

Parametric Study for the Effect of Suspension Composition on Electrophoretic Deposition of Zirconia

Alaa Abdul hasan Atiyah

Department of Materials Engineering , University of Technology
dralaadelta@gmail.com

Saad Badry Hasson Farid

Department of Materials Engineering , University of Technology
Dr.SaadBHF@gmail.com

Awham Jumah Salman

Babylon Technical Insitute ,Al-Furat Al-Awsat Technical University
awhamjumah@yahoo.com

Abstract

Electrophoretic deposition (EPD) is continued to be a major activity of science and technology in view of its advantages of controlling the deposit thickness, morphology, and microstructure. The method enables the formation of films on substrates of complex geometry that suits for various applications including biomaterials. However, the method involves controlling of various process parameters, which are of vital effects on the properties of the deposit.

Besides, deposition of zirconia nano particles is vital for production of parts used in restorative dentistry. This work was a study the effect of polyethylene glycol polymer as a binder-suspension agent, the amount the solid loading (zirconia particles), and the effect of the toluene as a dielectric liquid, on the final thickness and green density of the deposited parts.

Keywords: Electrophoretic deposition, Polyethylene glycol, Stabilized zirconia, Green density.

الخلاصة

الترسب الكهربائي (EPD) لا يزال احد الانشطة الرئيسي للعلوم والتكنولوجيا نظراً لمزاياه في السيطرة على سمك الترسيب، والتشكل، والبنية المجهرية. تمكن هذه الطريقة من تشكيل الأغشية على الأسطح الأشكال الهندسة المعقدة والتي تكون ملائمة لتطبيقات متنوعة بما في ذلك المواد الحيوية. ومع ذلك، فإن الطريقة تتضمن السيطرة على عوامل العملية المختلفة، والتي تكون ذات تأثيرات حيوية على خصائص المادة المترسبة.

إلى جانب ذلك، ترسب دقائق الزركونيا النانوية أمر حيوي لإنتاج الأجزاء المستخدمة في طب الأسنان التعويضي. هذا العمل هو دراسة تأثير بولمر البولي إيثيلين جلايكول كعامل تعليق، ومقدار المادة الصلبة (دقائق الزركونيا)، وتأثير التولوين كسائل عزل كهربائي، على السمك النهائي والكثافة الخضراء للأجزاء المترسبة.

الكلمات المفتاحية: - الترسيب الكهربائي، البولي إيثيلين كليكول، الزركونيا المثبتة، الكثافة الخضراء.

Introduction

The fabrication of thin layers of zirconia-based materials has attracted great attention because of the wide number of applications in the fields of electro-ceramics and structural ceramics. Partially stabilized zirconia (PSZ) are used as ionic conductive oxides for the development of electrochemical devices such as solid oxide fuel cells, oxygen sensors, etc., whereas, tetragonal zirconia polycrystalline (TZP) is used as a structural material due to its high toughness and modulus of rupture (Ferrari and Morenoz, 2000; Peng and John, 2013; Albert, 2009). Colloidal forming methods such as slip casting or pressure casting are widely described for the manufacturing of tailored

microstructures of either PSZ or TZP. These include physical, chemical, or electrochemical vapor deposition methods, electrodeposition, plasma technologies, etc. It is also possible to shape these ceramic/ ceramic or ceramic/metal structures using the colloidal approach by means of the electrophoretic deposition (EPD) method. (Van *et.al.*, 2006 ; Bailey *et.al.*, 2017)

The electrophoretic deposition (EPD) is a suitable technique for manufacturing ceramic pieces with complex geometry shapes. With EPD, ceramic bodies are shaped directly from a stable colloidal suspension by a electric field (Linda , 2014; Pouya *et.al.*, 2015; Caproni *et.al.*, 2011; Laxmidhar *et.al.*, 2008). The mechanism of EPD has two main steps: firstly, a DC electric field is applied to a suspension of the ceramic particles forcing the charged particles to move towards an oppositely charged electrode; secondly, the particles are deposited onto the electrode producing a relatively dense and homogeneous compact or film (Stefan *et.al.*, 2014; Ferrari and Moreno , 1997).

