

Parametric Study of the Effect of the Suspension Composition on the Electrophoretic Deposition of Alumina

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Abstract

Electrophoretic deposition (EDP) is gaining increasing attention both in science and industry because this method has allowed the formation of thin films or multilayer films of controlled thickness and morphology. The method enables the formation of films on substrates of complex geometry that suits for various applications.

This work is a study the effect of polyethylene glycol as a binder-suspension agent, the amount the solid loading (alumina particles), and the effect of the toluene as a dielectric liquid, on the pH of suspension, the final thickness and green density of the deposited parts.

It has been shown that a certain amount of polyethylene glycol when added to ethanol or ethanol-30%toluene has given good results for both the green density and the thickness.

Keywords:- Electrophoretic deposition, Polyethylene glycol, Alumina, Green density.

الخلاصة

أكتسب الترسيب الكهربائي اهتماما متزايدا في كل من العلوم والصناعة لأن هذا الأسلوب قد سمح بتشكيل الأغشية الرقيقة أو الأغشية متعددة الطبقات بسبك ومورفولوجيا مسيطر عليه. يمكن الطريقة من تشكيل الأغشية على ركائز باشكال هندسة معقدة والتي تناسب مختلف التطبيقات .

هذا العمل هو دراسة تأثير البولي إيثيلين جلايكول كعامل تعليق ، ومقدار المادة الصلبة (دقائق الألومينا)، وتأثير التولوين كسائل عزل كهربائي، على حامضية المعلق ، على السمك النهائي والكثافة الخضراء للأجزاء المترسبة.

وقد تبين أن هنالك كمية معينة من البولي إيثيلين جلايكول عند اضافتها الى الايثانول او الايثانول-30% تالوين قد أعطت نتائج جيدة لكل من الكثافة الخضراء والسمك.

الكلمات المفتاحية:- الترسيب الكهربائي، البولي إيثيلين كلايكول ، الألومنا، الكثافة الخضراء.

Introduction

Electrophoretic deposition (EPD) has attracted a great deal of interest in recent years due to its wide range of coating and part forming applications. It can be applied to any fine powder in a colloidal suspension, including oxides, carbides, nitrides, metals, and polymers. Electrophoretic deposition (EPD) is proved as a powerful method for the formation of both uniform thin and thick films on substrates of complex geometry (Atsushi and Fumihide , 2003 ; MARIJAS *et.al.*, 2004 ; Vesna , 2012). With EPD, ceramic bodies are shaped directly in 2012 from a stable colloidal suspension by a DC electric field, which causes the non metallic-surface charged particles to move to and deposit on an oppositely charged electrode (Linda , 2014 ; Pouya *et.al.*, 2015 ; Caproni *et.al.*,2011). 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 (Laxmidhar and Meilin , 2007; Shaohua *et.al.*, 2005). According to this, the EPD process requires the control of the suspension properties as well as the selection and optimization of the electrical parameters involved during forming (Ferrari and R. Moreno , 1997) .

Several papers have described the preparation of alumina deposits by EPD. These studies focus on the deposition rate and yield as a function of the process parameters; however, less is known about how the process parameters and the suspension composition and properties relate to the behavior of the deposit. 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 this work, the effect of the suspension's composition on the properties of the formed deposit layers is investigated. The goal was to determine the composition for effective and reproducible suspension for fabrication of alumina parts, and hence to encourage industrialization of the process. The effect of polyethylene glycol in ethanolic suspensions was analyzed.

Materials and methods

For the synthesis of Alumina samples, α -Al₂O₃ (Hefei EV NANO Technology , Chain; particle size 20-30nm) powder was used. Ethanol (Scharlab S.L.,Spain) was used as the suspension medium. Polyethylene glycol 4000 (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 (60-120V) is used in the course of the process, explicitly, the starting voltage was 60V and increased by 10V every 10 minutes, i.e. the total process time is 70 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 a suspension of alumina in ethanol (4g for each 100 ml) and varied weights of PEG (0, 0.5, 0.75, 1) g. The second group was of a constant amount of PEG (1g) for a 100 ml of ethanol and varied weight of alumina (4, 5, 10, 15) g. The third group was composed of a mixture of ethanol and 30% toluene with a varied weight of ethylene glycol (0, 0.5 , 0.75 , 1) g for each 100 ml of the mixture.

