Penetration of Liquid Glass Phase into Porous (3Y-TZP/Al$_2$O$_3$) Composite

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Abstract

3 mol% yttria stabilized tetragonal zirconia polycrystalline/Alumina (3Y-TZP/Al$_2$O$_3$)-Glass composite was fabricated successfully by infiltration of liquid glass phase into (3Y-TZP/Al$_2$O$_3$) porous body. Porous (3Y-TZP) was prepared first by pressing the powder mixture of (3Y-TZP) with 20 wt.% of nano alumina Al$_2$O$_3$ and (0,10,20,30,40) wt.% of graphite particles uniaxially to form cylindrical shaped specimens. The specimens were sintered at 1500 °C for 2 hours. The glass mixture of (18wt.% of hydroxide lithium, 78wt.% feldspar and 10 wt.% of nano titanium dioxide) were melt infiltrated into porous (3Y-TZP/Al$_2$O$_3$) at 1185 °C by capillary pressure to form composites. The glass amount penetrated into porous 3Y-TZP/Al$_2$O$_3$ depends on the amount and size of porosity exist in the alumina toughened zirconia (ATZ) structure. The results showed that the amount of glass infiltrated through (3Y-TZP/Al$_2$O$_3$) structure depends on the amount and size of pores. Increasing the amount of porosity and its size led to decrease the capillary action within pores through specific time. The porosity of the specimens was in the range of (7-50.5) %, while the diametrical strength values were in the range of (80.1-26.7) MPa. The glass infiltration process shows a very low values of shrinkage ranging from (0.21% to 0.31%).

Keywords: Glass penetration ceramic, diametric strength, glass, zirconia /Alumina composite.

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نفاذية الطور الزجاجي السائل في متراكب (3Y-TZP/Al₂O₃)

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الخلاصة

تم تحضير متراكبات (3Y-TZP/AL₂O₃)-زجاج بنجاح عن طريق ترشيح الطور الزجاجي السائل في الجسم المسامي لـ (3Y-TZP/AL₂O₃). تم تحضير المركب المسامي (3Y-TZP/Al₂O₃) اولا عن طريق كبس خليط من مسحوق (3Y-) TZP مع 20 Wt.% نانو الومينا و (40،30،20،10،0 wt.%) لجزيئات الكرافيت بطريقة الكبس اللامحوري لتكوين عينات ذات شكل اسطواني. تمت عملية تلبيد العينات عند حرارة C°1500 لمدة ساعتين لانتاج عينات مسامية ليتم لاحقا ترشيح الطور الزجاجي السائل. تم ترشيح خليط الزجاج من خلال (3Y-TZP/Al₂O₃) المسامي بدرجة 1185°C عن طريق الضغط الشعري لتكوين المتراكب (3y-TZP/Al₂O₃) كمية الزجاج النافذة من خلال الجسم المسامي تعتمد على كمية وحجم المسامات الموجودة في المركب (3Y-TZP/Al₂O₃,) اظهرت النتائج ان حجم المسامات يزداد بزيادة كمية المسامات في المادة مما يؤدي الى نقص في الخاصية الشعرية وبالتالي نقص في كمية الزجاج النافذ خلال زمن محدد. المسامية تراوحت بين (50.5-7) بينما قيم الكسر المحوري تراوحت (80.1-26.7MPa). عملية ترشيح الزجاج اظهرت نتائج واطئة جدا في قيم الانكماش الطولي تراوحت بين (0.31-0.21%).

الكلمات المفتاحية: الزجاج النافذ في السيراميك، المقاومة القطرية، زجاج، متراكبات زركونيا -الومينا.

