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Effect of CO_2 Laser on Glazed Zircon Surface Complemented by ZrO_2 Oxide

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K E Y W O R D S

CAD/CAM; Dental ceramics; CO_2 laser; Zirconia; Glazing; Residual stress, Grain size.

A B S T R A C T

In this paper, the effect of CO_2 laser on glaze-dental Zirconia_ceramics after adding ZrO_2 nanoparticles to glaze is introduced, and its improvement methods are studied. Specimens have been prepared using CAD/CAM dental machines and sintered at 1530o-C. Then the surface was glazed with VITA glaze plus (5% and 10%) Nano ZrO_2. A 15W continuous CO_2 laser was used as the indicator power to irradiate the glaze layer. The main phase of the ceramic substrate is tetragonal Zirconia, and the alumina content in the corundum phase is a certain percentage. The appearance of the varnish on the ceramic substrate changes the X-ray diffraction pattern through the appearance of new phases, which changes the crystallite size and the percentage of lattice strain. The range of grain size measured by atomic force microscopy was 88.46 nm to 62.18nm. In addition, the surface roughness was changed_due to the appearance of crystal cores and grain growth. In addition, the addition of ZrO_2 and laser irradiation changed the residual stress on the surface, which was reflected in the hardness value increased from 575 kg/mm2 to 1215 kg/mm2 after the laser treatment with the addition of 5% ZrO_2. Generally, in terms of the structure and hardness of the surface of the glaze layer, the addition of 5% ZrO_2 is better than 10%. SEM tests also showed no cracks in the central part of the treated area. These characteristics increase hardness.

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1. INTRODUCTION

The CO_2 laser was introduced into the alternative dental industry, by improving the surface of ceramic teeth, particularly zircon teeth [1]. This laser is perfectly suitable for the treatment of ceramic materials because it absorbs the wavelength emissions almost entirely of ceramic [2].

Zirconia is a very important engineering material because of its unique physic mechanical and refractory properties such as corrosion and wears resistance, high refractoriness, high fracture toughness and hardness, low coefficient of friction, low thermal conductivity at high temperature, ionic conduction, and high thermal shock resistance. Zirconia (ZrO_2) is a crystalline oxide of Zirconium and its mechanical properties are similar to those of metals, so, its color is similar to the teeth color [3]. Different oxides can be added to zircon to stabilize it, such as (ZrO_2), allowing the tetragonal form to exist at room temperature after sintering. Interaction between laser light and ceramic materials is of general importance for many different applications. The wavelengths of infrared (10.6 pm, CO_2-laser) and UV-radiation result in high absorption levels of oxide ceramics [4, 5]. Therefore, the purpose of this study is to evaluate the surface hardness, surface morphology, and structure of the Zirconia dental surface glaze layer. The originality of this subject is to add nano-ZrO_2 and perform laser treatment to obtain better specifications, to convert part of the amorphous glaze layer into crystalline glass-ceramics.

2. EXPERIMENTAL WORK

I. Materials and Sample Preparation

The used Zircon material is from (Easy co) company - Origin of high purity (99.9%) Zircon. The specimens were cut into small cylinders by CAD/CAM dental machine with a diameter (0.5 cm) and height (0.5 cm). The specimens were sintered at a temperature of 1530^o C for 120 minutes as hold time, by using an electrically programmable furnace type (Nabertherm-P310-Germany). The glazing process of the surface uses a mixture of the following, Vita Akzent plus Glaze LT (Low Temperature). A weight of (0.060 g) with 3 drops of (Vita Akzent Plus, Powder Fluid) and with two different percentages of ZrO_2 (5% and 10%) nanoparticles (< 26 nm), purity ≥ 99.8% supplied by Cheng Du Micxy Chemical Co Ltd – China was added to glaze mixture to produce two different glazing layers. After the completion of the glazing process on the specimen's surfaces, the specimens were dried and fired up to 800^o C.

The surface of the sample was irradiated by using a CNC CO_2 laser machine with a beam of 10.6 µm wavelength. The spot size used in treatment was 145µm and the output power range from 1-70Watt. Because of the high temperature of irradiated porcelain with this wavelength and the safety of ceramics, the value of energy output power used is 15Watt, with an Overlapping space of 0.01 mm. The irradiation done offline After 10 mm the size of the beam spot is aimed at the entire specimen surface with a speed of 500 mm/min and time 4.14 min. The laser handpiece was kept perpendicular to the irradiated surfaces.

