

## PREPARATION AND CHARACTERIZATION OF NOVEL COMPOSITES INCLUDING BORON NITRIDE

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### ABSTRACT

Dental composites are commonly used as restorative materials in dentistry, which helps to maintain a healthy tooth structure. In this work, dental composites were prepared by blending hexagonal boron nitride nanoparticle (h-BN) with glycidyl methacrylate (GMA) / Triethylene glycol dimethacrylate (TEGDMA) based on modified nanoparticles as fillers. The surfaces of the hexagonal boron nitride nanoparticles were modified by glycidyl methacrylate (GMA). The prepared materials were characterized by analyzing their mechanical and physical properties. According to the mechanical results, the obtained nanocomposites have good mechanical stability. The h-BN as fillers were well dispersed in the organic matrix. The homogenous distribution of the inorganic fillers throughout the matrix was observed by SEM. The dental composites, prepared in this work, are promising materials and the results indicate that these materials, can be used as restorative materials in dentistry.

**Keywords:** dental composites, hexagonal boron nitride, surface modification, blend

### INTRODUCTION

Dental materials were started to be commonly used in the Dental amalgams for tooth restorations during the 7<sup>th</sup> century in China and 16<sup>th</sup> in Germany (Bharti, Wadhwani, Tikku, & Chandra, 2010). Amalgams, which are known as dental fillings, are an alloy mixed with Hg, Cu, Sn, and/or Ag. Evethough, in terms of longevity, amalgams show better results compared to composite resins, composite resins bond directly to tooth, which demands less drilling and offers a better seal (Shenoy, 2008). Additionally, pale color makes them blend into existing tooth, which make them more aesthetically appealing (Shenoy, 2008). Because of that, composite materials have started to replace Hg amalgam fillings. Resin-based composites used for restorative purposes contain fillers with high atomic mass elements such as barium, zirconium, strontium or ytterbium. Composite resins were first started to be used in the 1960s. At the beginning, the resin mixtures were not easily polished, but their durability were stronger than amalgam and they did not require mercury and silver to be used. Composite resins adhere to the tooth better and their color is very close to the existing teeth (Bowen, 1963).

Some metal nanoparticles (Ag, Au, Cu) can be used alone in dentistry or they are used after mixing with other agents due to their antimicrobial and antibacterial properties. The positively charged Ag ions provide antibacterial activity by creating electrostatic attraction on the negatively charged bacterial cell membrane (Kim et al., 2007) and Ag ions inhibit the respirator enzyme of the bacterium. As the results of this process, free radicals are formed and the cell membrane is damaged (Pal, Tak, & Song, 2007). Titanium dioxide is widely used as a photocatalytic and antimicrobial compound. For this purpose, this material can be used in dentistry. Although TiO<sub>2</sub> is non-toxic at the usage concentrations, some recent studies showed that this nanoparticle may cause various problems in dentistry (Blake et al., 2008).

In recent years, biocompatible polymers are commonly used in the dental processes. Chitosan is a biocompatible, hydrophilic and bio-absorbable natural polymer, and, therefore, it is especially used for root canal disinfection in endodontics (Lin, Skribner, & Gaengler, 1992). An important

area of the uses of carbon nanomaterials in dentistry is carbon fiber reinforced epoxy resin posts (Bonon, Weck, Bonfante, & Coelho, 2016). Another application of the carbon/graphite fiber in dentistry is a reinforced polymethyl methacrylate (PMMA) denture base (Larson, Dixon, Aquilino, & Clancy, 1991). They observed that the flexural strength of the PMMA increases by adding nanoparticles. However, the negative aspect of the use of carbon materials is to show aesthetically unwanted image due to its black color (Meng, & Latta, 2005). Nanotechnology in dentistry started with the addition of nanoparticles to filling materials. Many dental materials, especially resin-based composite materials, cements and implant materials as the most important of this group contain nanoparticles. Today, resin composites are the most widely used in dental fillings (Althaqafi, Satterthwaite, & Silikas, 2020). Composites contain two or more basic materials and create a new material with feature. They have some advantages of each individual material. Dental resin-based composites contain different sizes of inorganic fillers such as supra-micron, sub-micron and nano-sized (Abdel Hamid, Esawi, Sami, & Elsalawy, 2008). Nano-sized filler particles are added to fill the space between larger particles and reduce the resin content of resin-based composites. Since they contain both large and very small particles, these resin-based composites are called hybrid composites or nanohybrid composites.