Introduction

The dental industry has long searched for a material and processing technology for making all-ceramic crowns and bridges. Currently, these restorations consist of a fitted, metal coping (substrate) for strength with a porcelain coating for aesthetics [1]. The replacement of metal by ceramic will improve the optical properties and biocompatibility of the restoration however processing difficulties and brittle behaviour hampered attempts to fabricate and use all ceramic crowns [2]. Ceramic crowns were fabricated using dental porcelain prior to the mid-l960s, appearance and biocompatibility were good, but dental porcelain was too weak (flexural

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strength of -60 MPa), many researches have been directed toward enhancing the strength of dental ceramics since the mid-l960s [3]. Alumina-reinforced porcelain and glass ceramic dental materials were developed with increased strength (both have flexural strength of - 150 MPa); but improvements in the reliability and processing ease are needed before dental ceramics are accepted as replacements for metals [4]. A promising method for making all-ceramic crowns by melt infiltration of glass into porous ceramic was recently developed by Vita Zahnfabric (a German company), a thin crown is prepared by brushing a 65 vol% alumina slip onto a gypsum mold, firing the alumina at 1100°C for 2 h to develop a skeleton of fused alumina particles, and infiltrating the porous skeleton with a borosilicate glass at 1100°C for 2 h [5]. The chemical strengthening or the diffusion-based process leads to grain coarsening over a longer treatment time and thereby deteriorates the property. Ion exchange method causes a compositional gradient and stress variation in the coating [1]. In hot-pressing and pre reaction methods, ceramic particles dispersed in a glass matrix are drawn closer together by capillary pressure and significant shrinkage occurs [4,6]. By contrast, composites by the melt-infiltration process densify as the pores of ceramic skeleton are filled with glass. Molten glass is drawn in from the exterior of the sample by capillary action, as illustrated in Fig.1. Since the ceramic particles are fused together prior to infiltration, they are not free to move, and very little shrinkage can occur [7].
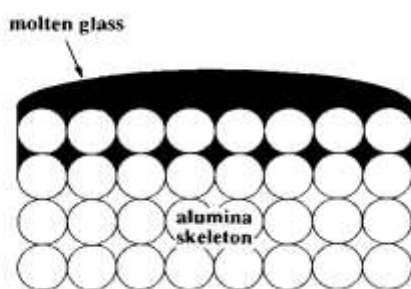

Figure 1: Densification in the melt-infiltration process occurs by drawing

the glass into the small pores of the alumina skeleton [7].

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Alumina toughened zirconia (ATZ) ceramics are widely used in many fields and particularly in dental restorations because it exhibits transformation toughening at high temperature, which greatly improves the fracture toughness, strength, and stability [8]. In this work, (3Y-TZP/Al$_2$O$_3$)-glass composites have been prepared by glass-infiltration into (3Y-TZP/Al$_2$O$_3$) composite. The 20 wt.% of (3Y-TZP) was replaced by alumina. The addition of Al$_2$O$_3$ is to control the specimen's dimensions. Mechanical and physical properties were investigated.

Materials and Method

2.1 Preparation of (3Y-TZP)/Al$_2$O$_3$

80Wt.% 3Y-TZP fine powder (3 mol.% Ytteria (Sky Spring Nanomaterials, USA) was mixed with ZrO$_2$ (Riedel de Haen, Germany, purity 99, particle size <5 μm) and with 20wt.% of nano Al$_2$O$_3$ (α-Al$_2$O$_3$ purity >99/99%, the average particle size of 13 nm, Sigma Aldrich, U.S.A) for the preparation of (3Y-TZP/Al$_2$O$_3$) composite also known as alumina toughened zirconia (ATZ). In order to prepare a porous structure, graphite particles supplied by (State Company of Geological Survey and Mining, particle size 32 μm) were added in 5 different ratios (0, 10, 20, 30 and 40) wt.%. The powder mixture was pressed uniaxially in steel die to form pellets (10 mm diameter and 5 mm thickness). The pressure was 2 ton for 30 seconds. The resulted specimens were sintered at 1500 °C for 2 hours.

