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RESEARCH ARTICLE

PRODUCTION AND CHARACTERIZATION OF COLORING LIQUIDS FOR DENTAL ZIRCONIA FRAMEWORKS

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Abstract

The coloring of the dental zirconia prostheses is performed by immersing the pre-sintered zirconia framework in the coloring liquid and waiting for a while. There are various problems that dental prosthesis laboratories experience regarding coloring processes. The most important of these is that the desired color can not be obtained in the dental zirconia framework. This may be due to the different density and porosity values of the zirconia frameworks prepared from different commercial zirconia blocks, the thickness of the framework or the unstable coloring liquids. In the present study, it was aimed to produce coloring liquids suitable for Zirmax brand dental zirconia blocks and characterization of zirconia samples that were colored using these liquids. For this purpose; coloring liquids were produced by dissolving different metal salts, pure water and two different organic solvents. Zirconia samples are colored by immersing the samples into prepared coloring liquids. The colored zirconia samples were sintered at 1500 °C for 2 hours and then the colors of the samples were visually determined by a specialist dental technician. Crystallographic analyzes of zirconia frameworks were performed with X-ray diffraction (XRD) technique. Microstructures of zirconia samples were examined by scanning electron microscope (SEM). According to the color results of the zirconia samples that were analyzed visually, the coloring liquids were successfully produced in accordance with the Ivoclar Vivadent IPS e.max Ceram Dentin A-D color scale.

Introduction:-

Zirconia (ZrO_2) stabilized with 3 mol % yttrium oxide (3Y-TZP) is currently used in dentistry for the fabrication of crowns, fillings, bridges, dental implant abutments and fixed partial dentures as it provides excellent biocompatibility, as well as showing superior strength and toughness in dentistry [1-2]. The production of these zirconia-based dental frameworks is done by milling full or partial sintered blocks using Computer-aided design / Computer-aided production (CAD / CAM) system. The purpose of developing this system is to produce higher quality products of dental zirconia restorations and to obtain these dental products at a more relevant cost [3]. Therefore, some attempts have been made to change the color of Y-TZP ceramics to capture the natural appearance

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of the teeth [4]. The most common coloring method is the immersion of the ZrO_2 prosthesis, which is milled from a white and pre-sintered zirconia block, into the coloring liquid or application of the coloring liquid on the zirconia framework using a brush. This coloring process is an advantage in terms of time and aesthetics [5-6-7].

In the study of Tabatabaian et al., the effects of the thickness values of monolithic zirconia ceramics on the final product color had been investigated, and therefore, the pellets produced by transparent monolithic zirconia powders at different thickness values (0.7, 0.9 and 1.1 mm) obtained from two different manufacturers (DDcubex, CopraSmile), were colored in a coloring liquid corresponding to the A4 scale and CIE L*a*b* values were measured. Based on their study findings, they stated that the thickness of zirconia was effective on the color of the final product. Researchers have determined that zirconia ceramic thickness should be 0.9 mm in order to obtain a color that is acceptable in terms of application (8). Kaplan et al. produced samples of dental zirconia powders, which were made semi-stabilized with mol 3% yttria, in disc form, and they colored these samples in coloring liquids that were prepared using $NiCl_2$, $MoCl_3$ and $NiCl_2$ + $MoCl_3$ metal salts in different concentrations (Weight. 0.1%, 0.25% and 0.5%) for different waiting times (5, 30, 60 seconds), and they examined the effects of the applied process on the mechanical properties of the discs, such as micro hardness, fracture toughness, flexural strength and rate of attrition, and their final colors. Their results revealed that the concentration of coloring liquids had an effect on the final color, while the waiting time in these liquids did not have a significant effect on the final color formation [9].In the study of Ozturk et al., it was observed that the products with different sample thicknesses exposed to coloring liquid had different colors obtained after repeated firing process. They used DC-Zircon and IPS e.max Press branded zirconia framework in their study. With the increase in the thickness of the ceramic, there was a significant decrease in the L* value of the DC-Zircon branded framework and an increase in the a* value, while there was no significant change in the b* value. While IPS e.max Press branded framework showed significant decreases in L* value, they found an increase in a* and b* values [10]. In the study of Kao et al., they observed that coloring liquids containing different amounts of iron oxide formed a color spectrum on the zirconia frameworks, ranging from yellow to red [11]. In the study of Wen et al. using CeO_2 and Er_2O_3 colorants, they found that these colorant species went from yellow to green and yellow to red on the zirconia framework [12]. In their study, Lee et al. demonstrated that the brand of the zirconia block used was effective on the color of the final product [13]. In the study conducted by Son et al, it was noted that the color differences in the final product depended on the thickness of the framework, as well as the block brand [14]. Due to the different porosity and density values of different brand zirconia frameworks, even if coloring is performed with the same coloring liquid, obtaining different results creates a complicated situation in terms of coloring zirconia frameworks, and this situation is a factor that limits the use of coloring liquids in practice. In the present study, the coloring liquids suitable for Zirmax brand dental zirconia blocks were produced and it was aimed to characterize of zirconia samples colored using these liquids.

