells were seeded on each sample in the 24 well plate for checking the compatibility of NIH3T3 cells with the samples. After that, 1 mg of composite sample was added into 1 mL of DMEM media. To compare the values with the cells produced in the presence of composite materials, the cells were also grown on tissue culture plastic plates without samples to serve as control.

Alamar Blue assay

The cell biocompatibility with the composite samples was assessed by taking readings with a fluorescent plate reader from the Alamar Blue assay after 3 days. Alamar Blue has a redox indication depending upon cellular metabolic activity that varies as the cells absorbed the substrate, from an oxidized

(blue) form to a reduced (red) form. Cells were seeded in 0.5 mL of 1 mM solution of Alamar Blue (Sigma Aldrich, UK) and were incubated for 3 to 4 hours at 37 °C. The PR4100 Absorbance Microplate Reader BIO RAD, UK, was used at the absorbance of 570 nm to measure the fluorescence.[22]

RESULTS AND DISCUSSION

Fourier transform infrared spectroscopy (FTIR)

FTIR was performed to evaluate the DC of various groups. Figure 1a shows the FTIR peaks of all six dental composite groups before curing, and Figure 1b – after curing. Different peaks in the curves show specific functional groups of the constituents of monomers found in RBCs. One of the most important peaks that can be seen at 1711 cm^{-1} (sharp and intense in the case of UDMA) corresponds to the stretching of C=O group in the monomers. The peaks at 1636 and 1400 cm^{-1} correspond to the C=C stretching and C-H group attached to C=C, respectively, in unpolymerized methacrylate of monomers. An UDMA specific peak due to N-H deformation group can be identified at 1509 cm^{-1}. A peak at 1034 cm^{-1} corresponds to the Si-O-Si group, which is mainly due to the silicate groups of some ingredients in the composites, whereas the peak at 1106 cm^{-1} corresponds to the stretching of the C-O-C group. A broad band in the regions of 2800 to 3700 cm^{-1} is due to the contribution of silicate filler particles in the composites.[23] The bands in the regions of 2859 to 2934 cm^{-1} show the contribution of CHX particles in the composite. The peaks in the regions of 3000 cm^{-1} to 3600 cm^{-1} are mainly due to the stretching of O-H and $-NH_2$ functional groups, while the peak at 1005 cm^{-1} is due to the stretching of SO^{2-}_4. All these peaks indicate the presence of miswak powder in the composite.[24]

DC is a significant parameter for predicting the final physical, mechanical and biological behavior of dental composites,[25] and FTIR is one of the extensively used techniques for studying the DC of RBCs. The exact value of DC for optimal intraoral performance of dental composite restoration has not been established yet. However, values ranging between 55-65% are considered for adequate clinical performance, whereas values of DC in the range of 35-77% are also reported in the literature.[26] Generally, the properties of RBCs are enhanced with increased DC. Besides this, a reduction in DC may result in elution of residual unreacted monomers from intraoral dental composites, which in turn may have the potential to trigger irritation, inflammatory response, or cytotoxic effects in the oral cavity.[27] The residual unreacted monomer from dental composite restoration may serve as a plasticizer and may decrease the mechanical behavior of the restoration.[28]

Figure 1b shows changes in the spectra after curing with light for 60 seconds. In the spectra, the peaks in the regions of 1636 cm^{-1} and 1606 cm^{-1}, which correspond to aliphatic and aromatic (C=C) groups of methacrylate-based monomers, respectively, decreased in intensity, which in turn shows the conversion of C=C to C-C groups (DC). A statistically significant difference (p-value ≤ 0.05) was observed among the DC of all composite groups for Group D (78%); likewise, Group D possessed higher DC after polymerization, which is better illustrated in Figure 2. The value of the degree of conversion (DC) of the commercial dental composite in Group E (Z250) was 60%, which is approximately in the same range as previously mentioned in the literature.[29] Also, the DC value of the other commercial composite in Group F (Nexcomp) is in the range of 40%.[30] Meanwhile, the experimental dental composite groups showed degree of conversion (DC) values in the range of 38-78%.