2.2 Preparation of lithium silicate glass

Lithium hydroxide (manufactured by Strem Chemicals, Inc. USA) was mixed in an amount of 18 wt.% with 72 Wt.% of feldspar powder which was brought from State Company of Geological Survey and Mining-Iraq, and nano titanium oxide (26nm, purity > 99.8% supplied by Cheng Du Micxy Chemical Co. Ltd-China) in the amount of 10Wt.%. First, lithium hydroxide was dissolved in distilled water by magnetic stirrer followed by adding the other component of glass mixture (feldspar and titanium oxide) to the solution and mixed for 3 hours

for achieving homogenization. The powder mixture will form lithium aluminosilicate glass (Li_2O-Al_2O_3-SiO_2), TiO_2 was added as nucleating agent.

2.3 Preparation of (3Y-TZP)/Al₂O₃- glass

By mixing the glass powder with a little amount of water to prepare the glass slurry, slurry then was pasted on the surface of the sintered porous (3Y-TZP/Al₂O₃) specimen uniformly using a brush. Then the pasted specimens were dried in oven at 50 C. After drying, the specimen was heated in air using electrical programmable furnace (type Nabertherm-P310-Germany), the rising temperature rate was 17min/ °C up to 1185 °C for 2.5 hours. The glass starts to melt at certain temperature and filled the pores by capillary action.

Methods

1. Physical Properties for porous (ATZ)

1.1. Bulk density and porosity

Archimedes principle is used to calculate the apparent porosity (A.P) and bulk density (B.D) of (3Y-TZP/AL₂O₃) composite after sintering at 1500°C as follows:

$$A.P\% = \frac{W_s - W_d}{W_s - W_i} \times 100 \tag{1}$$

$$B.D\ (g/cm^3) = \frac{W_d}{W_s - W_i} \tag{2}$$

Where W_d is referred to the mass for dry sample, W_s is referred to the mass for sample being infiltrated with water, and W_i is referred to the mass for sample being immersed in water.

1.2 Linear Firing Shrinkage

A micrometre was used to calculate the length before and after sintering to determine the linear firing shrinkage for (3Y-TZP/Al₂O₃)-glass specimens. After drying and sintering of ceramic specimens, the total linear shrinkage was calculated as a percentage of plastic length, as follows:

$$\text{L. F. SH}\% = \frac{L_o - L}{L_o} \times 100 \tag{3}$$

Where L_o is the specimen plastic length (before firing) and L is the specimen fired length.

2. Mechanical properties (Diametrical strength)

Diametrical strength for porous ATZ specimens was measured by Brazilian test. The Brazilian test, is a disc of the tested material loaded across a diameter. The test was done by hydraulic pressing (Ley Bold Harris NO.36110). The thickness and diameter of the specimens have been calculated. Diametric strength then is calculated by using following equation:

$$\textbf{Diametric Strength (MPa)} \qquad \sigma_D = \frac{2F}{\pi DL} \tag{4}$$

Where the applied load (N) is F, the disk diameter is D and the thickness of disk is L.

3. Microstructural (Scanning electron microscope).

Scanning electron microscope (SEM) was used to investigate the microstructure of glass-(3Y-TZP/Al₂O₃) specimens. Scanning electron microscope (SEM), (FEI company, INSPECT S50), equipped with a field emission gun was used to assess the microstructure of the samples and the interface between glass and ATZ. The etched samples were coated with gold before being tested.

Results and Discussion

Porosity and bulk density were determined using Archimedes method. The properties of ceramics are strongly affected by porosity. Graphite is volatile material and it is used as a pore forming agent. The sintering process will burnout the graphite leaving pores behind.

Fig.2 shows the variations of apparent porosity and bulk density of (3Y-TZP/Al₂O₃) composite specimens sintered at 1500 °C with different graphite additives. An increase in the apparent porosity was noted when graphite addition increases. The percentage of porosity was increased