Materials and Method:-

Preparation of Zirconia Pellets

Ready-to-press 3Y-TZP zirconium oxide powder and a 13 mm diameter press mold were used for preparing zirconia pellets. In order to see the effects of zirconia samples with different thickness on the final color, 0.15, 0.4 and 1.5 g zirconia powder were pressed at 500 PSI, respectively. In addition, 0.4 g zirconia powder was shaped by applying 250 PSI pressure to determine the effect of the forming pressure difference on the final color. After the forming process, the final pressing of all samples was done with cold isostatic press (CIP) at 2000 bar. According to the firing program presented in Table 1, zirconia pellets with CIP treatment were subjected to binder removal and pre-sintering processes and they were made ready for coloring.

Table 1:- Pre-sintering program of zirconia samples.

Program Code	Initial temperature (°C)	Heating rate (°C/min)	Binder burn out temperature (°C)	Dwell time (min)	Heating rate (°C/min)	Pre-sintering Temperature (°C)	Dwell time (min)	Cooling time (min)
PS-1	25	0.5	700	180	5	900	180	175

Preparation of Single-Unit Zirconia Frameworks in Tooth Form

Zirmax brand dental zirconia blocks (produced by Seramdent Dental Ceramics and Nanomaterials Ltd., Eskisehir / Turkey) were used for the preparation of single-unit zirconia frameworks in tooth form where coloring liquids were applied. Blocks were shaped using a milling device with CAD / CAM system.

Preparation of Coloring Liquids

In the preparation of coloring liquids, 5 different coloring salts (CS1-CS5) and distilled water, organic solvent-1 (OS1) and organic solvent-2 (OS2) were used (Table 2). Initially, the color after using only coloring salts in increasing amounts was determined. Then, three different liquids were prepared by dissolving 0.01 g/ml CS1 in three different solvents (distilled water, distilled water + OS1 and distilled water + OS2) in order to examine the effects of solvents used on liquid properties. Finally, 37 different coloring liquids were prepared for A series, 14 for B series, 118 for C series and 35 for D series using coloring salts, distilled water and OS2 according to Table 3.

Table 2:- Raw materials and their properties used for the preparation of coloring liquids.

Raw materials	Brand and Purity
CS1	MERCK
CS2	Rare Metal Compounds, 99.9%
CS3	Unknown, 99%
CS4	Unknown, 99%
CS5	ZAG Chemistry, 98%
Distilled water	-
OS1	MERCK
OS2	A.D.R. Group

Table 3:- Salts and solvents used for the preparation of coloring liquids.

Receipt Code	Raw materials						
	CS1	CS2	CS3	CS4	CS5	Distilled water	OS2
B1	+	-	-	-	-	+	+
B2	+	-	-	-	-	+	+
B3	+	-	-	-	-	+	+
B4	+	-	-	-	-	+	+
A1	+	+	-	-	-	+	+
A2	+	+	-	-	-	+	+
A3	+	+	-	-	-	+	+
A3.5	+	+	-	-	-	+	+
A4	+	+	+	+	-	+	+
C1	+	-	+	-	+	+	+
C2	+	-	+	-	+	+	+
C3	+	-	+	-	+	+	+
C4	+	-	+	-	+	+	+
D2	+	+	+	-	+	+	+
D3	+	+	+	-	+	+	+
D4	+	-	+	-	+	+	+

According to the recipes, organic solvents were added to the 20 ml beaker with a total liquid amount of 10 ml and mixed by the aid of a magnetic stirrer at 500 rpm. Colorized salts weighed separately were added into the beaker in the proportions specified in the recipe and mixed until a clear liquid was obtained. The coloring liquids obtained afterwards were stored in a plastic tube.