The higher values of DC for experimental composites may be due to the use of UDMA as base monomer, instead of *bis*-GMA in commercial dental composites. *bis*-GMA has higher viscosity, in the range of 600-1000 Pa.s, which is due to its increased intermolecular hydrogen bonding interactions, inherent aromatic structure of the molecule, and higher molecular weight (MW) value – of 512 g/mol.[31] Another reason for the lower value of DC in the commercial dental composite groups may be a mismatch in the refractive index value between *bis*-GMA (1.55) and silica particles (1.47).[32] A reason for using UDMA as base monomer in the experimental dental groups may be its lower value of viscosity – 23 Pa.s, inherent aliphatic molecular structure with flexible spacer group, which increases the mobility of the monomer, and lower molecular weight (MW) – 470.6 g/gmol.[33] Group D in the experimental dental composites has the highest DC – of 78%. The reason for this high DC value may be attributed to the close similarity in the refractive index of silica particles (1.47) and that of the base monomer *i.e*., UDMA (1.48).[34]

The lowest value of DC among experimental dental composites was 38%, which may be due to greater mismatch of the refractive index between chlorhexidine diacetate (CHX) (1.66),[35] and UDMA (1.48). Group B also showed a DC value in range of 44%, which may be due to a slight mismatch in the refractive index of miswak powder (1.33),[36] with UDMA (1.48); while Group C showed an intermediate value, of 60%. Another reason for the lower DC between Groups A and B might be the incompatibility of the initiator to absorb photons of visible light, producing an insufficient number of free radicals to initiate the polymerization process and thus polymerization might not have been completed properly.

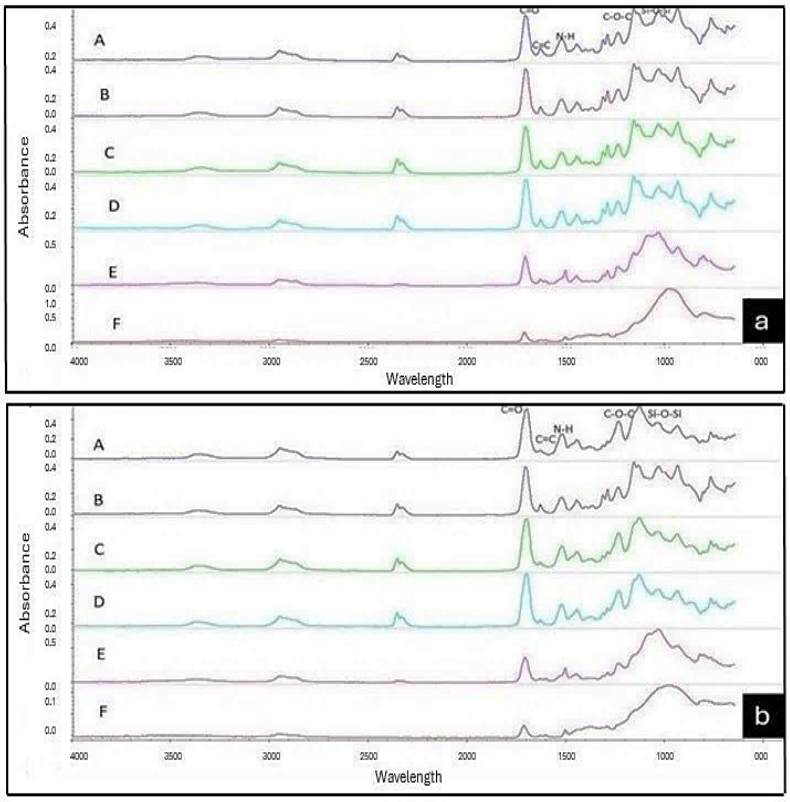

Figure 1: FTIR spectra of all dental composite groups: a) before curing, and b) after curing

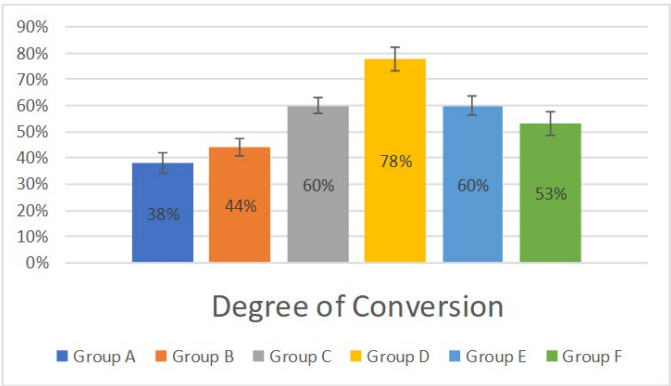

Figure 2: Degree of conversion (DC) among different groups of dental composites

Scanning electron microscopy (SEM)