from 7.6% (without graphite additive) to 51.8% for 40 Wt.% graphite additive specimen while bulk density decreased from 4. 9 to 2.8 g/cm^3.
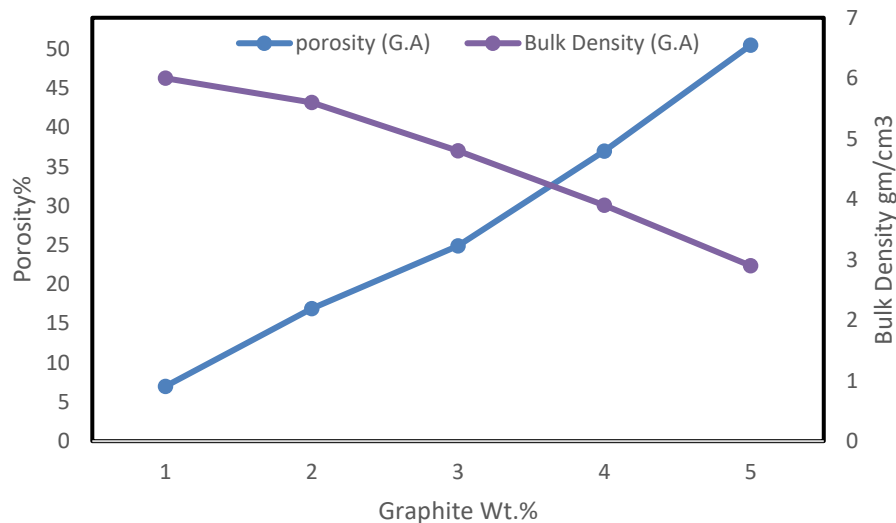

Figure 2: Apparent porosity % and bulk density variation with graphite additives.

The total increase of weight for each sample is shown in table (1), it is indicated that there is new material (glass) added to the ATZ structure and led to change the total weight of specimens, depending on porosity level, the times of infiltration can be determined. As the porosity level increases, the times of infiltration glass stages into ATZ matrix increased in order to occupy all the porosity into the structure, subsequently more glass amounts are needed to fill the porous structure.

The process stops when the specimens are saturated and the excess glass remained on the surface and cover the surface of the specimen.

In A1 as observed in fig.3 the sample is saturated from the second infiltration stage, the reason might be due to the low porosity content, and these porosities might close and hinder the flow of the glass into the structure [9]. As the porosity content increased, the samples need more infiltration stages and more time to be saturated, and then more weight percentage is gained.

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Table 1: Weight gain % for (3Y-TZP/20wt.%Al$_2$O$_3$) specimen per glass infiltration stage

Specimen	Weight gain %				
	S1	S2	S3	S4	S5
A1	2.8	4.1	4.1	4.1	4.1
A2	3.1	5.4	7.6	7.6	7.6
A3	4.8	6.2	9.8	12	12
A4	7.1	10	17	20.1	30
A5	11	15.3	20.7	38.5	43

Legend:

- S refers to stage, duration of each stage was 2 hrs.
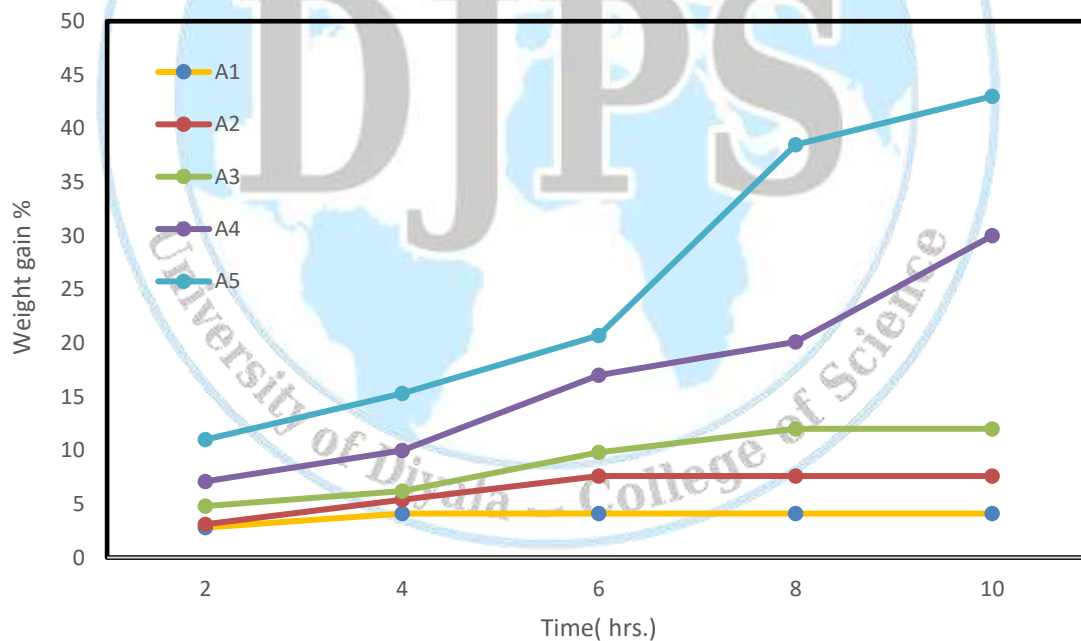- A is numeral no. of specimen.