Suspension preparation is a crucial step in this technique. It is very important to consider the solvent- dispersant – binder system, in terms of the solubility of binder and additives, the chemical compatibility of the components (Shaohua *et.al.*, 2005). According to this, the EPD process requires the control of the suspension properties as well as the selection of the electrical parameters involved during forming (Ferrari and Moreno, 1997).Suspension parameters are bound to the bath composition and determine the transport properties (particle charge or zeta potential, viscosity, conductivity) and the structure and cohesion of the green deposit (Fre´de´ric and Alain , 1999)

It is well known that the packing density of ceramic green parts is of utmost importance in the production of advanced ceramics. For example, the particle packing density in green parts determines the sintering shrinkage, the density of the sintered ceramics, and the number of defects. In order to fully exploit the advantages of the EPD process, these factors that affect the structure of the green parts need to be better understood (Sasa and Katja, 2009; Eduardo *et.al.*, 2013).

In our work, we investigated the effect of the suspension's composition on the properties of the deposits formed. Our goal was to determine the composition for effective and reproducible suspension for fabrication of zirconia, and hence to promote industrialisation of the process. We analysed the effect of polyethylene glycol in ethanolic suspensions.

Materials and methods

For the synthesis of zirconia samples, 3 mol% Y₂O₃–ZrO₂ (Hongwu International Group Ltd; Chain; average particle size 70-80 nm) powder was used. Ethanol (ScharlabS.L.,Spain) was used as the suspension medium. Polyethylene glycol 4000 Mw (sinopharm chemical reagent Co.,Ltd ,Chain) was added as a binder material and to enhance particle charging. Toluene (Avantor Performance Materials B.V., Germany) was also used to study suspension properties.

Ethanol-based suspensions for the production of dense samples were used. Ethylene glycol (PEG 4000) was added to the ethanol and must continue to mix by a magnetic stirrer (SH-2 model) to complete dissolve. Then the ceramic powder was added to the solution and continues to mix in order to formation the suspension.

A power supply (H. T. model EISCO) was used as a DC supplier in the electrophoretic deposition process. The electrophoretic cell was a 100ml cylindrical glass beaker; and the electrodes were stainless steel (316L) plate with dimensions (2.5 × 2 ×

0.1) cm. The distance between the electrodes was 1.2 cm. A variable voltage (250-290V) is used in the course of the process, explicitly, the starting voltage was 250V and increased by 10V every 5 minutes, i.e. the total process time is 25 minutes.

Three groups of suspensions were prepared in this work in order to study the effect of the powder weight on green density and thickness of parts. The first group was of a constant amount of PEG (1g) for a 100 ml of ethanol and varied weight of zirconia (1.2, 2 and 4 g) a suspension of zirconia in ethanol (4g for each 100 ml) . The second group was of a constant amount of zirconia (4g) for a 100 ml of ethanol and varied weight of PEG (0, 1, 1.5 and 2 g). The third group was of a constant amount of PEG (1.5g) and 1.2g zirconia for a 100 ml of solution from ethanol and varied amount of toluene (0, 6 and 10) ml.

All dimension of the deposits were determined with a micrometer (TERMA,0-25 mm /0.001mm).the weight of deposits were determined with a Digital Scale (DM.3,500g×0.01g).

The weight of a unit volume of deposited part expressed in grams per cubic centimeter is the green density of the deposited part. It is calculated from dimensions evaluations and the weighting of deposited part as follows (Goupee, 2005):

$$\rho_g = \frac{m_g}{V_g}$$

Where (ρ_g) green density (g/ cm³) , (m_g) green mass of the deposit (g) and (V_g) volume of the deposit (cm³).