SEM images of all six dental composite groups (A, B, C, D, E, F) at the selected magnification power are given in Figure 3. These SEM images show that all surfaces were free of cracks and were smoothly polished. Some pits could be seen in the micrographs, which may be caused by the incorporation of air during the manipulation process, however, their number was insignificant and insufficient to produce large

changes in SEM results. Group A composites showed haphazard distribution of silica and chlorhexidine (CHX) filler particles in the form of agglomerates against a dark background made up of the matrix consisting of dimethacrylate based monomer. Group B composites showed homogenous distribution of silica filler particles and heterogeneous distribution of miswak powder filler particles in the form of large irregular chunks against a background of dimethacrylate based monomers. Group C composite showed non-homogeneous distribution of silica, chlorhexidine (CHX) and miswak powder in the shape of cloud like agglomerates against a matrix of dimethacrylate based monomers. Group D composites showed random distribution of both fine and coarse shaped silica filler particles against a dark background matrix of its constituent resinous materials. Group E (Z250) showed the size of silane treated silica particles in the range of 5-50 µm. The SEM image showed both regular and irregular distribution of its compositional filler particles in the form of compacted agglomerates. Moreover, it was observed that the distribution of silica particles is not the same in various groups of composites because of the variable percentage of silica and other fillers, such as CHX and miswak.

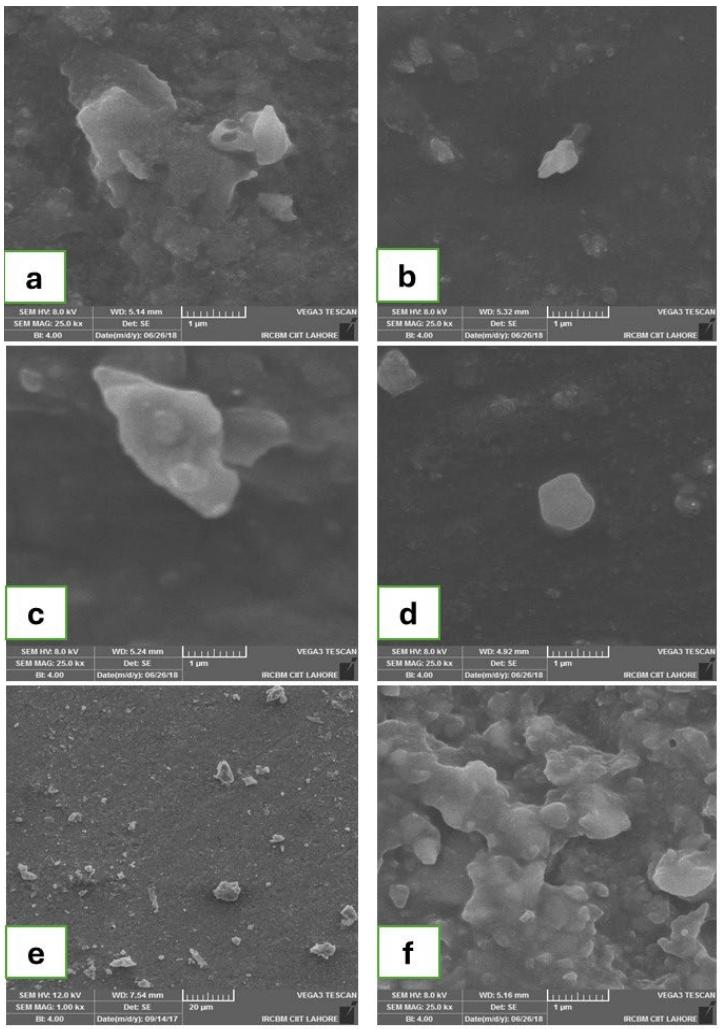

Figure 3: SEM images of all dental composites; a) Group A, b) Group B, c) Group C, d) Group D, e) Group E, and f) Group F

Compressive strength (CS) and compressive modulus (CM)

Compressive strength is another very important property in dental composite restorations as most of the intra-oral masticatory forces are compressive in nature. The CS also provides compressive modulus, which is a significant mechanical parameter and illustrates the rigidity of the material, which is important as different moduli of elasticity are required in

particular clinical situations.[37] The mean CS and CM values of six composite groups, along with their standard deviation values, are given in Table 2. The highest mean CS and CM were shown by Group E, whereas the least compressive strength was recorded for Group B. Statistical analysis showed Group E and Group B presented significant difference from the other groups. Statistical difference was also observed when comparing the experimental group (Group B) with other experimental groups: Group A, Group C and Group D. The difference was not significant between Group A and Group D.