II. Test Methods

The x-ray diffraction, XRD, measurements were conducted on a Shimadzu 6000-XRD with a scan range of (20-80 deg.). The mode of the scan was a continual scan rate of 10 deg. /min). The X-ray tube Cu (Filter). The crystallite size of the structure is estimated by using Scherer's formula:

$$D_c = \frac{K\lambda}{\beta\ \cos\theta} \tag{1}$$

Where K=0.94 and λ is the wavelength of X-Ray used, which is Cu K_α radiation (λ=1.54 A^o) and β is the full width at half maximum (FWHM) of the diffraction peak corresponding to a particular crystal plane. The lattice strain (ϵ) was calculated using the formula [6-, 8].

$$\boldsymbol{\epsilon = \frac{\beta}{4\ \tan\theta}} \tag{2}$$

The Atomic Force Microscopy device (AFM) type SPM-AA 3000 Angstrom (USA) is used in surface science laboratories to obtain grain size and distribution, as well as the images with atomic resolutions of 10 A^o. This type of microscopy can be effectively applied in the field of specimens to study the surface characteristics of a specimen. The instrument is based on the principle that when a tip, integrated into the end of a spring cantilever, is brought within the interatomic separation between the tip and sample, interatomic potentials are developed between the atoms of the tip and the atoms on the surface.

Roughness is one of the most important surface characteristics that can be measured with high precision using AFM.

Among Height Parameters, the roughness average (Ra) is the most widely used because it is a simple parameter to obtain when compared to others. The roughness average is described as follows:

$$\mathbf{Ra} = \mathbf{1}\boldsymbol{l} \int |(\mathbf{x})| \mathbf{dx}\ \boldsymbol{l}\ \mathbf{0} \quad (3)$$

The microhardness was measured by the Vickers microhardness apparatus. Before you perform a hardness test, the surfaces of all. For all specimens after surface roughness measurement to ensure that the specimens have the same hardness. The measurements done with a load of 30 kg for 15 seconds as a hold time. Three measurements were taken at the center of the laser spot and averaged to one value.

Scanning Electron Microscope, SEM, type Tescan Vega 3 SB, Magnification: Continuous from 6 xs to 100,000xs, accelerating voltage (200V to 30kV), made in Japan was used for measuring linear dimensions, shape, orientation and other parameters of nanostructures and microrelief of surfaces of various objects.

3. Results and Discussion

Figure (1) represents the x-ray diffraction pattern for all specimens sintered at 1530°C and glazed with different percentages of frit glass supported with Nano zirconium dioxide powder. Scheme TZP shows the main structures of the substrate ceramic material without glazing. It is clear that the components are tetragonal Zirconia (t-ZrO_2) and alumina (α-Al_2O_3). They represent the components of toughed Zirconia. Alumina modifies the toughness when added to Zirconia at a limited percentage [9]. We also noticed that the percentage of cubic phase is low. Also, the monoclinic phase was absent. After glazing without any addition of Nanomaterials and with laser surface treatment. Amorphous phases appeared between the crystalline phases as represented by the peaks of the TZP as in the scheme (TZP-Glazes). These amorphous phases led to the atrophy of the α-alumina phase (corundum) [10]. After adding 5% of ZrO_2 for glaze glass and without laser treatment, we noticed a simple increase in the glass layer components as shown in the scheme (TZP- glazes 5+ % for ZrO_2). Thus, the ratio of rutile enhanced the appearance of glass and ceramics. After the treatment of the surface by laser, the process led to the emergence of new crystalline phases clearly. This indicated the increase in the proportion of glass-ceramic composite. As in the scheme (TZP-Glazes+5% ZrO_2 with the laser).

The increase of the ZrO_2 to 10% without laser treatment led to the emergence of the rutile phase with notice appearance of crystalline phases in the glass mixture, or the transformation of part of the amorphous glass compound into crystalline phases, as shown in scheme (TZP-Glazes+10% ZrO_2). Thus, new characteristics may emerge that may help to improve the properties associated with synthetics such as mechanical properties. After laser treatment, all crystalline phases have become clear. This is evident from the scheme (TZP-Glazes+10% ZrO_2 with the laser).

