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INFLUENCE OF INCORPORATION OF SiO_2 AND ZrO_2 NANOPARTICLES ON SOME PHYSICAL AND MECHANICAL PROPERTIES OF THERMOPLASTIC RESIN MATERIAL

Mahmoud Abdellah *, Mohamed Shokry Ali **, Ramy Abdallah Abdelrahim***

ABSTRACT

Objective: This study was aimed to evaluate the effect of incorporation of two-different nanoparticles (NPs); silica (SiO_2), and zirconia (ZrO_2) on flexural strength, impact strength, as well as, water sorption and solubility of thermoplastic resin material. **Materials and Methods:** Two nanoparticles SiO_2-NPs, and ZrO_2-NPs (Nano-gate Company, Egypt) were incorporated into thermoplastic resin with different concentration (1.5 and 7 wt.%). This study was divided into five main groups; unmodified "control group" and four modified groups according to the type and concentration of the incorporated nanoparticles. **Results:** The results of this study revealed that the incorporation of 1.5 wt.% SiO_2-NPs significantly improves the flexural strength of thermoplastic resin. While the incorporation of SiO_2-NPs and ZrO_2-NPs nanoparticles insignificantly decreases its impact strength. However, they significantly increase water sorption and solubility of thermoplastic resin. **Conclusion:** The incorporation of nanoparticles into thermoplastic resin at different concentrations could negatively affect its flexural strength, impact strength, as well as its water sorption and solubility.

Key Words: Nanoparticles, Mechanical properties, Silica, Thermoplastic resin, Zirconia.

INTRODUCTION

To overcome the disadvantages of conventional heat-polymerized polymethylmethacrylate (PMMA) acrylic resin a wide range variety of alternative denture bases resin materials were lately addressed such as polycarbonate resins, acetal resins, polyamides, and thermoplastic resin material [1,2]. So, the new terminology of "metal-free" removable thermoplastic prosthesis was introduced in dental practice [1,3].

Recently the use of thermoplastic resin has an ongoing increase in dentistry [4]. This resin is a polymerized acrylate, fabricated via the blending of methyl methacrylate "MMA" with other co-polymers to improve the inherent low impact strength of conventional heat-polymerized PMMA resin [5]. The manufacturing technology of thermoplastic resin based on plasticizing the resin material via thermal transformation in the absence of any actual chemical reaction. The mold injected technology of this plasticized resin opens a new trend for the fabrication of complete and partial denture prosthesis [4].

These resins are mostly used for the construction of removable denture bases due to their advantages such as excellent esthetics, biocompatibility "monomer-free", and smoother surface texture [5].

* Assistant Lecturer of Dental Biomaterials, Faculty of Dentistry, Asyut, Al-Azhar University
** Professor and Head of Department of Dental Biomaterials, Faculty of Dentistry, Cairo, Boys, Al-Azhar University
*** Assistant Professor of Dental Biomaterials, Faculty of Dentistry, Cairo, Boys, Al-Azhar University

Despite these previously mention advantages of thermoplastic resin, but it was found that thermoplastic resin still has poor impact strength, however it has adequate flexural strength which renders them non-ideal [6]. So, many attempts have been made to improve the mechanical and physical properties of the acrylic resin material such as the incorporation of silica and zirconia nanoparticles as reinforcing additives [7-9].

Silicon oxide nanoparticles (SiO_2-NPs) are mostly used to reinforce the acrylic resin because of their excellent biocompatibility as well as their optical, thermal, and mechanical properties [7-10]. However, zirconia oxide nanoparticles (ZrO_2-NPs) received attention due to their white color as well as other beneficial properties such as excellent biocompatibility, toughness, and strength [9, 11].

Hence, the purpose of this study was to evaluate and compare the effect of silica and zirconia nanoparticles on some physical and mechanical properties of thermoplastic resin material. The hypothesis was that the reinforcements will improve the physico-mechanical properties of the thermoplastic resin.

MATERIALS AND METHODS

In this in-vitro study, one type of thermoplastic resin material (i-flex, TCS^{tm}, Inc., USA) as well as, two different nanoparticles (silica, and zirconia) with different concentrations (1.5 and 7 wt.%) (Nano-gate Company Egypt) were selected.

Samples Grouping:

A total of 150 samples were made from an unmodified and modified thermoplastic resin material and divided into the following five groups according to the type and concentration of the incorporated nanoparticles (n=30).

Group I: Unmodified thermoplastic resin (control group).

Group II: Thermoplastic resin modified with 1.5wt.% of SiO_2-NPs.

Group III: Thermoplastic resin modified with 7wt.% of SiO_2-NPs.