Coloring of Zirconia Samples

Coloring process was performed by immersing the samples in the coloring liquid and waiting for the desired time. In order to determine the effect of the holding time in the coloring liquid on the final color, the products in the form of pellets, which were subjected to pre-sintering, were colored in two different coloring liquids (A1 and A3) for 15 seconds and 60 seconds. The coloring of zirconia samples prepared in tooth form was done by keeping them for 60 seconds in the liquid. All the colored products were subjected to the final sintering process according to the program presented in Table 4.

Table 4:- Final sintering program of zirconia samples.

Name of program	Initial Temperature (°C)	Heating Rate (°C/min)	Sintering Temperature (°C)	Dwell Time (min)	Cooling Time (min)
Final sintering	25	10	1500	120	295

Characterization

Crystalline phases formed in colored zirconia samples were detected by using Rigaku brand Rint 2000 model X-Ray Diffraction (XRD) device with CuKα1 radiation. Microstructural analysis of the final sintered samples was performed using FEI NovaNanoSEM650 Brand Scanning Electron Microscope with field diffusion electron gun (FEG) in Kütahya Dumlupınar University İLTEM. Density measurement of zirconia samples was done with Archimedes principle and calculated according to the formula below.

Bulk Density=$\frac{W1}{(W2-W3)} \times P_{water}$

W1: Dry weight of sample

W2: Suspended weight of sample in water

W3: Wet weight of sample

P_{water}: Density of water;

Color analysis of single crown zirconia samples produced by using Zirmax brand zirconia block were performed by Dt. Saim Güler in accordance with IPS e.max Ceram Dentin A-D color scale. Color analyzes were carried out between 12:00 and 14:00 hours, in sunny weather, on a gray ground and using a lighting device that provides 5500K daylight support.

Results:-

When three different liquids prepared to examine the effects of solvents used on liquid properties were examined after 15 days, precipitation was observed in distilled water and distilled water + OS1 containing liquids, while no precipitation was observed in the distilled water + OS2 containing coloring liquid. It was determined that the coloring liquids with precipitation were blurred, and the liquid containing OS2 preserved the clarity as in the day it was produced (Figure 1).

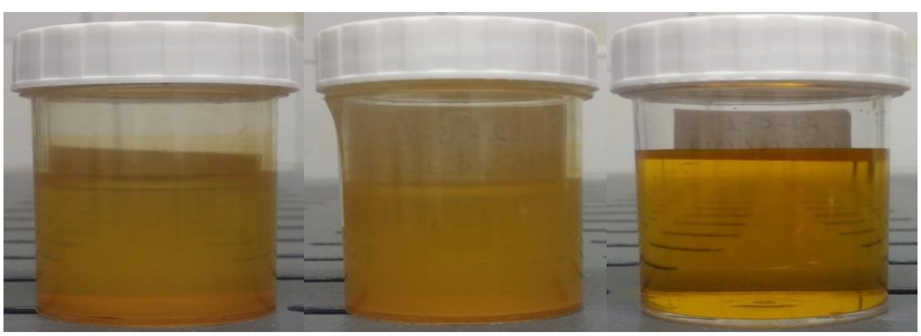


Figure 1:- Images of only distilled water (left), distilled water + OS1 (middle) and distilled water + OS2 (right) liquids after 15 days.

The shelf life of the coloring liquids used in dental prosthesis laboratories and their stability after preparation are extremely important in terms of color. Therefore, it was determined that it would be better to use OS2 in the preparation of coloring liquids. The colors of the zirconia pellets became darker, as expected, due to the use of coloring salts in increasing amounts alone (Figure 2 - Figure 6). According to these results, it was possible to determine the amount of coloring salts that should be used to reach the IPS e.max Ceram Dentin A-D scale.

Amount of Coloring Salt (g/ml)	0.001	0.01	0.05	0.1
Colored and sintered pellets				

Figure 2:- Colored pellets prepared by using CS1-coded salt.

Amount of Coloring Salt (g/ml)	0.001	0.01	0.05	0.1	1
Colored and sintered pellets					

Figure 3:- Colored pellets prepared by using CS2-coded salt.