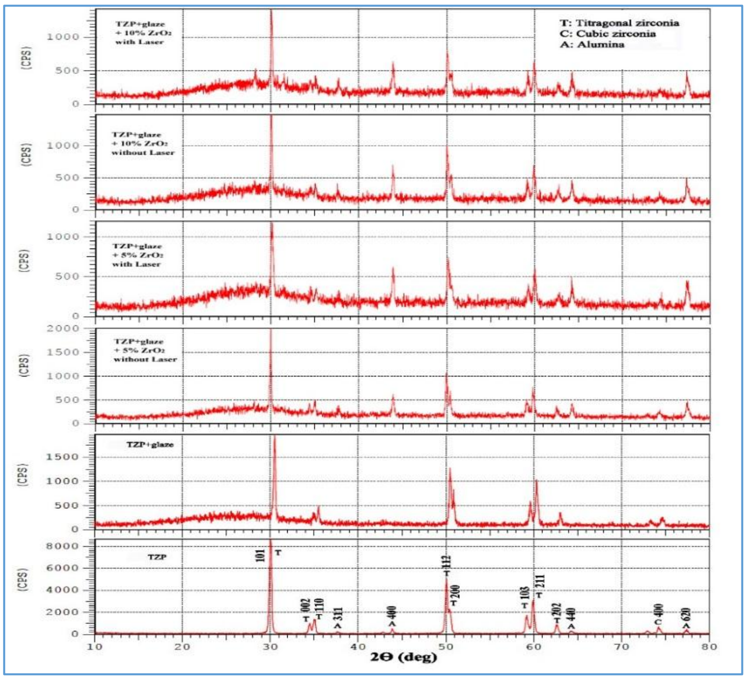

Figure 1: X-ray diffraction patterns of specimens

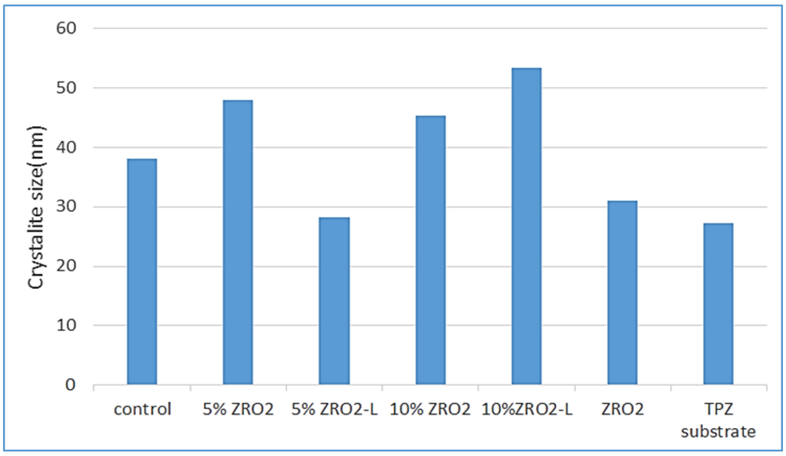

Figure 2: Crystallite size of specimens: (control: TZP with glazes and with laser treatment).

Figure (2) showed the results of crystallite size calculated from X-ray diffraction data. We noted from the figure that the crystallite size was 38.08 nm in the case of glaze with laser treatment of the zirconia surface without any addition. That value increased to 47.99 nm when 5% of Nano ZrO_2 powder was added and the change caused by ZrO_2 in terms of the appearance of other crystalline phases and increase surface transparency turned white.

The increase in the size of the crystal for (FWHM) declined the peak of diffraction corresponding to a certain crystal plane in the degree of 0.22 to 0.17 when you added 5% ZrO_2. Compared with the control sample, as shown in Figure (3), this got a balance in FWHM. The number of residual stresses between the glazing layer of glass and the surface of the substrate ceramic, where increasing the means FWHM residual stresses, showed that the laser treatment does not significantly affect the size of the metal flour and FWHM. Therefore, it is expected that the laser residual layer, between the pressures, will not affect paint and ceramic glaze coating layer and ceramics.

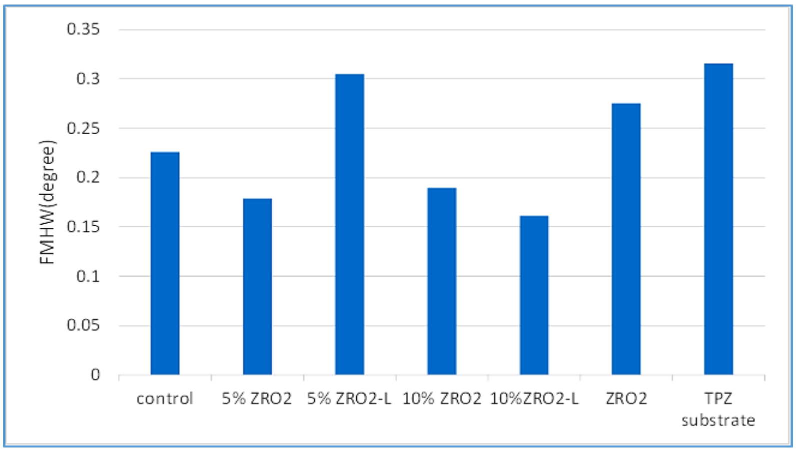

Figure 3: Variation of the full width at half maximum (FWHM) with ZrO_2 addition and laser treatment.