Results and discussion

Applying constant voltage (80V) in the electrophoretic process results in an alumina deposit with the formation of cracks after the drying process as seen in figure

(1-A). This result indicates low green density of the deposited alumina. Applying higher voltages leads to uneven or irregular deposit. The low voltage is sufficient for low thicknesses but not when the thickness goes higher. On the other hand, starting with higher voltages results in fast, unsteady deposition process. Thus, it is decided to use an incremental increase of the applied voltage as the thickness of the deposit increase (10V increase every 10 min). This method of applying variable voltages is not preceded by other works as far as we know. The resulting deposit was free of cracks after the drying process; figure (1-B) which indicates a homogeneous deposit with higher density.



A-



B-

Figure (1): Effect the type of DC voltage; A- constant voltage (80V), B- Variable voltages (60-120 V).

1- 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 (2). I.e. the PEG increases the OH^- concentration in the suspension. Thus, the PEG contributes in increasing the surface charge of the alumina particles in the suspension. Accordingly, the addition of the PEG is expected to enhance the density and the thickness of the alumina deposit.

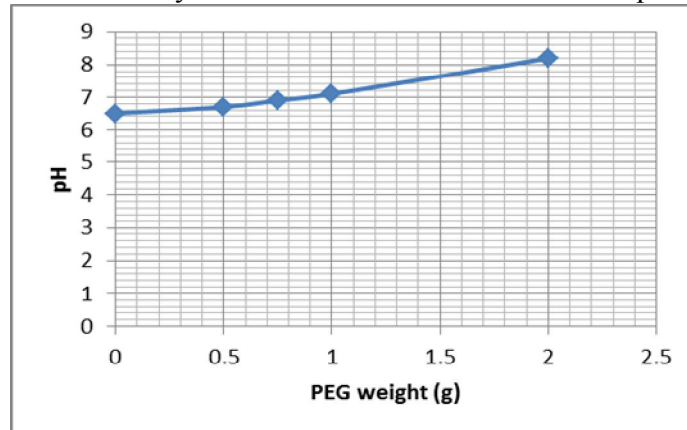
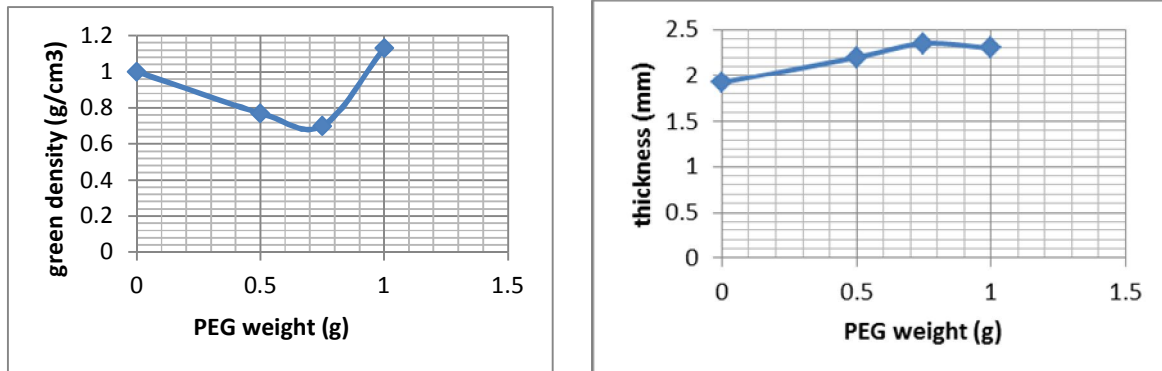


Figure (2): Effect of PEG on pH of (4g alumina in 100 ml ethanol) suspension.

As shown in figure (3-A), the density of deposit alumina was decreased with the increase the amount of PEG and reaches a minimum at 0.75g. For higher amounts of PEG, the thickness is increased. The increase of the thickness of PEG amount higher than 0.75 g may be attributed to the increase of the amount of the suspended alumina that

counterpart the negative effect of the increase of the suspension viscosity. The thickness of the deposit, figure (3-B) is increased with increasing PEG, i.e. it increased with the increasing amount of the suspended alumina. For PEG amount higher than 0.75 g, the increased density led to lower thickness.