Group IV: Thermoplastic resin modified with 1.5wt.% of ZrO_2-NPs.

Group V: Thermoplastic resin modified with 7wt.% of ZrO_2-NPs.

Then, each main group was divided into three subgroups according to the type of test(n=10).

Mold Fabrication:

For flexural strength test, milled stainless-steel metal plates with a rectangular form of 65mm x 10mm x 2.5 mm were used [12] and milled stainless-steel metal plates with a rectangular form of 75mm x 10mm x 10 mm for impact test was used [13], while, for water sorption and solubility test, a milled stainless-steel metal plates with disk form of 50mm x 0.5 mm was used [14].

Each stainless-steel metal plates were painted with a separating medium, and then flasked with stone into a metal flask, after the complete set of stone, the top half of the metal flask and the stainless-steel metal plates removed leaving mold spaces in stone of the bottom half of flask with the same dimension of each stainless-steel metal plate. Then, the stone mold was painted with a separating medium for the application of unmodified and modified thermoplastic resin of different main groups.

Samples Fabrication:

The unmodified thermoplastic resin prepared according to manufacturer's instructions and then the thermoplastic resin material was injected in the stone mold after painting by a separating medium, using an injection machine (Thermogen, Egyptian Engineering, Egypt) [15].

The modified thermoplastic/ SiO_2-NPs and thermoplastic/ ZrO_2-NPs granules were prepared by using electronic analytical balance device

(Sartorius, Sartorius AG. Germany) to measure each nanoparticle concentration to be added to the unmodified thermoplastic resin granules. Then the modified nanocomposite was stirred with an electric mixer at a rotating speed of 400 rpm at room temperature for 30 minutes to obtain a more homogenous and equal distribution of nanoparticles. The modified resins were processed according to the manufacturer's instructions and then injected using an injection machine in their specially designed stone mold [16].

All unmodified and modified samples were then trimmed using a tungsten carbide bur, ground with an emery paper 120, 200, 400, and 600 grain respectively, to remove any remaining small scratches and to get a smooth, highly polished surface [16].

Flexural Strength Test:

Each specimen was individually and horizontally mounted for a three-point bend test on a computer-controlled material testing machine (Model LRX-plus; Lloyd Instruments Ltd., Fareham, UK) with a load cell of 5 KN and data were recorded using computer software (Nexygen-MT; Lloyd Instruments).

Then, each specimen was statically compression loaded until fracture at a crosshead speed of 5 mm/minute. The maximum load exerted on the samples was recorded, and the flexural strength was calculated according to the following equation [17]:

Where; FS: flexural strength; W: load at fracture, L: span length of specimen (mm); b: width of the specimen (mm); and d: thicknesses of the specimen (mm).

Impact Strength Test:

Impact strength test was performed using a Charpy-type impact tester (Coesfeld, Pendulum Impact Tester, Dortmund, Germany). Impact strength (IS) was calculated using the following formula [18]:

Where; E: is the energy required to break the specimen (J), w: is the width (mm), and t: is the thickness of the specimen (mm).

Water Sorption and Solubility Test:

The completely dried samples were weighed using analytical digital balance and (M1) was recorded. Then, all samples in each group were transferred to separate glass vessel with 20 ml of deionized water and stored in a 37°C incubator for 1 week. The deionized water was changed daily. After water storage, the samples were removed from the water, blot dried with an absorbent paper and weighed to record (M2). Then, each disk was placed in a desiccator containing silica gel for 7 days and dry mass (M3) was recorded.

The maximum mass gain (apparent water sorption) and mass loss (solubility) at equilibrium were calculated for each disk using the following equations [19]:

Where; M1: the mass of the specimen, in microgram (μg), before immersion in water; M2: the mass of the specimen, in μg, after immersion in water; M3: the mass of the specimen, in μg, after immersion and desiccation, V: the volume of the specimen in cubic millimeters (mm^3).

The volume of the specimen in (mm^3) was calculated using the following equation [20]:

Where π; 3.14, r; the radius of the specimen in (mm), and h; thickness of specimen in (mm).

Statistical Analysis:

All resulted data were collected, tabulated, and statistically analyzed using IBM® SPSS® statistics Version 25. Numerical data were described as mean and standard deviation. Data were compared using the ANOVA test. The level of significance will be set at $P<0.05$.

RESULTS

Flexural Strength:

The values of flexural strength results measured in MegaPascal (MPa) of unmodified and modified thermoplastic resins were statistically significant (p-value = 0.006), Figure 1.

In the comparison between groups, Tukey's pairwise post-hoc test showed a statistically significant difference (p<0.05). Where there was a significant difference between the unmodified group and modified groups. The modified groups revealed a significant difference among the groups, except for group IV and group V were there was no significant difference.