Amount of Coloring Salt (g/ml)	0.001	0.01	0.05	0.1
Colored and sintered pellets				

Figure 4:- Colored pellets prepared by using CS3-coded salt.

Amount of Coloring Salt (g/ml)	0.001	0.01	0.05	0.1
Colored and sintered pellets				

Figure 5 :- Colored pellets prepared by using CS4-coded salt.

Amount of Coloring Salt (g/ml)	0.001	0.01
Colored and sintered pellets		

Figure 6:- Colored pellets prepared by using CS5-coded salt.

When the cross section surface of zirconia pellets colored at 15 and 60 seconds and sintered is examined, the color absorption has been observed to increase due to the raised coloration time (Figure 7). Therefore, it was considered to color zirconia dental prosthesis materials for 60 seconds for a homogeneous coloring.

(a) (b)

Figure 7:- a) Pellets colored in two different coloring liquids (A1 and A3) for 15 seconds, b) Pellets colored in two different coloring liquids (A1 and A3) for 60 seconds.

The densities of the pellets produced at different weights and pressure values and those subjected to pre-sintering are presented in Table 5.

Table 5:- Density values of pre-sintered pellets produced at different weights and pressure.

Zirconia pellets	Density (g/cm^3)
0.4 gram / 250 PSI	3.14
0.4 gram / 500 PSI	3.19
0.15 gram / 500 PSI	3.19
1.5 gram / 500 PSI	3.19

According to the density results, an increase in density value was observed with increasing press pressure, while the density did not change with increasing pellet thickness. This is due to the better packaging of zirconia powders due to the increase in press pressure.

Although they were colored at the same time (60 seconds), it was found that the colors of zirconia pellets with different weight and thickness were changed. The color obtained as a result of increasing the thickness of the sample had been darkened (Figure 8 and Figure 9). Although all the samples had the same density (3.19 g / cm^3), the relevant color darkening was due to the increase in the distance traveled by the light in the zirconia sample.

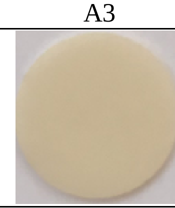
Color series	B1	B2	A2	A3
Colored and sintered pellets				

Figure 8:- Image of pellets produced at 0.15 gram with 500 PSI pressure and colored in B1, B2, A2 and A3 coloring liquids.

Color series	B1	B2	A2	A3
Colored and sintered pellets				

Figure 9:- Image of pellets produced at 1.5 gram with 500 PSI pressure and colored in B1, B2, A2 and A3 coloring liquids.

It was determined that the color of samples was lightened with increasing pressure value used in shaping zirconia samples (Figure 10 and Figure 11). This is due to the increase in the density of the sample with increasing press pressure, in other words, due to lower absorption of coloring liquid as a result of the decrease in the amount of porosity in the sample.

Color series	B1	B2	A2	A3
Colored and sintered pellets				

Figure 10:- Image of pellets produced at 0.4 gram with 250 PSI pressure and colored in B1, B2, A2 and A3 coloring liquids.

Color series	B1	B2	A2	A3
Colored and sintered pellets				

Figure 11:- Image of pellets produced at 0.4 gram with 500 PSI pressure and colored in B1, B2, A2 and A3 coloring liquids.

According to the findings of the visual analysis applied to the zirconia samples that were prepared in the form of teeth, it was determined that all the color series A, B, C and D in the IPS e.max Ceram Dentin AD scale could be obtained successfully with the coloring liquids prepared according to Table 3 (Figure 12-Figure 15).