The increase in ZrO_2 percentage up to 10%, in the glaze mixture led to a clear change in crystallite size. In addition, increased up to 45.34nm. This can be attributed to the increased crystalline phases, due to increased ZrO_2 interaction with alumina on the ceramic surface. The doubling of the ZrO_2 percentage has reduced the FWHM to 0.18 in degrees, which means that the residual stresses will decrease. The removal of the residual stresses significantly may be detrimental to the success of the glazing process, in which a certain amount of residual stresses, especially the compressive stresses should be present between the glaze layer and ceramic substrate. This undesirable decline of the residual stresses in the glazing of the ceramic surfaces has processed and improved by laser treatment. This was observed by decreasing the FWHM to 0.16 in degrees with increasing crystallite size up to 53.3 nm.

In the case of a stress-free material, the inter planner spacing d, for a particular reflection (hkl), is constant from one crystallite to another, when it is deformed elastically. The lattice spacing of the crystallite's changes from their stress-free values and causes a shift in the Bragg angle. The strain calculated from this shift is termed the lattice strain. The lattice strain will depend upon the orientation of the reflecting group of the crystallites with respect to the direction of stress [11]. The percentage of lattice strain behavior is like FWHM behavior and the effect of laser treatment is evident on the strain values as shown in Figure (4).

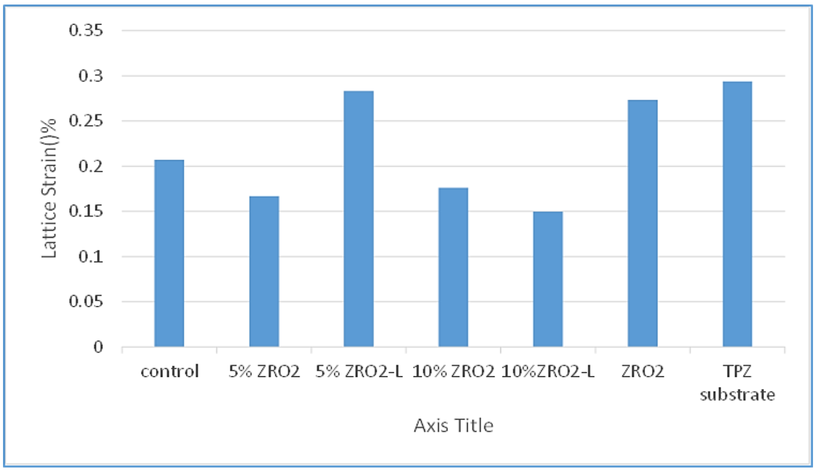

Figure 4: Variation of the lattice strain with ZrO_2 addition and laser treatment.

The average of grain size was determined using the Atomic force microscopy (AFM) device as well as the grain size distribution. Figure (5) (a) showed the statistical distribution grain size to the (TZP-Glazes) control specimen, the grain size ranged between 60 and 120.

