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Online Publication Date: 7 Sep 2019

URL: <http://www.jresm.org/archive/resm2019.132me0506tn.html>

DOI: <http://dx.doi.org/10.17515/resm2019.132me0506tn>

Journal Abbreviation: *Res. Eng. Struct. Mater.*

### To cite this article

Topuz M, Dikici B. Two simple methods for surface modification of lithium disilicate dental blocks with hydroxyapatite. *Res. Eng. Struct. Mater.*, 2020; 6(1): 97-104.

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Technical Note

## Two simple methods for surface modification of lithium disilicate dental blocks with hydroxyapatite

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### Article Info

#### Article history:

Received 06 May 2019

Revised 08 Aug 2019

Accepted 05 Sep 2019

#### Keywords:

Lithium Disilicate;  
Calcium Phosphates;  
Dental Block;  
Sol-gel;  
Coating;

### Abstract

Crowns are materials that attracting the attention and lithium disilicates among them widely used because of their high bulk mechanical features. Increasing the bioactivity of crowns has generally been a common aspect of all studies. In this study, lithium disilicate dental blocks were coated with bioactive calcium-phosphate components using sol-gel dip-coating methods to increase their surface bioactivity. Two coating methods called as slurry and powder methods were proposed to obtain dense and remarkable layer on the lithium disilicate substrates. The coating layers were characterized by scanning electron microscopy attached with energy dispersive spectroscopy. Coating procedures have successfully accomplished and show enhanced surface properties with terms of calcium/phosphate ratios. Tri-calcium phosphate obtained by the slurry method, which has 1.5 calcium/phosphate ratio, is a favorable structure for bio-absorbable. However, the powder method has approximately 1.67 calcium/phosphate ratio which necessary for hydroxyapatite (HA) structure for higher crystallinity and lower solubility properties.

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## 1. Introduction

In the area of dentistry, highly detailed studies have been carried out for the restoration of the teeth that have been decaying, ageing or lost their function. As the technology progresses, always there is another innovation in the field of health. Especially in recent years, CAD/CAM technologies which have come into our lives have caused many developments in the field of dentistry. With the help of the new computer-aided image and design technology developed within this scope, it is possible to have new teeth in a short time like 40min [1]. The crown is a method of restoring damaged teeth for various reasons. Crowns can be used as an implant superstructure to prevent weakened teeth from cracking, repair a broken tooth, make a bridge, overcoat a lost or deformed tooth due to excessive caries. It can be made of full-metal, metal-backed porcelain or full-porcelain. Crowns are prepared according to the model. If porcelain is used, the natural tooth tone is determined and the support adheres to the tooth [2]. Ceramic dental materials used in CAD/CAM processes are classified according to their chemical content; feldspathic, low leucite-containing glass, lithium disilicate and mica reinforced, alumina and magnesium ceramics. The lithium disilicate consists of several randomly interleaved layers. In terms of fracture resistance, the needle-like crystals prevent both cracking and divergence of the cracks [3].

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DOI: <http://dx.doi.org/10.17515/resm2019.132me0506tn>

Res. Eng. Struct. Mat. Vol. 6 Iss. 1 (2020) 97-104

On the other hand, the biggest advantage of lithium disilicate crowns is that it does not need any reinforcing elements such as metal or zirconium in its infrastructure. In other words, they are kind of a porcelain type that has been made more than 3 times more durable than classical porcelain. So, what brings us to this increase in endurance? The fact that a prosthesis can be made without the use of a substructure gives us a great advantage in mimicking the characteristic appearance of natural teeth under the light. Besides, the fact that it can be worked with thinner means that less tissue will be removed from the teeth on which it will be made. According to the location of the prosthesis to be made, dentists had to make difficult decisions to be durable or aesthetic [4].

Calcium phosphates have the ability to increase bone formation in the surrounding tissue and implanted in the human skeletal system. They are used extensively as coating components due to the higher osteoblast orientation capabilities. The apatite compound of calcium phosphates is almost identical to the mineral phase of bone and tooth, especially HA [5].