The compressive strength reported for the commercial groups was in the range of 217.37 MPa-248.45 MPa, whereas the compressive modulus was 3.59 GPa-4.19 GPa. The compressive strength and modulus values for composite Z250 are in the same range as previously mentioned in the literature,[38] while the value reported for commercial composite Nexcomp was also in agreement with the one previously reported in the literature. The CS of the experimental dental composites was between 87.60 MPa and 156.52 MPa, while the CM values were in the range of 1.69 GPa-2.68 GPa. The higher values of CS and CM for commercial dental composites may be due to use of *bis*-GMA as a base monomer, which has a high elastic modulus, of 1427 MPa, in contrast to UDMA as base monomer in experimental groups, having the elastic modulus of 1405 MPa.[39] Another reason for the difference in CS and CM values between commercial and experimental groups lies in the filler loading: commercial groups having a loading in the range of 60-90%, while experimental groups having a loading of 70%. Among experimental groups, both Groups A and D showed statistically insignificant values *i.e.*, 156.52 MPa ± 7.01 and 153.35 MPa ± 6.69, respectively.

Groups B and C showed lower CS and CM values than other experimental groups. The reason behind this lies in the use of 5% miswak powder as filler constituent, which decreases the compressive strength value of these composites. Secondly, miswak powder is hydrophilic, porous in nature, and has less resistance to moisture.[40] The absorbed water acts as plasticizer,[41] which in turn causes a decrease in compressive strength. Also, the light brownish color of miswak powder might not allow adequate penetration of light during the light curing process, which in turn decreases the compressive strength. The miswak powder was produced by lab ball milling, in contrast to the aerosol production of fillers, which may lead to the heterogeneous distribution of miswak powder in composites, which obviously decreases their compressive strength. The heterogeneous distribution of miswak is quite evident in SEM images, as discussed above. In the case of commercial dental composites, there is vacuum mixing of filler particles with constituent monomers under controlled conditions, which produces uniform distribution of the fillers, with less void formation; also, vacuum mixing produces perfect alignment of filler particles in the direction of masticatory forces. In the case of our experimental dental composites, room temperature mixing was performed under varied conditions, which might lead to the formation of voids. Also, in the experimental dental composites, there is non-uniform distribution of filler particles because of the incompatibly among different filler particles, which in turn decreases compressive strength.

Table 2
Mean compressive strength (CS) and compressive modulus (CM) values of all composite groups, along with their standard deviation (SD) values

Groups	Mean compressive strength (MPa) and SD	Mean compressive modulus (GPa) and SD	P value
A	156.52 ± 7.01	2.68 ± 0.60	≤ 0.05
B	87.60 ± 11.23	1.69 ± 0.77	
C	140.16 ± 6.65	2.01 ± 0.47	
D	153.35 ± 6.69	2.11 ± 0.50	
E	248.45 ± 14.95	4.19 ± 1.09	
F	217.37 ± 18.05	3.59 ± 0.56	

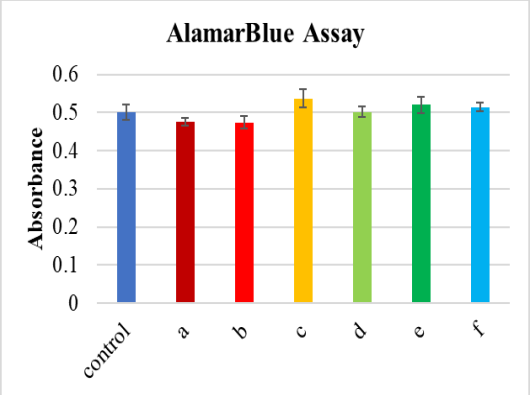

Figure 4: Evaluation of Alamar Blue assay for all the composite groups

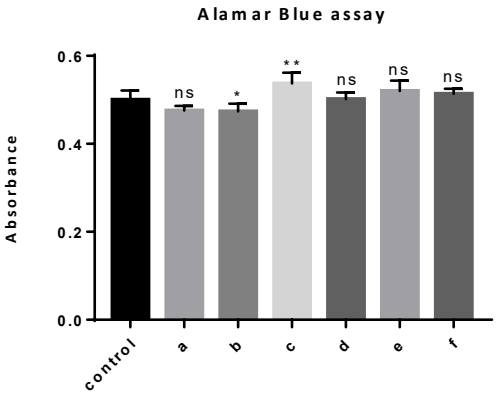

Figure 5: Statistical analysis of different dental composite groups as compared to control groups