Figure 3: Variation of weight gain % versus time of glass infiltration (3Y-TZP/20wt.%Al$_2$O$_3$).

The values of linear firing shrinkage of (3Y-TZP/Al$_2$O$_3$) are shown in figure (4). The glass infiltration process causes almost no shrinkage in composites with 20 wt.% Al$_2$O$_3$ additive < 0.3%, almost free shrinkage since alumina zirconia particles were in different sizes have formed

a rigid framework with an open porous structure after sintering at low temperature below1200 ^{o}C. The framework after initial sintering, not only prevents the alumina compact from shrinkage while the melted glass penetrates and occupies all of the open pores which exist in the infrastructure, but also increases strength and toughness of composite [10].
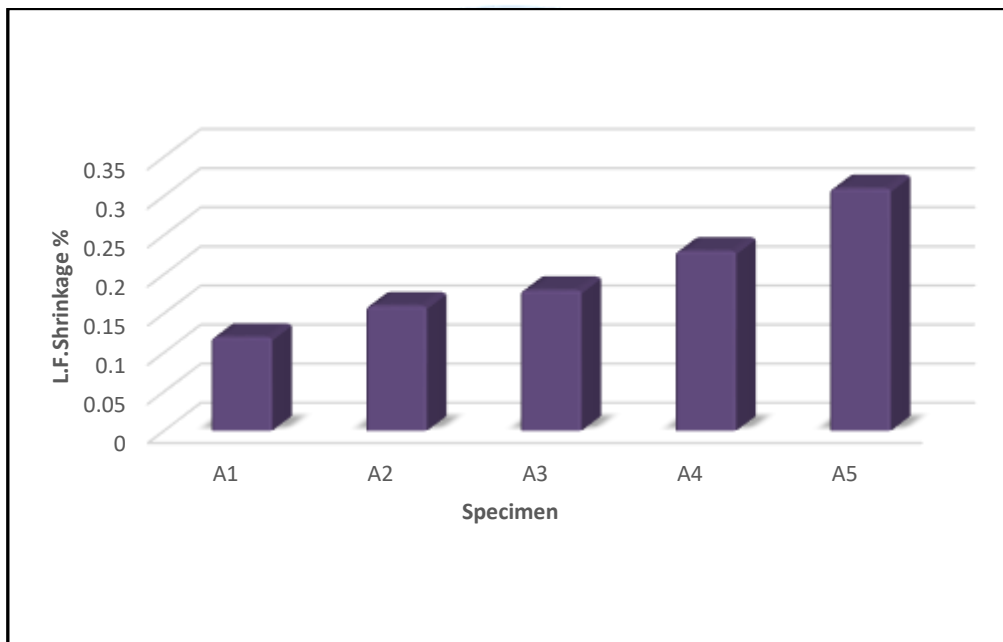

Figure 4: L. F. SH. % for (3Y-TZP/20wt% Al$_2$O$_3$) - glass

Diametrical strength for ATZ-glass composite specimens are shown in figure (5), the strength of specimens after glass infiltration was due to healing of cracklike defects of the ceramic structure, which are points of initiation of brittle failure. In addition, the pores themselves become more even and circular in form and create a decreased concentration of stresses [11]. The elastic property mismatch between the penetrated glass and ATZ produced residual compression in the surface region during cooling. This residual compression enhanced the composite strength [12]. When the strength was measured, the glass penetrated region was under tensile load. This region requires some extra force initially to neutralize pre-existing compressive stress, before the system actually undergoes tensile stress, hence more force is

required to break. In addition, the spheroidization of grains preferably minimizes the stress concentration zones in glass treated samples [13].