Many techniques can be used for the coating of ceramic dental blocks such as sol-gel dip-coating, plasma electrolytic oxidation and plasma spray [6-8]. However, among these techniques, sol-gel dip-coating has easier process steps and also cheaper than others.

A similar study was done on zircon blocks by Azari et al. [9] and compared thermal coating and air abrasion methods. As a result of the study, the crystalline HA structure was found in the thermal coating method while in the air abrasion method HA mini-islands were encountered. Another research conducted by Azari et al. [10], suggested that HA coatings on zirconia blocks could be a solution for increasing the adhesion strength of resin cement. In another study on bone tissue formation, it was observed that HA coatings on zircon block contributed to the formation of new bone tissue [11]. It has been stated by many researchers that there will be no problems with adhesion resistance at the abutment-crown interface with the use of hydrofluoric acid [12,13]. However, different surfaces are being studied to investigate the living cell reaction at the interface [14]. Therefore, it has become necessary to investigate whether the bioactivity can be increased by coating lithium disilicate surfaces.

In this study, lithium disilicate dental blocks were coated with calcium phosphate components to increase their surface bioactivity. Two different coating methods (called as a slurry (LD-S) and powder (LD-P) methods) were proposed to obtain dense and remarkable layer on the lithium disilicate substrates. The synthesis steps are discussed in details, comparatively.

## 2. Materials and Methods

Lithium disilicate materials with dimensions 14.4×14.4×18mm were used as a substrate for coating procedures (Ivoclar IPS e.max CAD). The standard structure of blocks was; 57.0-80.0% SiO<sub>2</sub>, 11.0-19.0% Li<sub>2</sub>O, 0.0-13.0% K<sub>2</sub>O, 0.0-11.0% P<sub>2</sub>O<sub>5</sub>, 0.0-8.0% ZrO<sub>2</sub>, 0.0-8.0% ZnO, 0.0-10.0% other oxides (Al<sub>2</sub>O<sub>3</sub> and MgO) and consists of 0.0-8.0% color oxides by means of weight. First of all, the samples were sandblasted by Al<sub>2</sub>O<sub>3</sub> particles of 150µm. The aim of sandblasting is to obtain better wettability between substrates and coating layer. Subsequently, the surface cleaning process was carried out in distilled water (DW) for 15min in order to remove surface residues after sandblasting.

Two different coatings procedures (LD-S: slurry) and (LD-P: powder) were used to examine the characteristics of HA coatings synthesized on the base materials. The first one is the dipping into the slurry solution and the other is powdered and sintered HA's into a solution and the surface is coated with a dipping process. Agar was used in order to obtain a higher wettability on the surface. Synthesis of the LD-S coatings, the sol-gel method was

used in which low sintering temperatures were sufficient, allowing complex shapes to be easily coated. In the synthesis of the sols, ethyl alcohol (Sigma-Aldrich), calcium nitrate tetrahydrate (Calcium source:  $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ , Sigma-Aldrich), di-ammonium hydrogen phosphate (Phosphate source:  $(\text{NH}_4)_2\text{HPO}_2$ , MERCK) and agar (MERCK) were used as precursors. The amount of precursors which used for procedures was determined from the Ca/P stoichiometric ratio of HA. 12.619g of calcium nitrate tetrahydrate and 4.457g of phosphorus pentoxide were dissolved in two separate 500ml breakers with 100 and 120ml DW, respectively. After one hour stirring 75ml ammonia was added to  $(\text{NH}_4)_2\text{HPO}_2$  solution and stirred 10min more. Then Ca solution, 1.708g agar (1wt%) was added to P solution slowly, and 10min stirred. The mixed solution was heated until boiling and stirred 10min while boiled. The boiled solution was waited in order to the aging of sol. for 24h. Unlike the LD-S procedure, LD-P procedure has different steps. Firstly, the slurry solution (LD-S procedures last step) filtered for 1 day to squeeze the excess amount of ammonia in the solution. Then dried in an oven at 200°C for 4h, to evaporate the residual ammonia and organic components in the wet cakes, and pulverized with the aid of a mortar. The CaP powders were sintered at 1000°C for 1h. 2g CaP powder was mixed for 2h in 50ml DW for obtaining coating sol. After these steps, both sols were homogenized ultrasonically for 20min at 25% duty rate to obtain homogenous sol. before dipping. The samples were dip-coated for three times to obtain thicker films on its, and every dip-coating steps were pre-dried at 40°C for 5min. Finally, samples were sintered for 2h at 300°C for LD-S and 200°C for LD-P procedures. In pre-sintering trials, small burn marks on the surfaces of lithium disilicate materials were effective in the selection of sintering temperatures. In the LD-S procedure, the sintering temperature is determined as 300°C, because the reactions between Ca and P can be occur. In the LD-P procedure, since the powders were sintered at 1000°C before coating, 200°C sintering temperature was considered sufficient. The algorithmic flow chart of both procedure is presented in Fig. 1.