Nanofiller also contains silica nano-particles (1-100nm) throughout the resin matrix. The surface of these particles is treated with a silane coupling agent such as MPTS (2-methacryloxypropyl-trimethoxylane). This silica ester binds to the inorganic surface at one end to prevent any agglomeration and clusters. Nanoclusters were formed by light sintering of nanomeric oxides to produce composites with good rheological properties (Mitra, Wu, & Holmes, 2003). Lee et.al. (Lee et al., 2020) worked on boron nitride nanoplatelets (BNNPs) and demonstrated their excellent mechanical properties (fracture toughness, flexural strength and wear resistance) and biocompatibility, as a suitable reinforcement for dental materials. Weidenbach Degrazia and coworkers (Degrazia, Leitune, Samuel, & Collares, 2017) used boron nanotubes and incorporated of boron nitride nanotubes into adhesive resin materials to improve their physical and chemical properties. Alqahtani (Alqahtani, 2020) enhanced mechanical properties of dental materials by curing PMMA with zirconia and boron nitride nanopowders. In the present research, a dental composite was prepared by using the modified h-BN nanoparticle as inorganic filler for dental process and its physical, mechanical and morphological properties were analyzed.

## **MATERIAL AND METHODS**

### **Materials**

Hexagonal boron nitride nanoparticles were obtained from Bortek Company. 3-amino 1, 2, 4 triazole ( $C_2H_4N_4$ , Mw: 84,08 g/mole) was supplied from Sigma. 2,2'-Azobis(2-methyl propionitrile) solution (AIBN) was purchased from Merck. Glycidyl methacrylate (chemical formulas:  $C_7H_{10}O_3$  and Mw: 142,15g/mole) and the other chemicals were obtained Sigma Aldrich. The solvents used in this work as toluene and N, N-dimethylformamide were purchased from Aldrich Company.

### **Methods**

Preparation of Hexagonal Boron Nitride Nanoparticles with Glycidyl Methacrylate (PGMA/h-BN-Nps)

2 g h-BN-Nps were dissolved in toluene by continuous mixing. Then, 6g of glycidyl methacrylate (GMA) and 0,130 g of Azobisisobutyronitrile (AIBN) were added to the solution and mixed until homogenous mixture. The mixture was refluxed at 75°C overnight under  $N_2$  gas. The resultant substances were washed with toluene to remove the unreacted materials. The washed materials were dried in an oven at 60°C and then kept in dark a place until used.

Modification process of PGMA/(h-BN-Nps)- with Triazole

1g of PGMA/(h-BN-Nps) and 0,463 g of 3-amino-1, 2, 4-triazole were mixed by magnetic stirrer, and, then, refluxed in dimethyl formamide (DMF) at 90°C for 24h under  $N_2$  gas. The resultant substance PGMA/(h-BN-Nps)/ATri was washed with distilled water and centrifuged. The sample was then dried in an oven at 50°C and then kept in dark a place until used.

#### Preparation of Dental Nanocomposite: PGMA/(h-BN-Nps)/ATri with TEGDMA

4,5 g of triethyleneglycol di methacrylate (TEGDMA) as a solvent was added to 1,0 g of PGMA/(h-BN-Nps)/ATri). 0,05g of Camphor Quinone were added to the mixture of h-BN/PGMA/ATri and TEGDMA. The composite was poured into the Teflon petri dishes. Finally, the composite paste was cured using LED light at the wavelength between 420- 480 nm for 40s.

#### The Characterization of the prepared composite

Strength Test is applied by using a universal testing machine. The samples were prepared as a disc shape with 5 mm diameter and 3 mm in thickness. Before applying this test the surfaces of the sample were polished. The sample was placed between the plates of the machine. The force was applied on the sample and the relationship between stress (MPa) and strain (%) of the sample was obtained. The dispersion of the inorganic filler in the nanocomposites was analyzed by SEM. The solubility of the cured composites were examined in 3 different media. For this purpose, the samples were first weighted ( $m_1$ ), then kept in distilled water and in buffer solutions at pH = 5 and pH = 9 at room temperature for a week.

The samples were removed from the distilled water and buffer solutions, and they were weighted until reaching a constant mass ( $m_2$ ). The solubility was found by the following equation:

$$SL \% = \frac{m_1 - m_2}{m_2} \times 100 \quad (1)$$

#### Cytotoxicity test

The sample was first sterilized for 30 mins under UV light. Then, they were washed with ethanol/deionized water (1/2) (v/v). The samples were shaken in DMEM F12 (1:1 mixture of Dulbecco's Modified Eagle's Medium (DMEM) and Ham's F-12 Nutrient Mixture obtained from Merck company) at 37 °C for 3 days in an incubator. L929 were seeded in the wells. DMEMF12 was placed on the cells and kept for 24 hours at 37 °C. The absorbance was measured by WST1 viability test.