Cytotoxicity

Figure 4 shows a comparison between the results of the control and of the experimental groups after 3 days of the cytotoxicity essay. The proliferation of cells increased over time for composite groups C, D, E and F, as compared to the control, which was shown as an increased absorbance rate. Among all the experimental composites, Group C exhibited the best results. However, all the groups demonstrated compatible and non-toxic behavior towards the living cells.

Each of the experimental groups provided five readings, thus every analysis was done at least twice to obtain accurate results. The statistical analysis of several composite groups in relation to the control is displayed in Figure 5. Apart from Groups B and F, which have p-values less than α, or <0.05, indicating a significant difference between the means of the composite groups, all groups in the graph have p-values larger than α, or >0.05, indicating that the difference between the means of the composite groups is not significant (ns) (* and **).

Cytotoxicity testing of dental composite groups was performed according to specifications mentioned in ISO-10933-5. To assess cytotoxicity, fibroblast cells were used because these are the predominant type of cells in human pulpal tissue, which get in contact with dental composite restorations in the oral cavity. The biocompatibility of a dental composite restoration depends on the type, its nature and on the amount of leachable monomer from its resinous matrix network and many adverse reactions caused by it due to an incomplete polymerization process.[42] The cytotoxicity is also influenced by dentine diffusivity and residual dentine thickness. The different cytotoxicity behavior exhibited by different results of both experimental and commercial groups might be influenced by their different compositions. The results of cytotoxicity of different composite groups might rely on the cytotoxic effects of their leachable monomers released from their organic polymeric organic matrix and might not depend on its filler matrix, *i.e.*, inorganic filler particles.

The results for the commercial composite groups showed an increase in cell proliferation, in contrast to the control cell population, as these composites had minimal organic component in their composition as compared to the filler percentage. Although these sample groups release Bisphenol-A (BPA), which is a by-product of *bis*-GMA, these BPA levels showed a peak value 3 hours after the composite restoration placement and returned to the baseline value in 24 hours. However, cytotoxicity reading was done on day 3, not on day 1, due to which, on day 3, both these groups showed an increase in cell proliferation, as compared to the control group. In the case of the experimental composite groups, Groups A and B showed a decreased cell proliferation rate, as compared to the control, as well as to other experimental composites – Groups C and D. The reason behind this may be a lower degree of conversion in both groups *i.e.*, in the range of 38-44%, due to which considerable amounts of UDMA, TEGDMA and HEMA were released from the composites and exerted cytotoxic effects on pulpal cells. The decrease in cell proliferation among these groups may be caused by the synergistic cytotoxic effect of both UDMA and TEGDMA, as previously reported in the literature; while HEMA reported less toxicity, as compared to both UDMA and TEGDMA, owing to its lower molecular weight. Moreover, Groups C and D showed an increase in the cell proliferation rate due to their high value of the

degree of conversion, as compared to Groups A and B.

CONCLUSION

The present study aimed to develop a novel dental composite material by integrating a complex filler mixture comprising miswak powder, chlorhexidine, and silica fillers in different proportions, into a resinous mixture that included urethane dimethacrylate, triethylene glycol dimethacrylate, and hydroxyethyl methacrylate. Then, mechanical, chemical and biological properties of the developed composites were investigated by appropriate techniques. Experimental dental composite formulations showed an improved degree of conversion, good fibroblastic cell biocompatibility and compromised mechanical properties in comparison with commercial composite formulations.