Results and Dissuasions

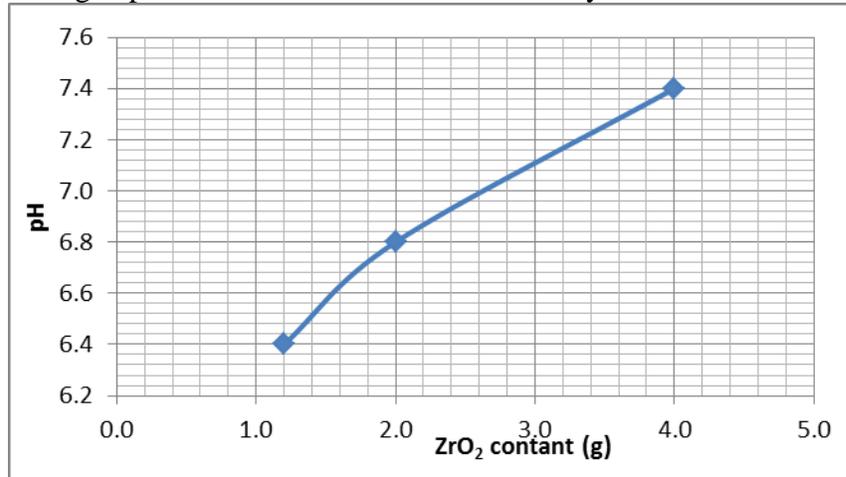
Applying constant voltage (250V) in the electrophoretic process results in a zirconia deposit with the formation of cracks after the drying process as seen in figure (1-A). These cracks occur due to various green density of the deposited zirconia. Thus, it is decided to use an incremental increase of the applied the voltage as the thickness of the deposit increase (10V increase every 5 min). This method of applying variable voltages is not preceded by other works as far as we know. The resultant deposit was free of cracks after the drying process; figure (1-B) which indicates a homogeneous deposit with higher green density.



Figure (1): Effect the type of DC voltage; A- constant voltage (250V), B- Variable voltages (250-290 V).

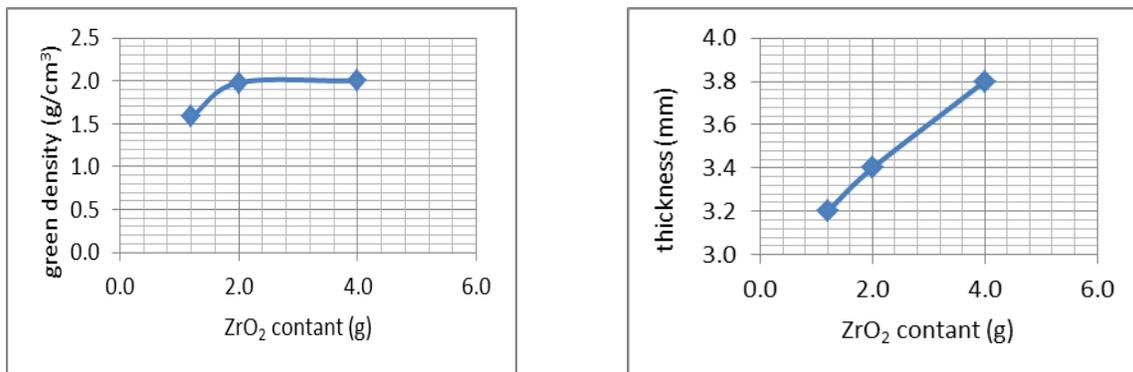
1- Effect of powder loading to the suspension :

The increase of zirconia powder loading also increases the pH of the suspension, as shown in figure (2). Thus, it is expected that this increase in pH helps in increasing the deposition density due to the increase of the surface charges. It is known that zirconia also increase the dielectric constant of the suspension; this may also give rise to the mobility of the charged particulates and increases the density.



Fig(2): effect of ZrO₂ on pH of 1g PEG in 100 ml ethanol suspension.

Figure (3-A) shows an increase in the deposit density with the increase of powder loading, but this increase become very low after the 2g of powder loading. This effect is the reduction of the electrical conductivity of the suspension with the increase of the zirconia content. On the other hand, the increase of the powder loading continues to increase the deposit thickness, figure (3-B), although the deposit density does not increase significantly.