Phase formations in the coatings were investigated by X-Ray diffraction (XRD, Bruker D8). XRD analyses were performed on a stage which using a radiation wavelength  $\lambda=1.54056\text{\AA}$  X-ray source with a scan rate of  $0.06^\circ\text{s}^{-1}$  between 10 to 90°. Microstructural characterization of the coatings was performed using scanning electron microscopy (SEM, JEOL JSM-6335F) and energy dispersive spectroscopy (EDS, Oxford Inca). Because the coatings are ceramic-based, they were coated with Au-Pd alloy prior to the examination to improve the image quality for SEM and EDS examinations.

#### 4. Results and Discussions

SEM surface morphology and EDS result of LD-S coated samples are given in Fig. 2. When the SEM surface morphologies are examined, it is seen that there are micron-size pores in some parts of the coating structure. It is thought that these micron-sized pores formed because of the lack of surface wetting by the LD-S solution. It was also mentioned by other researchers that the nano-sized crystal structures in the pore were lithium disilicate [15]. Researchers reported that the nano-sized lithium disilicate crystals which noodles-like shaped could improve the strength of the material because of the different orientation of these crystals [3]. This situation was also determined by EDS analysis that the electron energy distribution of Si element was observed at 1.739 keV.

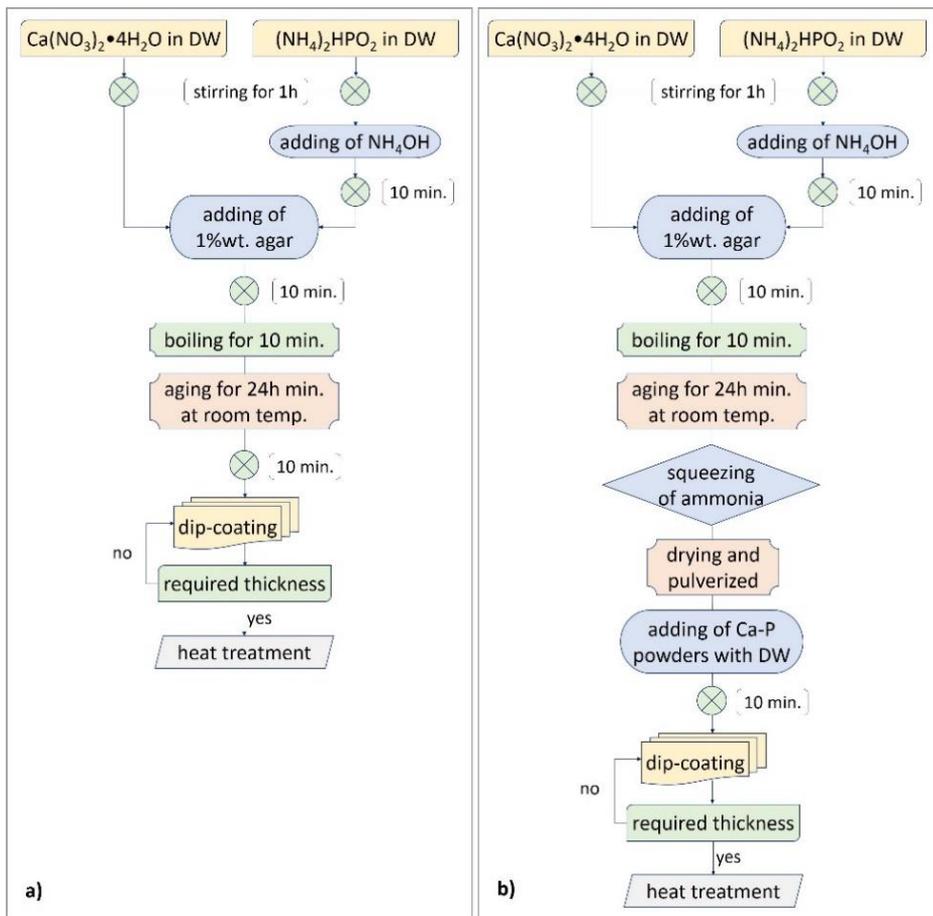


Fig. 1 Algorithmic flow charts of (a) LD-S and (b) LD-P synthesis of HA sols

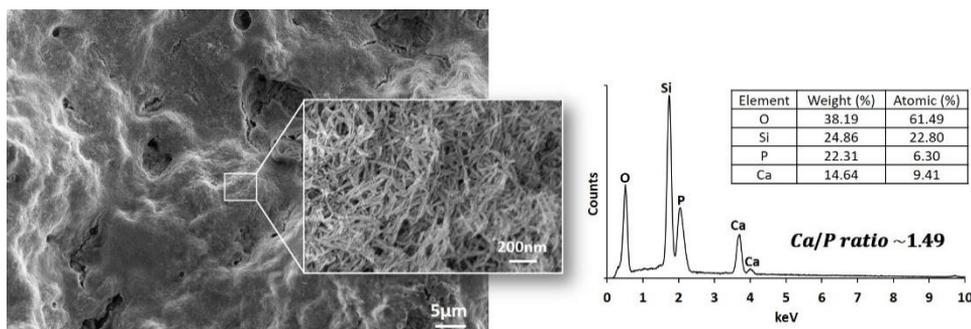


Fig. 2 SEM surface morphology of LD-S coated sample and EDS result

On the other hand, calcium and phosphate elements on the surface have been proven by EDS analysis, and the Ca/P atomic ratio (~1.49) was determined to be tri-calcium phosphate ( $\beta$ -TCP, Ca/P ratio 1.5) [5]. It has been mentioned by many other researchers that this  $\beta$ -TCP structure has lower crystallinity compared to the HA structure and resulted in higher solubility but has similarly improved osseointegration properties [16]. The low Ca/P ratio and low crystallinity values achieved may be due to the fact that coating components are made at low sintering temperatures (300°C for LD-S). On the other hand,

high sintering temperatures to be used in the coating affect the long-term service life as it causes permanent damage to the substrate or changes in the grain structure.