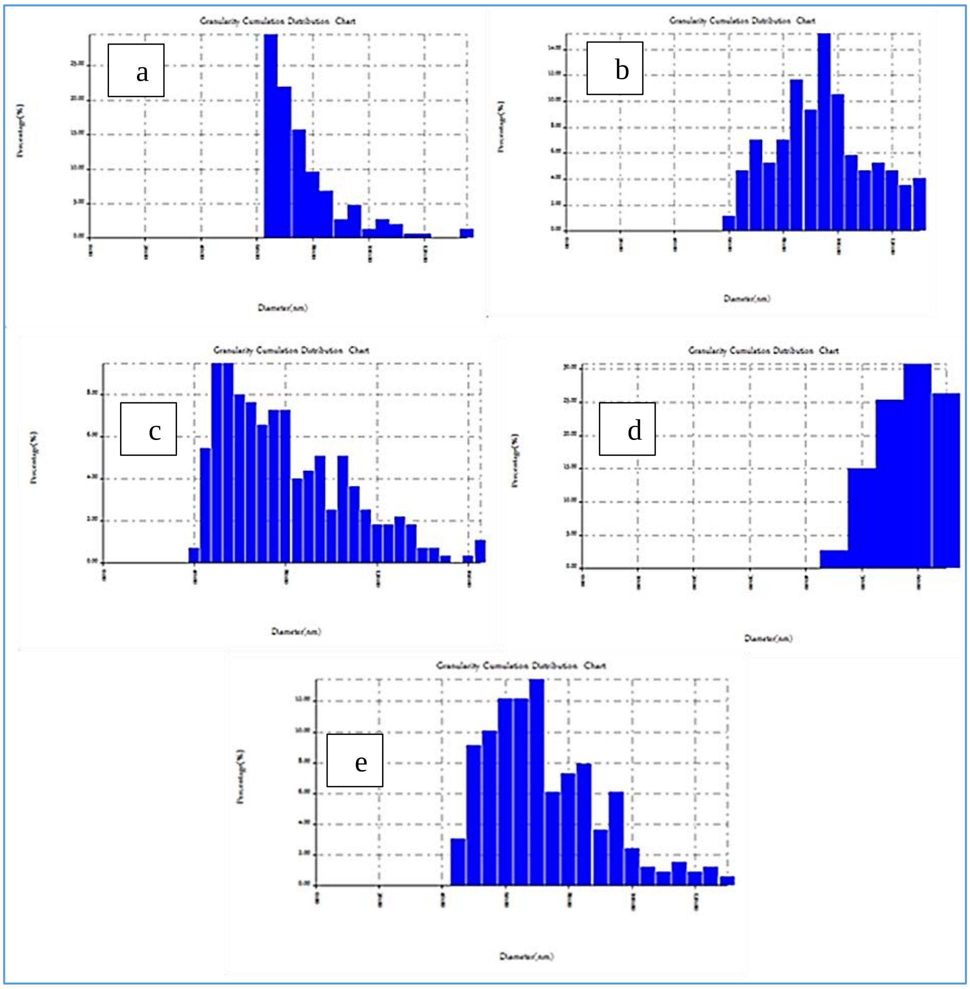

Figure 5: Grain size distribution for: (a) control specimen (TZP + glazes with laser), (b) TZP+glazes+5% ZrO_2, (c) TZP+glazes+5% ZrO_2 with laser, (d) TZP+ glazes + 10% ZrO_2, (e) TZP+glazes+10% ZrO_2 with the laser.

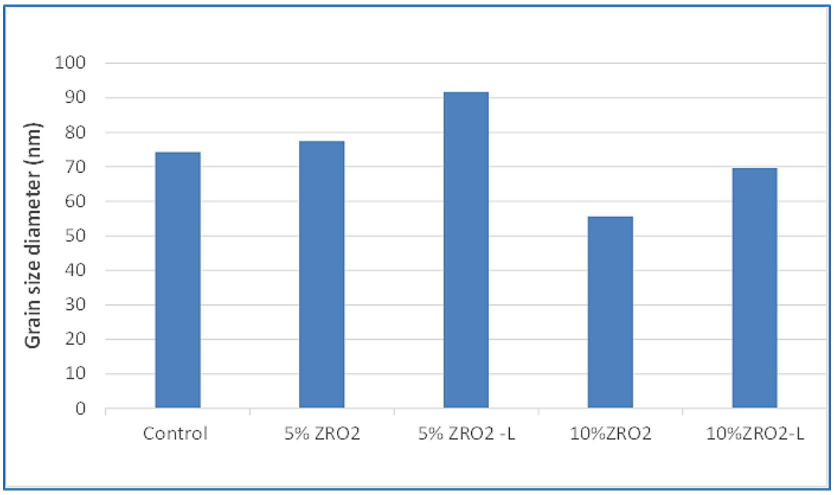

Figure 6: Variation average of the grain size diameter for different specimens.

The statistical distribution diagram of glazes without the addition of ZrO_2 is more skew to the right and the average grain size of this specimen is 74.26 nm as shown in Figure (6). The addition of 5% of Nano ZrO_2 for the specimen (TZP-Glazes+5% ZrO_2), led to a change in the grain size distribution of the glaze layer and led to decreasing the amount of the Nano grains. The effect of the addition seems clear in the distribution of granular sizes to extend from 50 nm to greater than 140nm and the shape of a statistical distribution is like to bimodal and symmetrical due to the wide range of granular sizes, as shown in Figure (5) (b). The average grain size increased up to 77 dues to the emergence of crystallized phases of ZrO_2 reaction with glazing mixture components as shown in Figure 6.