#### RESULTS AND DISCUSSION

The chewing process is satisfied with the highest mean value of flexural strength ( $590 \pm 64$  Mpa) (Fig. 1).

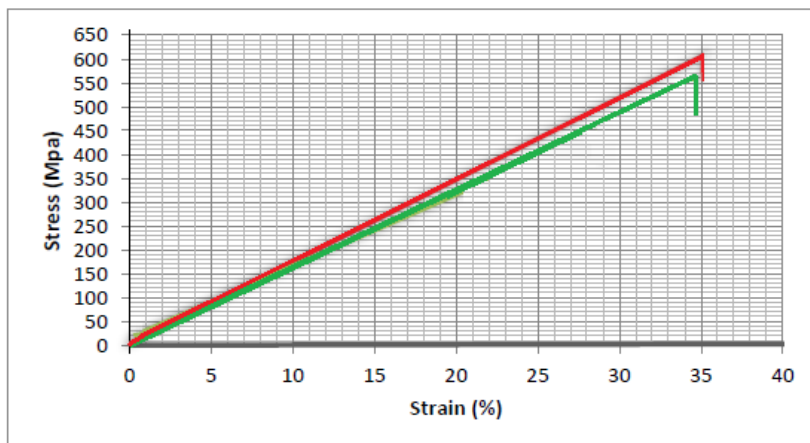


Figure 1. Relationship between stress and strain. Standard deviation of the flexural strength (green line) and flexural modulus (red line).

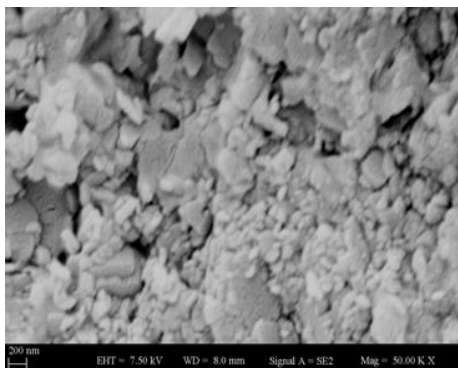


Figure 2. SEM micrograph of the prepared sample.

The homogenous distribution of the inorganic filler throughout the organic matrix, which was due to the attachment between the organic matrix and inorganic filler, was observed from the morphological analysis (Figure 2).

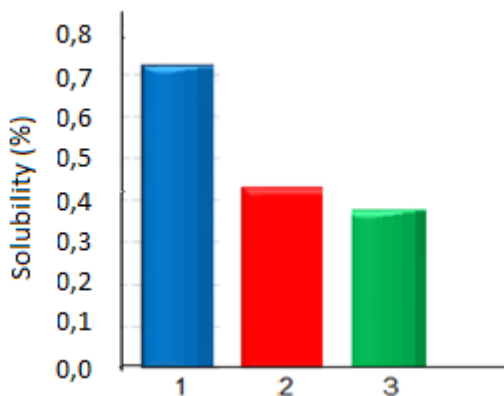


Figure 3. The % of solubility of the sample is seen in distilled water and in different buffer solution (1. Solubility percentage of cured nanocomposites in the distilled water (0.72); 2. Solubility percentage of cured nanocomposites in the acidic medium (0.42); 3. Solubility percentage of cured nanocomposites in the basic medium (0.37)).

The result of the solubility test is shown in Figure 3. The low solubility values of the composite in different media showed that this prepared material can be used in the dental processes.

Figure 4 shows the results of cytotoxicity tests obtained from the sample and the control group. By comparing the control group with the sample due to the cytotoxicity test it can be observed that the prepared composite shows nontoxic property.

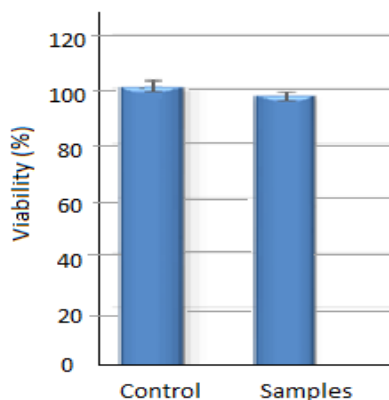


Figure 4. Cytotoxicity property of the sample comparing with control group.

## CONCLUSIONS

In this study, h-BN-Nps were used to prepare dental nanocomposite. The experimental results showed that the sample is non-toxic and it has good properties as hardness. The sample also performed better flexural strength, cytotoxicity, solubility properties than a tradition dental fillings.

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