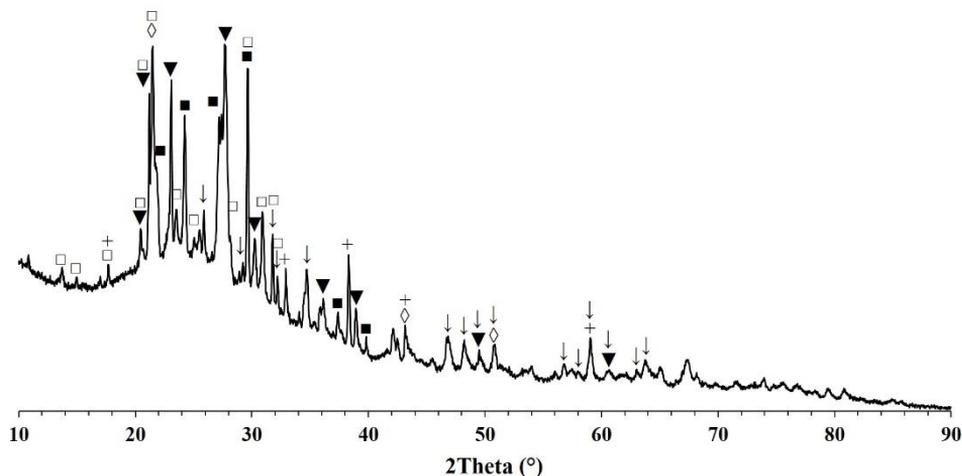


Fig. 3 XRD result of LD-S coated sample, legends are; ↓ HA, ◊ Cristoballite (SiO<sub>2</sub>), ■ Li<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>, ▼ Trdymite (SiO<sub>2</sub>), + Li<sub>2</sub>SiO<sub>3</sub>, □ β-TCP

XRD analyze of LD-S coated samples was given in Fig. 3. As shown from XRD result of LD-S sample, β-TCP structure was confirmed with 09-0348 JCPDS card number. This result agrees with EDS analyze result. LD-S coated samples show small amount of HA peaks (09-0432) which may be observed as it is sintered at the β-TCP-HA transition temperature. On the other hand, different phases such as Cristoballite (SiO<sub>2</sub>), Trdymite (SiO<sub>2</sub>), Li<sub>2</sub>Si<sub>2</sub>O<sub>5</sub> and Li<sub>2</sub>SiO<sub>3</sub> were found from the base material (JCPDS card numbers are; 01-0424, 01-0378, 040-0376 and 010-0017 respectively). It is thought that phases which contain lithium and silicate come from the substrate materials, due to the inhomogeneous surface morphology of the coating.

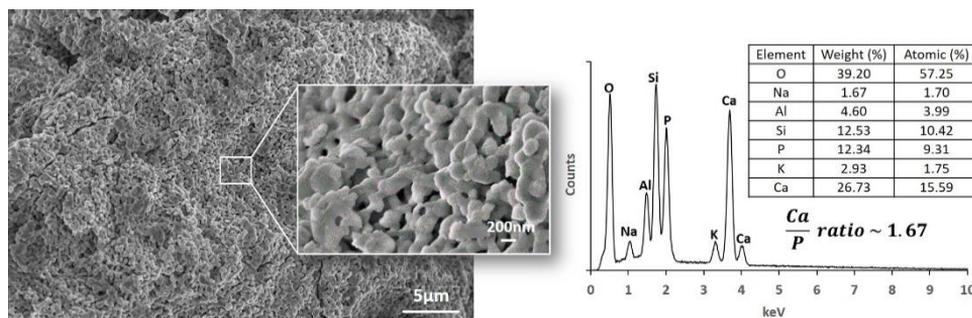


Fig. 4 SEM surface morphology of LD-P coated sample and EDS result

The SEM surface morphology and EDS results of the coating synthesized by the LD-P method are shown in Fig. 4. It can be shown that from Fig. 4 the surface is smoother than LD-S and has nano-sized pores. These nano-sized pores may responsible for enhanced biocompatibility. Unlike the LD-S method, the Ca/P ratio was measured as 1.67 in this method, atomically. In order to determine whether HA which another calcium phosphate structure is formed, the researchers decide either the Ca/P ratio or the XRD test. As it is known from the literature, the Ca/P ratio of HA is 1.67 and it is the same as the value obtained in this method [17,18]. It was determined that the Na, Si, and K elements of the

EDS results from the blocks which used as substrate material [2], Al element was determined due to the blasting process applied before the coating. The high crystallinity of the HA shows that it will not dissolve easily and have a long life in the physiological environment. The surfaces similar to the surface of the LD-P were also obtained by other researchers [19,20].