It was noticeable that the strength decreased with ATZ porosity due to the increasing presence of residual porosity closed in the (3Y-TZP/Al$_2$O$_3$)-glass structure which is responsible for the initiation of the crack. This behaviour agreed with other strength values reported by Shenyong Shia et al. as the flexural strength of (3Y-TZP)-glass composites was 600 for 5% porosity in the 3Y-TZP matrix and it turns into 400 when the porosity of 3Y-TZP matrix was about 25% [10]. Another theory to explain decrement of strength with increasing ATZ skeleton porosity is due to the pore size in ATZ structure, because there is a relation between pore size and capillarity height and time [14] so capillary rises faster (higher) in A1 specimen which had a pore size of about (1.20 μm) than A5 specimen which had a pore size of about (19.95 μm), for this reason the structure containing larger pore size will need more time for infiltration. Figure (6) shows the ATZ pore size for 3 different porosity contents.
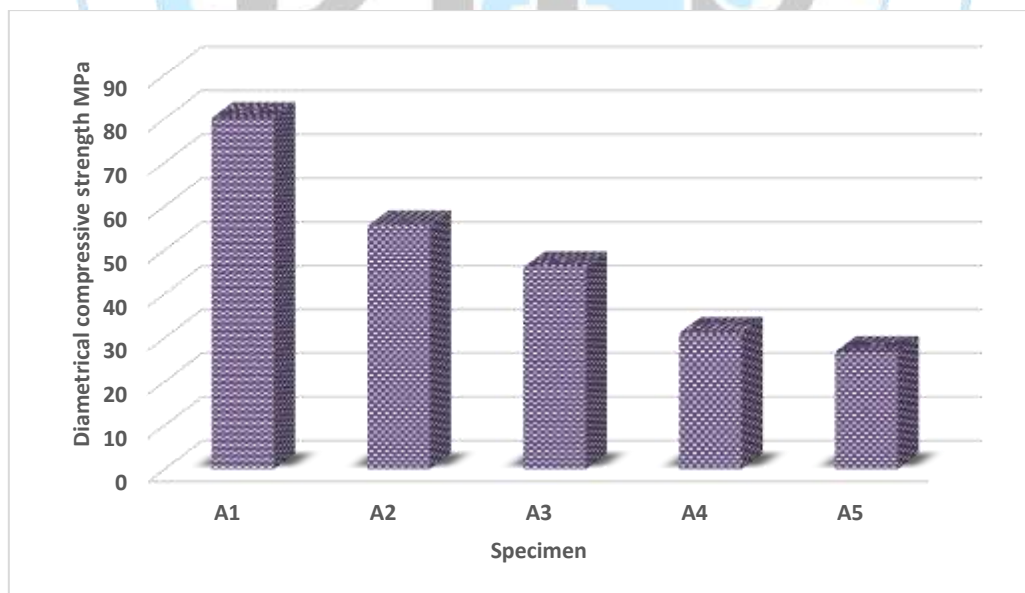

Figure 5: Diametrical strength values for (3Y-TZP/20Wt.% Al$_2$O$_3$) -glass.