REFERENCES

[1] R. A. Giacaman, C. E. Fernández, C. Muñoz-Sandoval, S. León, N. García-Manríquez *et al*., *Front. Oral Health*, **3**, 764479 (2022), https://doi.org/10.3389/froh.2022.764479

[2] A. Slimani, S. Sauro, P. Gatón Hernández, S. Gurgan, L. S. Turkun *et al*., *Materials*, **14**, 6272 (2021), https://doi.org/10.3390/ma14216272

[3] A. S. Khan and M. R. Syed, *Dent. Mater*., **38**, 163 (2019), https://doi.org/10.4012/dmj.2018-039

[4] D. Bompolaki, E. B. Lubisich and A. P. Fugolin. *Dent. Clin*., **66**, 517 (2022), https://doi.org/10.1016/j.cden.2022.05.003

[5] M. Shahmoradi, B. Wan, Z. Zhang, M. Swain and Q. Li, *J. Mech. Behav. Biomed. Mater*., **125**, 104942 (2022), https://doi.org/10.1016/j.jmbbm.2021.104942

[6] S. Agarwalla, S. K. Singh, M. A. Ibrahim, H. Noothalapati and S. Duraiswamy, in "Functional Coatings", edited by R. K. Arya, G. D. Verros and J. P. Davim, John Wiley and Sons, 2024, pp. 231-274, https://doi.org/10.1002/9781394207305.ch9

[7] A. Priya, N. H. Prasath, N. Malligarjunan and S. K. Pandian, "Pharmacological Studies in Natural Oral Care", Scrivener Publishing LLC, 2023, pp. 723-773, https://doi.org/10.1002/9781394167197.ch40

[8] S. Ali, L. Sangi, N. Kumar, B. Kumar, Z. Khurshid *et al*., *Techn. Heal. Care*, **28**, 165 (2020), https://doi.org/10.3233/THC-181568

[9] A. Caiafa and L. Visser, "Restorative Dentistry", edited by H. B. Lobprise and R. Johnathon, 2019, pp. 357-386

[10] A. F. Nur Diyana, A. Khalina, M. S. Sali, C. H. Lee, H. A. Aisyah *et al*., *Phys. Sci. Rev*., **8**, 5089 (2023), https://doi.org/10.1515/psr-2022-0043

[11] L. Zhang, L. Guo and G. Wei, *Materials*, **14**, 5390 (2021), https://doi.org/10.3390/ma14185390

[12] F. A. Adam, N. Mohd, H. Rani, M. Yusof and B. Baharin, *J. Ethnopharm*., **302**, 115863 (2023), https://doi.org/10.1016/j.jep.2022.115863

[13] H. A.-R. Khalaf and M. J. Al-Fahaam, *IOSR J. Dent. Med. Sci*., **14**, 32 (2015), https://doi.org/10.9790/0853-14163234

[14] L. Singer, G. Bierbaum, K. Kehl and C. J. M. Bourauel, *Materials*, **13**, 5352 (2020), https://doi.org/10.3390/ma13235352

[15] F. Rehman, P. L. Volpe and C. J. Airoldi, *J. Environ. Manag*., **133**, 135 (2014), http://dx.doi.org/10.1016/j.jenvman.2013.11.042

[16] S. Hadi, R. Tjandrawinata and E. J. Fibryanto, *J. Int. Oral Health*, **16**, 150 (2024), https://doi.org/10.4103/jioh.jioh_198_23

[17] A. H. Delgado and A. M. Young. *Plos ONE*, **16**, e0252999 (2021), https://doi.org/10.1371/journal.pone.0252999

[18] A. L. S. Borges, A. M. Dal Piva, S. E. Moecke, R. C. de Morais and J. P. Tribst, *J. Compos. Sci*., **5**, 322 (2021), https://doi.org/10.3390/jcs5120322

[19] A. Butera, M. Pascadopoli, S. Gallo, M. Lelli, F. Tarterini *et al*., *Polymers*, **13**, 2740 (2021), https://doi.org/10.3390/polym13162740

[20] B. Pratap, R. K. Gupta, B. Bhardwaj and M. J. Nag, *Mater. Today Proc*., **33**, 2567 (2020), https://doi.org/10.1016/j.matpr.2019.12.142

[21] M. Sabir, N. Muhammad, U. Siddiqui, A. S. Khan, M. R. Syed *et al*., *Polym. Bull*., 1 (2022), https://doi.org/10.1007/s00289-022-04503-9

[22] M. Sabir, A. Ali, U. Siddiqui, N. Muhammad, A.S. Khan *et al*., *J. Biomater. Sci. Polym. Ed*., **31**, 1806 (2020), https://doi.org/10.1080/09205063.2020.1777827