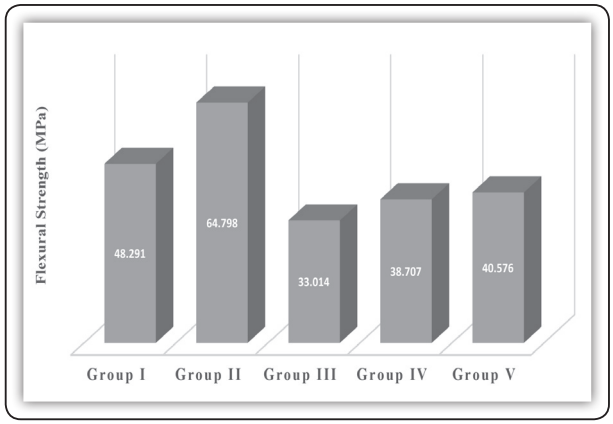

FIG (1) Column chart of the mean values of flexure strength among all tested groups.

Impact Strength:

The values of impact strength results measured in joule/mm^2 (J/mm^2) of unmodified and modified thermoplastic resins were statistically non-significant (p-value = 0.151). The highest mean value was recorded for unmodified thermoplastic resin, followed by modified groups, Figure 2.

Water Sorption:

The values of water sorption results of unmodified and modified thermoplastic resins were statistically significant (p-value = 0.00000), Figure 3.

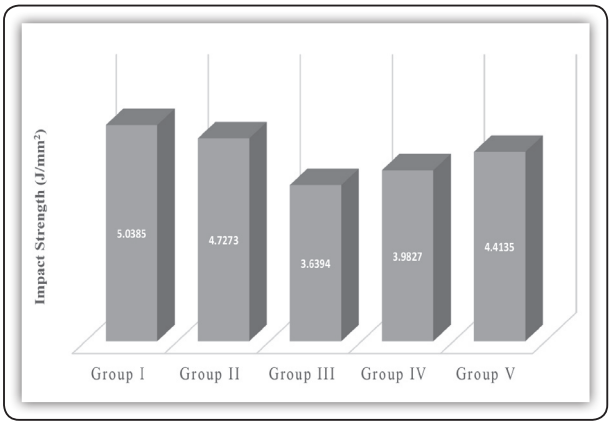

FIG (2) Column chart of the mean values of impact strength among all tested groups.

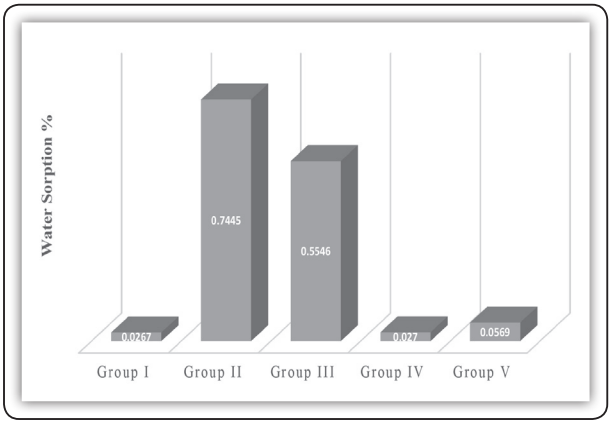

FIG (3) Column chart of the mean values of water sorption among all tested groups.

In the comparison between groups, Tukey's pairwise post-hoc test showed a statistically significant difference (p<0.05). Where there was a significant difference between the unmodified group and ZrO_2-NPs modified groups. While no significant difference between the unmodified group and SiO_2-NPs modified groups.

Water Solubility:

The values of water solubility results of unmodified and modified thermoplastic resins were statistically significant (p-value = 0.00000), Figure 4.

In the comparison between groups, Tukey's pairwise post-hoc test showed a statistically

significant difference (p<0.05). Where there was a significant difference between the unmodified group and modified groups. Furthermore, there was a significant difference between all modified groups.

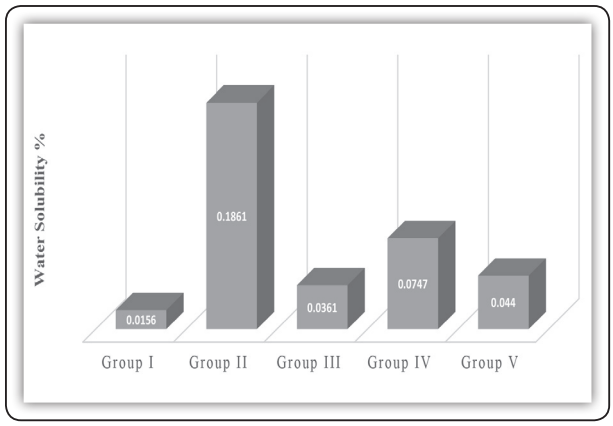

FIG (4) Column chart of the mean values of water solubility among all tested groups.