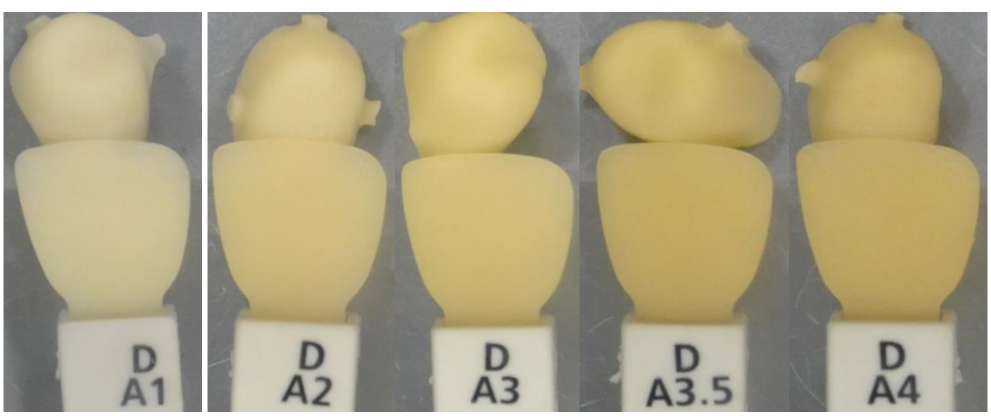


Figure 12:- Samples for A-series according to IPS e.max Ceram Dentin A-D scale.

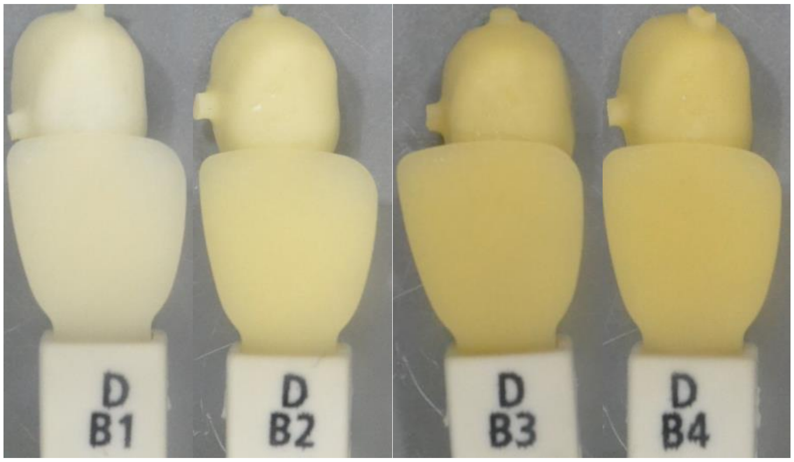


Figure 13:- Samples for B-series according to IPS e.max Ceram Dentin A-D scale.

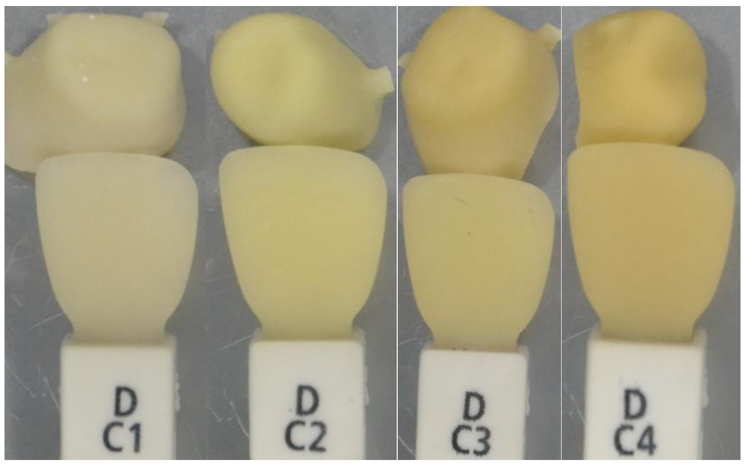


Figure 14:- Samples for C-series according to IPS e.max Ceram Dentin A-D scale.

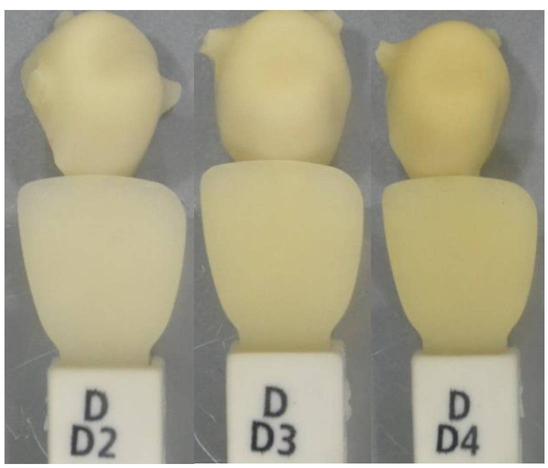


Figure 15:- Samples for D-series according to IPS e.max Ceram Dentin A-D scale.