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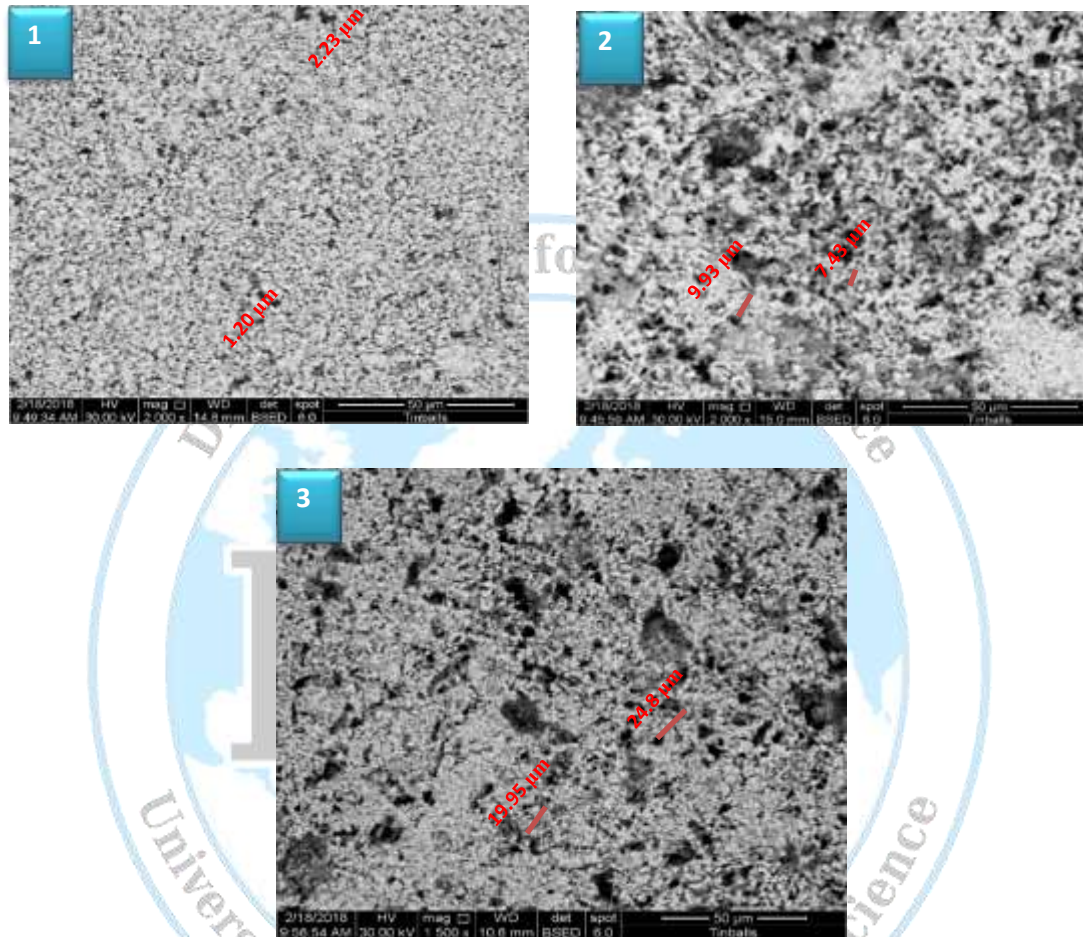**Sara N. Ibrahim, Shihab A. Zaidan and Mudhafar A. Mohammed**

Figure 6: SEM images for 3 different (3Y-TZP/Al₂O₃) porous specimen.

Conclusion

(3Y-TZP/Al₂O₃) specimen was successfully fabricated using pressureless infiltration of (3Y-TZP/Al₂O₃) with a lithium aluminosilicate glass. The most important fact is that the infiltration temperatures are lower than 1200 °C. The shrinkage of the composite was about (<=0.3%) which could be used in the near net-shape fabrication of dental restorations.

Diametrical strength depends on the amount of glass infiltrated into the (3y-TZP/Al₂O₃) skeleton, the amount of glass penetrated will not only be affected by the amount of porosity but also on the size of pores.

Small pore size has the capillary rise faster than larger pore structure. The filtration process replaced the process of surface coating and diffusion of the glass into the depth of the ceramic body gives the ceramic properties that qualify it for biological applications such as dental industry.

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