A- Green density

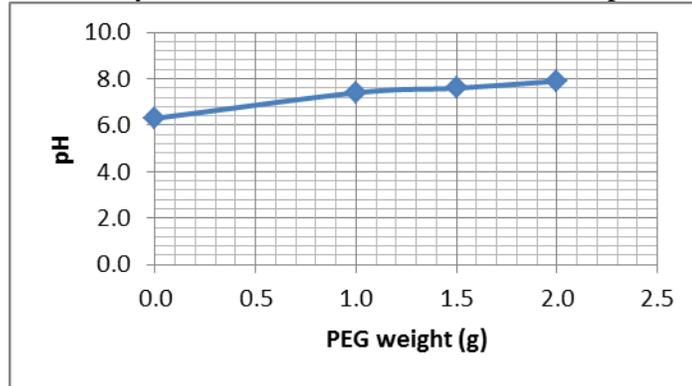
B- Thickness

Fig(3): effect of ZrO₂ on green density and thickness of deposited zirconia from 1g PEG in 100 ml ethanol suspension. A- Green density, B- Thickness.

2- The effect of PEG addition to the suspension:

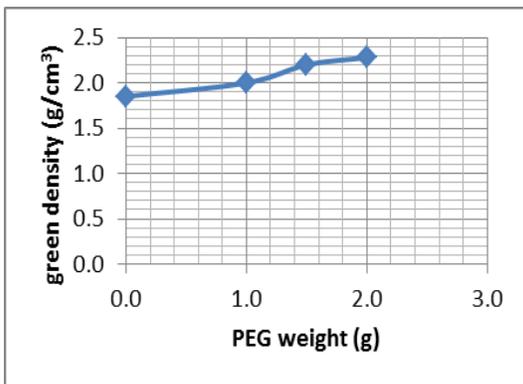
At first, the pH of the solution is monitored. It is found that the addition of PEG increases the pH of the suspension, as shown in figure (4). The PEG increases the OH⁻ concentration in the suspension. Thus, the PEG contributes in increasing the surface

charge of the zirconia particles in the suspension. Accordingly, the addition of the PEG is expected to enhance the density and the thickness of the zirconia deposit.

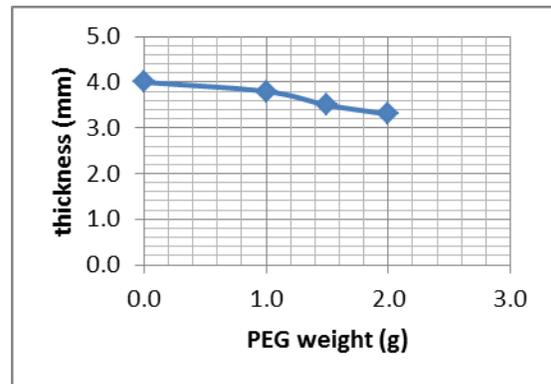


Fig(4): effect of PEG on pH of 4g zirconia in 100 ml ethanol suspension.

As shown in figure (5-A), the density of deposited zirconia was increased with the increase the amount of PEG, this increase is due to the increased particle charge in the suspension. For increase amount of PEG, the thickness is decreased. This decrease in thickness occurs due to the increase the packed of deposit by increasing the amount of PEG. The thickness of the deposit, figure (5-B) is decreased with increasing PEG.



A-Green density

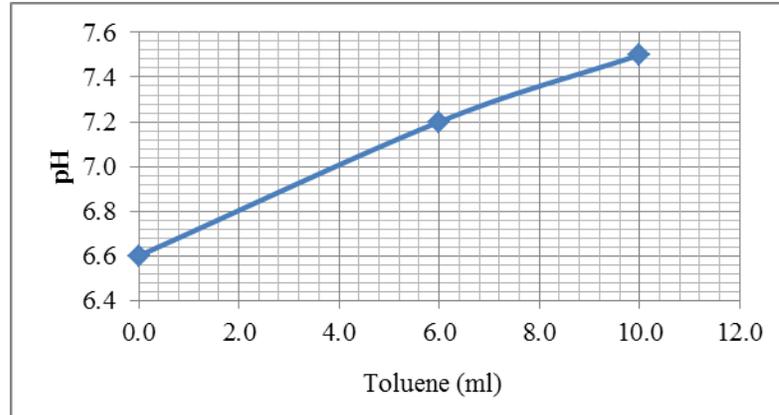


B-Thickness

Fig(5): effect of PEG on thickness and green density of deposited zirconia from 4g zirconia in 100 ml ethanol suspension. A- Green density, B- Thickness.