After the treatment by the laser of the specimen (TZP-Glazes+5% ZrO_2 with the laser), there is a clear change in the grain distribution of the glaze surface [12]. The statistical distribution is narrow to a specific range within Nanoscale, which is the ranges from 65 nm to 100 nm as shown in Figure (5) (c). While the average grain size was decreased to 76.4 nm due to the formation of nuclei formed between the glazes and ZrO_2 components with alumina separated on the surface of the ceramic substrate as shown in Figure 6. The statistical distribution of grain size was repeated by adding 10% of ZrO_2, and this distribution with max Nano size granular range less than 100 nm, as shown in Figure (5) (d). The symmetry distribution means more homogeneity in grain size and obtaining uniform surface in granular distribution, and inhibits the continued growth of granular of glaze mixture. The average grain size was at its lowest value of 55.66nm, which is due to increased crystallization due to the increase for ZrO_2, which is an important factor in the nucleation and glass-ceramic formation. The laser effect on the 10% ZrO_2 enhanced glaze was clear and the grain size distribution of the specimen, in this case, is called negatively or left-skewed as shown in Figure (5) (e). So that the extent of the distribution of grain sizes is wider to the range. The average grain size increased up to 69.69 nm, because of the laser on the structure of the glaze layer through the fusion of crystalline nuclei together.

Figure (7) shows the 3D topography profiles under different ZrO_2 additives, where the max high (peak-peak) of the grain size was increased with 5% ZrO_2 compared to the ZrO_2-free coating layer. The value has become 45.32 nm for glaze layer without ZrO_2 and decreased to 6.04 nm when 5% ZrO_2 was added due to the emergence of crystallization nuclei. The laser effect was evident in removing the high peaks of the grains to reach a value of 5.49 nm. The height of the peaks decreased much when added 10% ZrO_2 of the glaze mixture and reach to 3.09 nm. The maximum value of the peak-peak of the grain size was obtained when the 10% ZrO_2 surface was treated with a laser to reach 57.32 nm. This meant that there was a large variation in the grain size distribution due to the grain growth caused by laser treatment.

The roughness was clearly dependent on the ZrO_2 content and laser treatment as shown in Figure (8). The interpretation of the variation in the roughness values corresponded to the change in grain size peak-peak with a grain size average and statistical distribution.

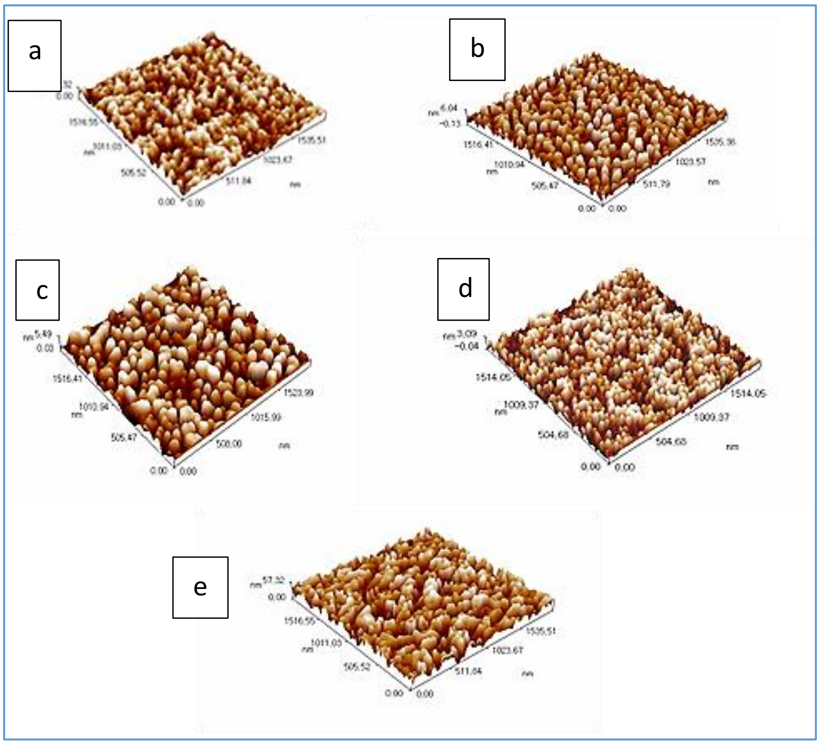

Figure 7. 3D AFM scan data for: (a) control specimen (TZP + glazes with laser), (b) TZP+glazes+5% ZrO2, (c) TZP+glazes+5% ZrO2 with laser, (d) TZP+ glazes + 10% ZrO2,

(e) TZP+glazes+10% ZrO2 with a laser.