DISCUSSION

Thermoplastic resins are lately used as an alternative to the conventional heat-cured PMMA because of the newly introduced injected molded technique which eliminates the need of using powder/liquid system that can cause allergic for patients due to residual free monomers [2,5, 21]. Also, they are the non-porous structure, so, no bacterial growth, however, they can retain a little amount of water to keep some comfortable against soft oral tissues[22,23]. Furthermore, the polymer chains of thermoplastic resins bond via intermolecular forces[2,4,24].

Flexural strength is an important property of the denture base materials that can reflect the ability of these materials to functionally resist the masticatory forces during clinical services [25]. The three-point flexural (bending) test is the test used to compare the denture base materials as it simulates the original stresses that are applied to the denture during service [25].

Generally, the properties of polymer/nanocomposite materials are superior over the conventional pure polymer matrix. The effect of nanoparticle inclusion is dependent on their distribution, aggregation potential, and interaction with the resin matrix as well as their crystalline or amorphous nature [9,26].

In the present study, it was found that the incorporation of 1.5 wt.% SiO_2-NPs significantly improve the flexural strength of thermoplastic resin. This may be because silica (SiO_2) is a porous structure and can adsorb various ions and/or molecules, so, it can form an additional intermolecular bond with the thermoplastic resin and eventually improve its flexural strength [24,27]. Also, it could be attributed to the high interfacial shear strength between silica nanoparticles and polymer matrix as a result of the formation of "supra-molecular bonding" which in turn covers or shield the silica nanoparticles and prevent crack propagation [24].

However, the incorporation of 1.5 wt.% ZrO_2-NPs significantly decreases the flexural strength of thermoplastic resin. This may be due to zirconia nanoparticles has an extremely high surface activity that causes these particles to aggregate and from clustering in the resin matrix which may weaken the material and enhance the propagation of crack [28-30]. Furthermore, the incorporation of 7 wt.% ZrO_2-NPs significantly decreases the flexural strength of thermoplastic resin. This in agreement with the previous studies which revealed that the increase in the concentration of nanoparticles beyond 5 wt.% resulted in aggregation of nanoparticles and cluster formations that eventually resulted in weakened material [11].

Moreover, the significantly decrease the flexural strength of thermoplastic resin in the present study with the incorporation of 7 wt.% SiO_2-NPs and ZrO_2-NPs, maybe also due to incomplete wetting of the nanoparticles by the thermoplastic resin which leads to decrease in its flexural strength [31,32]. This in agreement with the previous studies which found that when the concentration of nanoparticles exceeded over a particular percentage of 2.5 wt.%

in the resin matrix, an adverse effect was observed and the values of flexural strength significantly decreased in the thermoplastic resin material [33].

The results of the present study revealed that the incorporation of both SiO_2-NPs and ZrO_2-NPs insignificantly decrease the impact strength of thermoplastic resin. This may be due to that the incorporation of hard SiO_2 and ZrO_2 nanoparticles into thermoplastic resin can increase the brittleness of the samples because of limiting the free movement of the polymer chains due to interruption of intermolecular forces, which would reduce the impact strength [10,34]. Also, the inhomogeneous distribution of the nanoparticles with frequent clustering could affect the impact strength negatively[10,33,35].

Furthermore, the incorporation of SiO_2 and ZrO_2 nanoparticles into thermoplastic resin significantly increase its hydrophilicity (water sorption). This can explain as the incorporation of these nanoparticles between the linear polymer chain of thermoplastic acrylic resin separate the polymer chains further apart from each other and increase spaces between the polymer chains [1,15,36]. Furthermore, they can increase their porosity which leads to an increase in water sorption of thermoplastic resin. Additionally, the increased porosities in this resin may be due to the inclusion of air during the injection procedure of the modified resins [37].

However, the significant increases in water solubility of thermoplastic resin may be due to their lower cross-linkage as its chemical structure which mainly linear polymer structure with intermolecular bonding mechanism which adversely affected by the incorporation of these nanoparticles [1,15, 24, 37].

CONCLUSION

The incorporation of silica nanoparticles at optimum concentrations could improve the flexural strength, and water solubility of thermoplastic resin, while both nanoparticles at different concentrations negatively affect its impact strength, as well as its water sorption and solubility.

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