The results of XRD analysis applied to zirconia samples in colored tooth form are presented in Figure 16-Figure 19. According to the results of XRD analysis, it was determined that the crystal phases in all samples were tetragonal zirconia and coloring liquids had no effect on the crystal structure of zirconia.

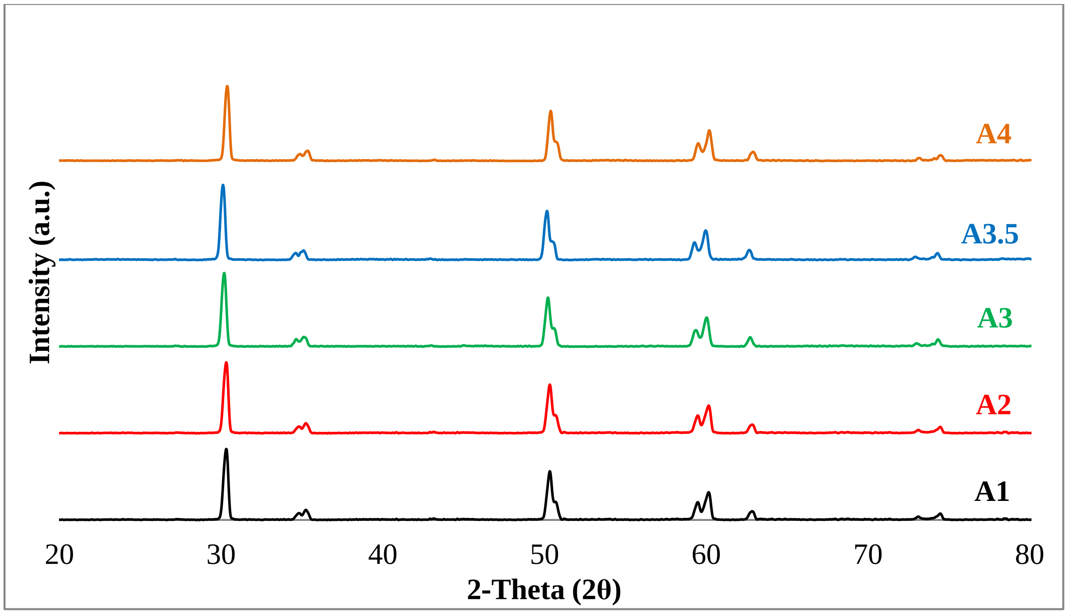


Figure 16:- XRD patterns of zirconia pellets colored with A1-A2-A3-A3.5-A4 coloring liquids (all peaks are tetragonal ZrO_2).

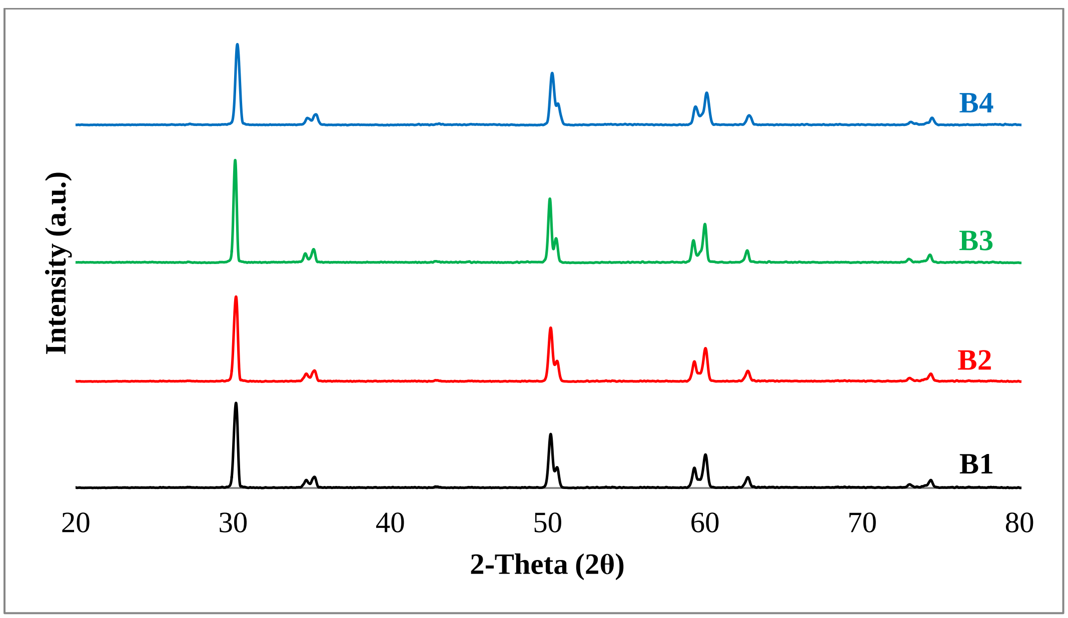


Figure 17:- XRD patterns of zirconia pellets colored with B1-B2-B3-B4 coloring liquids (all peaks are tetragonal ZrO_2).