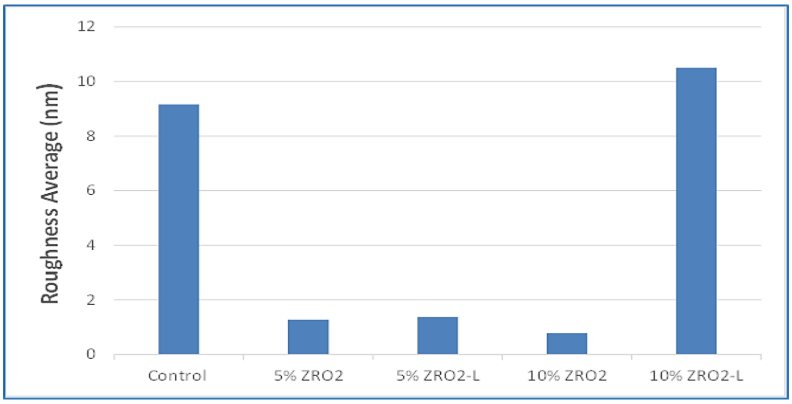

Figure 8. Variation of roughness average for different specimens

Hardness is a measure of the surface roughness of the scratch and wears resistance. The increase in hardness as shown in Figure (9) is due to increased crystalline phases for the added glazes 5% ZrO_2 and irradiated by laser. However, the hardness dropped after adding 10% of ZrO_2 and resumed rising after laser treatment. Increasing hardness is not necessary to increase the toughness of the glazed surface because the toughness is heavily dependent on the grain sizes and distribution. In glazed surfaces, increased hardness can be the result of an increase in residual surface stresses arising from the reaction between the glaze components and ceramic substrate. This is useful if the remaining stresses are in a compressive state and this is due to the difference between the coefficient of thermal expansion of the ceramic substrate and glaze layer [13].

Figures (10) (b), and (10) (d) displayed SEM images of the microstructures of the samples with large grains, the formation of the grain boundary. The appearance of pores before laser irradiation and after irradiation with different laser energies are shown in Figures 10 (c), 10 (e). This indicates that porosity decreased after laser irradiation with Additions of 5% and 10% percentage of ZrO_2 the irradiated region was uniform and homogeneous. It was found that the central part of the treated area is free of cracks. These characteristics increased hardness. After comparing the different laser energies from optical microscopy and SEM, irradiation with laser energy of 15Watt was determined to obtain the best microstructural feature.

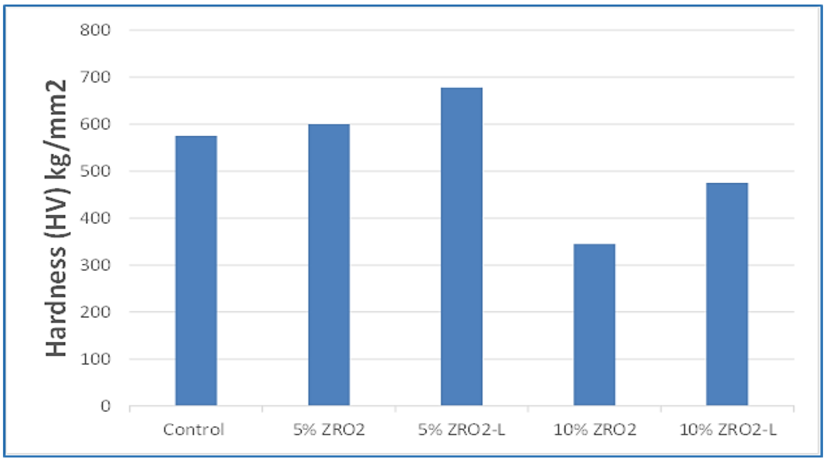

Figure 9: Variation of Hardness for different specimens.