[23] A. S. Khan, H. Khalid, Z. Sarfraz, M. Khan, J. Iqbal *et al*., *J. Appl. Spectr. Rev*., **52**, 507 (2017), https://doi.org/10.1080/05704928.2016.1244069

[24] R. Chaaben, R. Taktak, B. Mnif, N. Guermazi and K. Elleuch, *J. Therm. Comp. Mater*., **35**, 1815 (2022), https://doi.org/10.1177/0892705720939167

[25] D. Kaisarly and M. E. Gezawi, *Odontology*, **104**, 257 (2016), https://doi.org/10.1177/089270572093916710.1007/s10266-016-0264-3

[26] P. Yılmaz Atalı, B. Doğu Kaya, A. Manav Özen, B. Tarçın, A. A. Şenol *et al*., *Polymers*, **14**, 4987 (2022), https://doi.org/10.3390/polym14224987

[27] M. Moldovan, R. Balazsi, A. Soanca, A. Roman, C. Sarosi *et al*., *Materials*, **12**, 2109 (2019), https://doi.org/10.3390/ma12132109

[28] W. Potiprapanpong, W. Thepveera, C. Khamsuk, S. Channasanon, S. Tanodekaew *et al*., *Polymers*, **13**, 2742 (2021), https://doi.org/10.3390/polym13162742

[29] A. L. S. Borges, A. M. Dal Piva, S. E. Moecke, R. C. de Morais and J. P. M. Tribst, *J. Compos. Sci*., **5**, 322 (2021), https://doi.org/10.3390/jcs5120322

[30] S. Omer, A. Khan, M. Ejaz and M. Zeeshan, *J. Rehm. Coll. Dent*., **4**, 20 (2023), https://doi.org/10.52442/jrcd.v4i2.84

[31] L. Hong, Y. Wang, L. Wang, H. Zhang, H. Na *et al*., *J. Dent*., **59**, 11 (2017), https://doi.org/10.1016/j.jdent.2017.01.006
[32] D. L. Del Rio and W. M. Johnston, *J. Dent*., **118**, 103949 (2022), https://doi.org/10.1016/j.jdent.2022.103949
[33] A. Szczesio-Wlodarczyk, M. Domarecka, K. Kopacz, J. Sokolowski and K. Bociong, *Materials*, **14**, 2727 (2021), https://doi.org/10.3390/ma14112727
[34] S. B. Lone, R. Zeeshan, H. Khadim, M. A. Khan, A. S. Khan *et al*., *J. Mech. Behav. Biomed. Mater*., **151**, 106398 (2024), https://doi.org/10.1016/j.jmbbm.2024.106398
[35] A. Aljabo, W. Xia, S. Liaqat, M. Khan, J. Knowles *et al*., *Dent. Mater*., **31**, 1279 (2015), https://doi.org/10.1016/j.dental.2015.08.149
[36] N. Chelli-Chentouf, A. T. T. Meddah, C. Mullié, A. Aoues and B. J. Meddah, *J. Ethnopharm*., **144**, 57 (2012), https://doi.org/10.1016/j.jep.2012.08.025
[37] K. Cho, G. Rajan, P. Farrar, L. Prentice and B. G. Prusty, *Compos. B Eng*., **230**, 109495 (2022), https://doi.org/10.1016/j.compositesb.2021.109495
[38] M. Saleem, S. Zahid, S. Ghafoor, H. Khalid, H. Iqbal *et al*., *J. Appl. Polym. Sci*., **138**, 50336 (2021), https://doi.org/10.1002/app.50336
[39] I. M. Barszczewska-Rybarek, *Materials*, **12**, 4057 (2019), https://doi.org/10.3390/ma12244057
[40] A. F. Nur Diyana, A. Khalina, M. S. Sali, C. H. Lee, H. A. Aisyah *et al*., *Phys. Sci. Rev*., **8**, 5089 (2023), https://doi.org/10.1515/psr-2022-0043
[41] I. F. Ghazi, J. K. Oleiwi, S. I. Salih and M. A. Mutar, in *IOP Conf. Ser.*: *Mater. Sci. Eng*., 2021, https://10.1088/1757-899X/1094/1/012169
[42] E. Ohlsson, C. Bolay, S. Arabulan, K. M. Galler, W. Buchalla *et al*., *Dent. Mater*., (2024), https://doi.org/10.1016/j.dental.2024.02.015