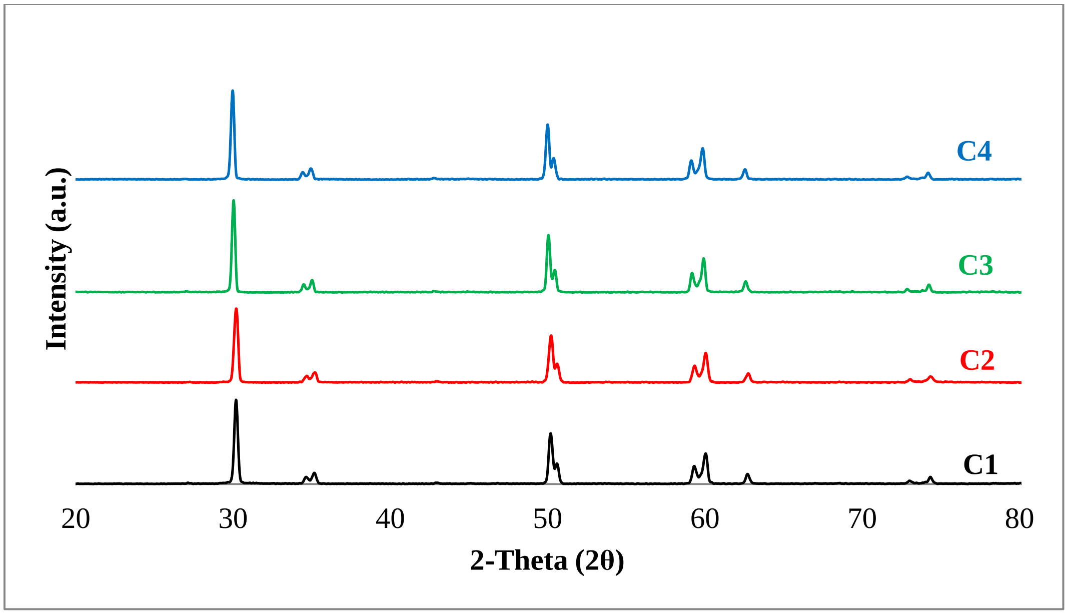


Figure 18:- XRD patterns of zirconia pellets colored with C1-C2-C3-C4 coloring liquids (all peaks are tetragonal ZrO_2).