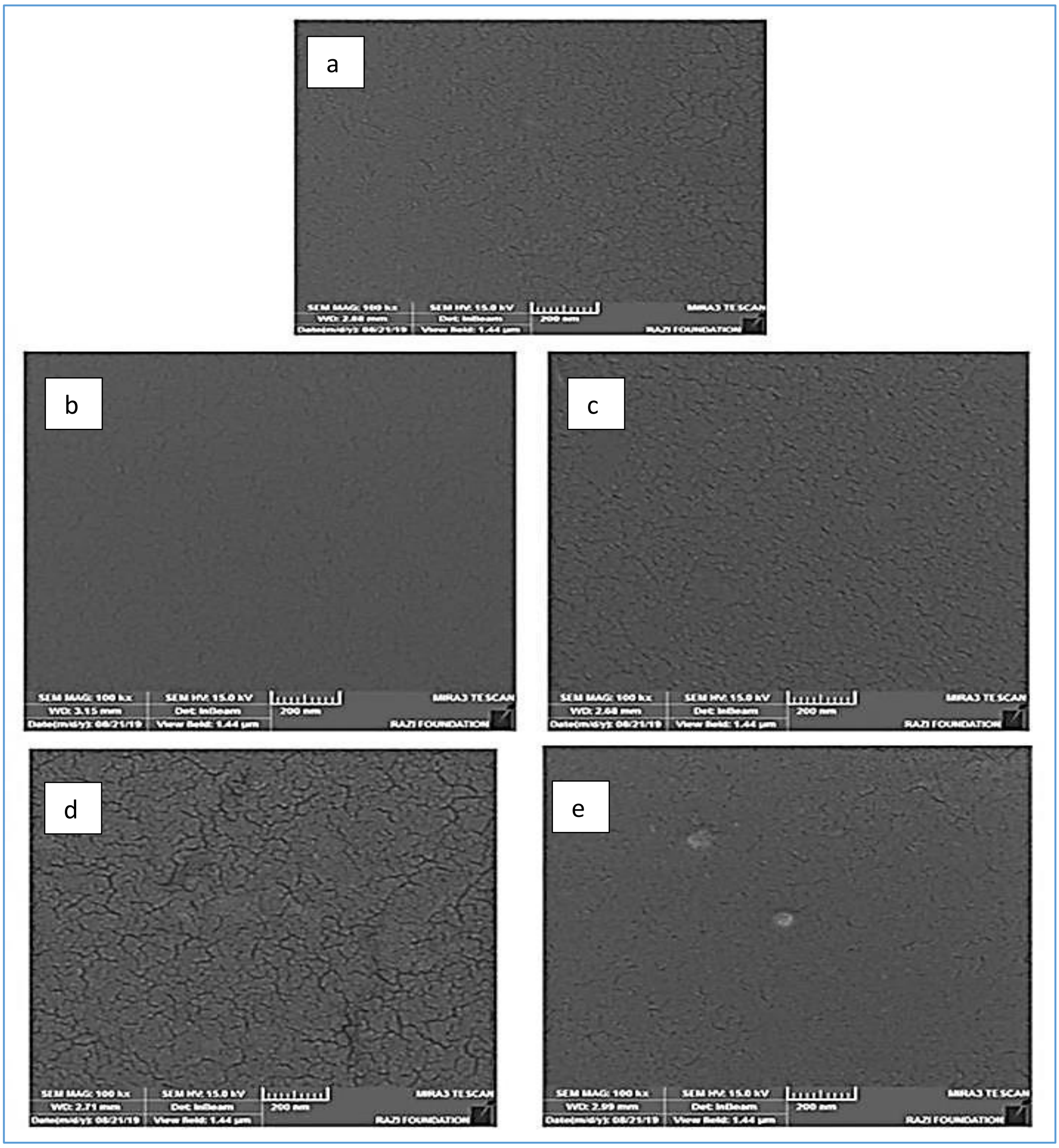

Figure 10: SEM data for a- control specimen (TZP + glazes with laser) b- TPZ+glazes+ 5% ZrO_2 c-TPZ+glazes+5% ZrO_2 with laser15watt d- TPZ+glazes +10% ZrO_2 e- TPZ+glazes+ 10% ZrO_2 with laser15watt

4. Conclusions

The addition of Zirconia is an important factor in the crystallization of the ceramic phase and the appearance of the nucleating agent due to the interaction with the components of the glaze mixture. The alumina in the Zirconia component type TZP. CO_2 laser surface treatment can enhance the appearance of the crystal phase. The appearance of the cover layer of glass-ceramics. In addition to controlling the grain size, especially when a large amount of titanium oxide (10% ZrO_2) added, the laser will increase the grain growth of the glaze layer, so the average grain size will increase.

One of the most important effects of laser irradiation is the control of residual stress. If their values are high due to grain growth or different coefficients of thermal expansion, the laser will reduce these stresses. If the residual stress is very low, the laser irradiation will increase the residual stress and keep the glaze layer under permanent compressive stress, which is the most important factor for successfully completing the glaze processing.

The CO_2 laser can reduce the surface roughness of the 5% ZrO_2 sample. When the amount of ZrO_2 doubles to 10%, it cannot be changed due to the increase of glass-ceramic core centers and the need for higher laser energy or longer irradiation time. The power of 15 W used for polishing, and because the coating layer needs high hardness, the SEM of the sample is displayed after reducing the porosity after laser irradiation and the area is uniform. Therefore, the best condition to achieve this is to add 5% Nano ZrO_2 ceramics, and the power of laser irradiation is 15 watts.

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