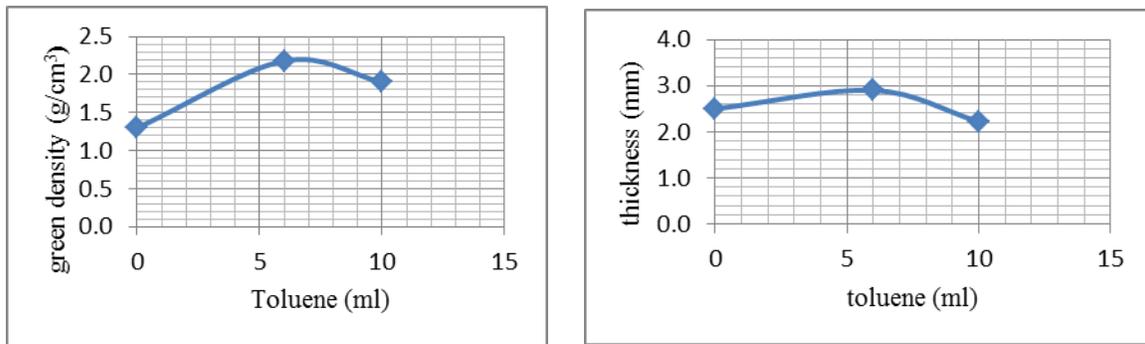
3- Effect of toluene addition to the ethanol- 1.2g zirconia – 1.5g PEG suspension.

The toluene was added to the ethanol in order to decrease the viscosity and dielectric constant of the suspension. The toluene also increases the pH of suspension. Figure (6) shows the increase of the pH of the suspension with increasing toluene.



Fig(6): effect of toluene on pH of 1.2g zirconia in 1.5g PEG -100 ml ethanol suspension).

The green density of deposited part were increased with addition 6 ml toluene to suspension of zirconia and reached 2.18g/cm^3 . This result may be attributed to the higher pH of the suspension due to the including of the toluene and accompanied increase of the surface charges. When the amount of the toluene is increased from 6ml to 10ml, it seems that the pH of the suspension is increased beyond the optimum and led to slowing down the particles mobility and accordingly, drops in the deposit density.



A- Green density

B- Thickness

Fig(7): effect of toluene on thickness and green density of deposited zirconia from 1.2g zirconia in 1.5g PEG -100 ml ethanol suspension. A- Green density, B- Thickness.

The thickness of deposit starts at value (2.5mm) at zero toluene content. Again, the increase of the particles surface charge enhances the deposition rate. The later drop in thickness for higher values (2.9mm) of the toluene 6 ml to (2.22mm) at 10 ml toluene was due to increase of the pH., i.e. due to the increase of packing of the deposit. Once more, the unhelpful increase of pH of the suspension at toluene content of 10ml led to depress the deposition rate and the drop of both the density and the thickness of the deposit.

Conclusions

- 1- The pH of the suspension increase with the increase of PEG content, zirconia powder loading, and with the addition of toluene. This increase of pH enhances the particles surface charge and enhance deposition rate, which led to the increase of density of the deposit.

- 2- The exaggerated content of PEG led to an increase of the mobility particles of the suspension with a positive effect on the deposition rate. Also, the exaggerated amount of toluene led to an increase of the pH of the suspension that counterpart the deposition rate.
- 3- Both the deposit thickness and density increase with the increase of the deposition rate and reach an inflection point where the increase in density led to higher particle packing and thus, reduced deposit thickness.