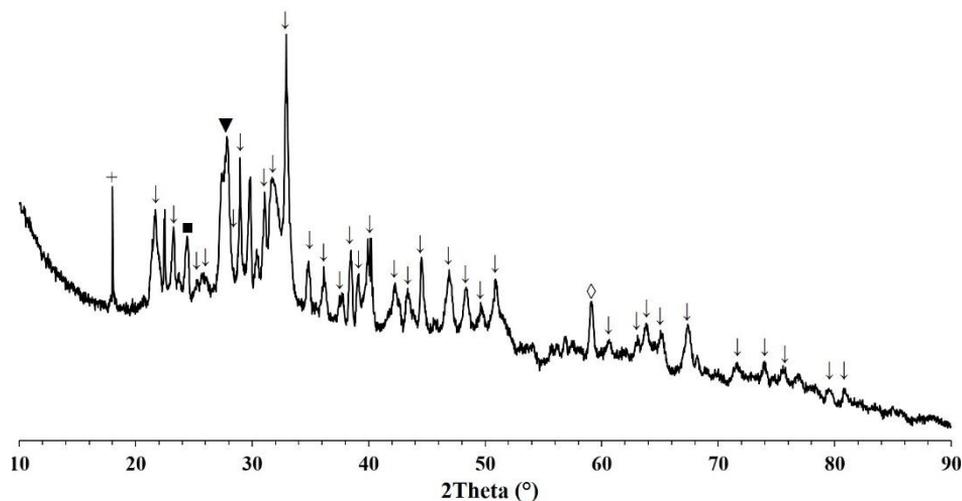


Fig. 5 XRD result of LD-P coated sample, legends are; ↓ HA, ◇ SiO<sub>2</sub>, ■ Li<sub>2</sub>Si<sub>2</sub>O<sub>5</sub>, ▼ SiO<sub>2</sub>, +Li<sub>2</sub>SiO<sub>3</sub>

Phase formations and transformations (XRD result) of LD-P coated samples are presented in Fig. 5. These results were in agreement with SEM/EDS results. When compared with XRD results of LD-S samples, it is seen that XRD peak widths of LD-P samples are narrower and this may be caused by pre-sintering process applied in LD-P method. Unlike LD-S method's XRD result, LD-P coated sample has completely HA phases on it.

Findings from the studies emphasized that agar causes Ca deficiency in HA structure [21]. Ouyang et al. [22] were found that the amount of agar decreased to about 30% by weight, but not completely decomposed by heat treatment at 300°C. The difference in Ca/P ratios between LD-S and LD-P procedures is thought to be due to the agar's removal from the coating composition after 1000°C of heat treatment for powdering in the LD-P procedure. As can be seen from the researches, the sintering temperature of the LD-S procedure was 300°C, did not completely decompose the agar from the coating composition. Thus, it is evident that a small amount of agar remaining in the coating composition leads to Ca deficiency and a low Ca/P ratio. Besides SEM/EDS analyses, phenomenon which formation of both  $\beta$ -TCP and HA, proved by XRD results. Li<sub>2</sub>Si<sub>2</sub>O<sub>5</sub> was major phase from substrate materials but there is a small amount of SiO<sub>2</sub> and Li<sub>2</sub>SiO<sub>3</sub> peaks which may occur nucleation of Li and Si during sintering. It's thought that such higher intensities, as can be seen in XRD results, are favorable to human body due to the lower soluble behavior.

Also, as Narayanan et al. [23] mentioned,  $\beta$ -TCP stated that it could be absorbed in the body and HA could not be absorbed. While absorption in the body is a more important condition, due to the unpredictable dissolution behavior of  $\beta$ -TCP, HA which has high stability properties, is more desirable. After all these results and discussions, the LD-P procedure is thought to be an alternative coating procedure for lithium disilicate ceramics due to its HA structure and surface homogeneity. HA formation on LD-P coated samples were proved by XRD results. In addition, it is thought to have a positive effect on cell growth at the

abutment–crown interface with its optimal Ca/P ratio and will not cause any adverse reactions.

## 5. Conclusion

In this study, lithium disilicate blocks used in the field of dentistry were investigated by using two different methods and their ability to form a possible calcium phosphate component was investigated. The coatings obtained in the LD-P method were found to be more preferable both as surface morphologies and Ca/P ratio. In the literature, there are a few studies which conducted on surface morphologies of HA coatings on lithium disilicates but it is suggested that osteoconductivity or osteoblast formability tests of the coatings synthesized by both methods will be made and investigated in the future.

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