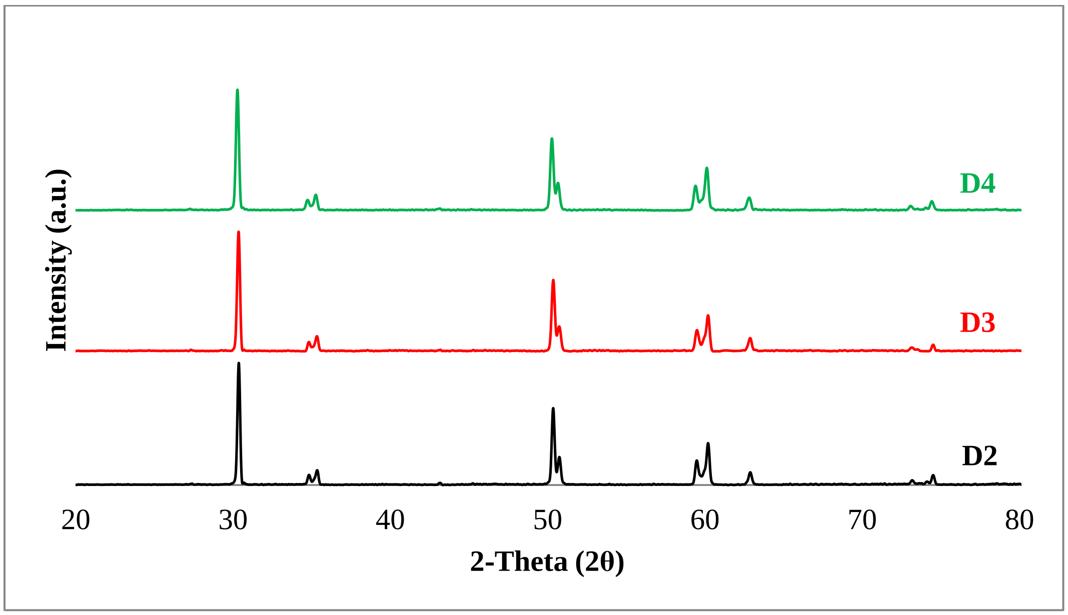


Figure 19:- XRD patterns of zirconia pellets colored with D2-D3-D4 coloring liquids (all peaks are tetragonal ZrO_2).

Final SEM images of sintered uncolored zirconia pellets and those colored with D3 liquid with the highest coloring salt content are presented in Figure 20 and Figure 21, respectively.

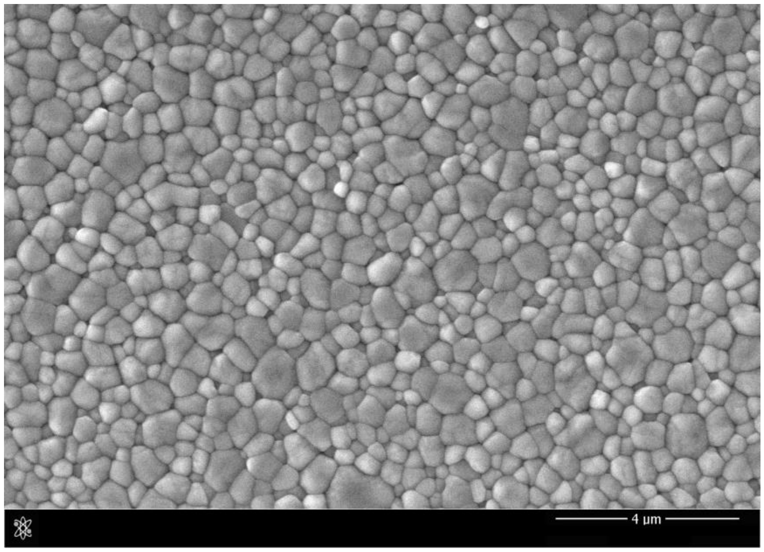


Figure 20:- Final SEM images of sintered and uncolored pellet.

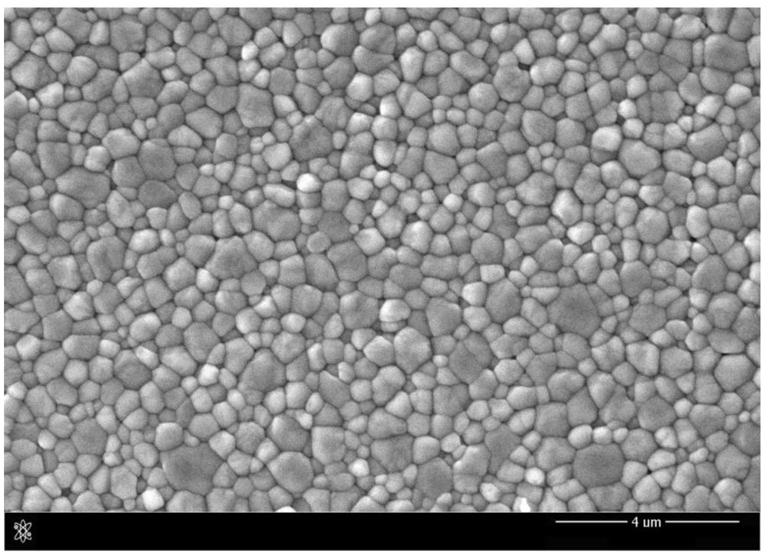


Figure 21:- Final SEM images of sintered pellet colored with D3 coloring liquid.

According to the results of SEM analysis, it is observed that uncolored and colored products are totally sintered and concentrated at 1500 °C. As a result, it was determined that the coloring liquids did not have a negative effect on the zirconia microstructure.

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