References:

- Albert Tarancón, 2009 , “Review Strategies for Lowering Solid Oxide Fuel Cells Operating Temperature” *Energies*, Vol. 2 , pp .(1130 -1150) .
- Bailey Moore, EbrahimAsadi, and Gladius Lewis, 2017, ” Deposition Methods for Microstructured and Nanostructured Coatings on Metallic Bone Implants: A Review”, *Hindawi ,Advances in Materials Science and Engineering*.
- Caproni E., Gouvea D. and Muccillo R., 2011 , “Yttria-stabilized zirconia closed end tubes prepared by electrophoretic deposition”, *Science Direct ,Ceramics International* ,Vol.37 ,pp.(273–277).
- Eduardo Antonelli, Ronaldo Santos da Silva, MariaInês Basso Bernardi and Antonio Carlos Hernandez , 2013 , ” Electrophoretic Deposition of BaTi 0.85 Zr0.15O3 Nanopowders”, *Materials Research*, Vol. 16, No. 6, pp.(1344-134).
- Ferrari B. and Moreno R., 1997 , ”Electrophoretic Deposition of Aqueous Alumina Slips” ,*Journal of the European Ceramic Society*, Vol. 17, pp.(549-556).
- Ferrari B. and Morenoz R., 2000, “Zirconia Thick Films Deposited on Nickel by Aqueous Electrophoretic Deposition” ,*Journal of The Electrochemical Society*, Vol. 147 , No.(8) ,pp.(2987-2992).
- Frédéric Bouyer and Alain Foissy , 1999 , ” Electrophoretic Deposition of Silicon Carbide”, *J. Am. Ceram. Soc.*, Vol. 82 ,No.(8),pp.(2001–2010).
- Goupee A. , 2005 , "Methodology for the Thermo-mechanical Simulation and Optimization of Functionally Graded Materials", M. Sc. Thesis, Mechanical Engineering, p. (6).
- Laxmidhar Besra, Tetsuo Uchikoshi, Tohru S. Suzuki, Yoshio Sakka, 2008 ,”Bubble-Free Aqueous Electrophoretic Deposition (EPD) by Pulse -Potential Application”.*J. Am. Ceram. Soc.*, Vol.91, No. (10), pp.(3154–3159).
- Linda Berg Aas , 2014 , ” Corrosion protection of metals/alloys by EPD (electrophoretic deposition)”, *Norwegian University of Science and Technology, Department of Materials Science and Engineering*.
- Peng Li, John T.S. Irvine , 2013 , “Fabrication of anode-supported zirconia thin film electrolyte based core-shell particle structure for intermediate temperature solid oxide fuel cells” ,*Materials International* , Vol. 23, No.(3), pp.(302–307).
- Pouya Amrollahi, Jerzy S. Krasinski, Ranji Vaidyanathan, Lobat Tayebi, and Daryoosh Vashaei , 2015 , “Electrophoretic Deposition (EPD): Fundamentals and Applications from Nano- to Micro-Scale Structures”, *Handbook of Nanoelectrochemistry*, Springer International Publishing Switzerland.
- Sasa Novak and Katja König , 2009 , ”Fabrication of alumina parts by electrophoretic deposition from ethanol and aqueous suspensions”, *Science Direct ,Ceramics International* , Vol.35, pp.(2823–2829).

- ShaohuaZHANG , C. C. Sorrel and F. Y. Li , 2005 , “Electrophoretic Deposition of Ceramic Powder”,*J.Mater. Sci. Tehnol.*, Vol.21 , pp.(107-110).
- Stefan Frank , Carolina Mochales , Martin Heimann , Felix Kochbeck , Rolf Zehbe , Claudia Fleck and Wolf-Dieter Mueller , 2014 , ” Electrophoretic Deposition of Zirconia Nanoparticles ”, *Symbiosis , Nanoscience & Technology: Open Access.*
- Van der Biest O., Joos E. and Vleugels J., 2006 , ”Electrophoretic deposition of zirconia layers for thermal barriercoatings”,*J Mater Sci*, Vol. 41,pp.(8086–8092).