A- Green density

B- Thickness

Figure (3): Effect of PEG on thickness and green density of deposited alumina.

2- Effect of powder loading of the suspension :

The increase of alumina powder loading also increases the pH of the suspension, as shown in figure (4). 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 alumina also decrease the dielectric constant of the suspension; this may also give rise to the mobility of the charged particulates and increases the density.

Figure (5-A) shows an increase in the deposit density with the increase of powder loading until a counterpart effect is evolved that cause a reduction in the deposit density. This effect is the reduction of the electrical conductivity of the suspension with the increase of the alumina content. On the other hand, the increase of the powder loading continues to increase the deposit thickness, figure (5-B), although the deposit density goes lower. The optimum powder loading for alumina, as shown in figure 5, is 5 g that gave a balance between deposit density and thickness.

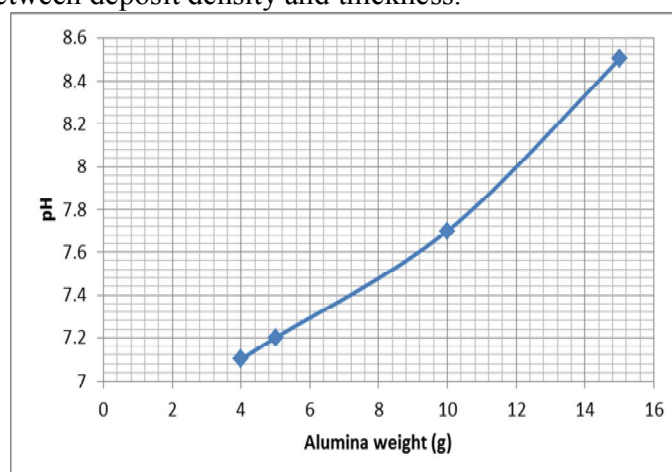
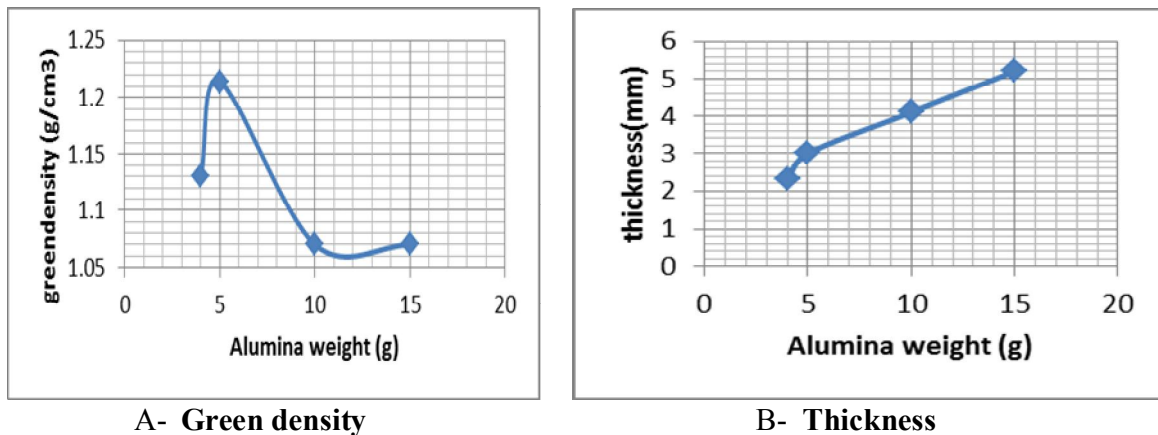


Figure (4): Effect of alumina powder loading on pH of suspensions.



A- Green density **B- Thickness**
Figure (5): Effect of alumina powder loading on the green density and the thickness of deposited alumina.

3- Effect of PEG addition to the ethanol-30% toluene 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 the suspension. Figure (6) shows the increase of the pH of the suspension with the existence of 30% toluene, which is much higher without it, as compared with that shown in figure (2).

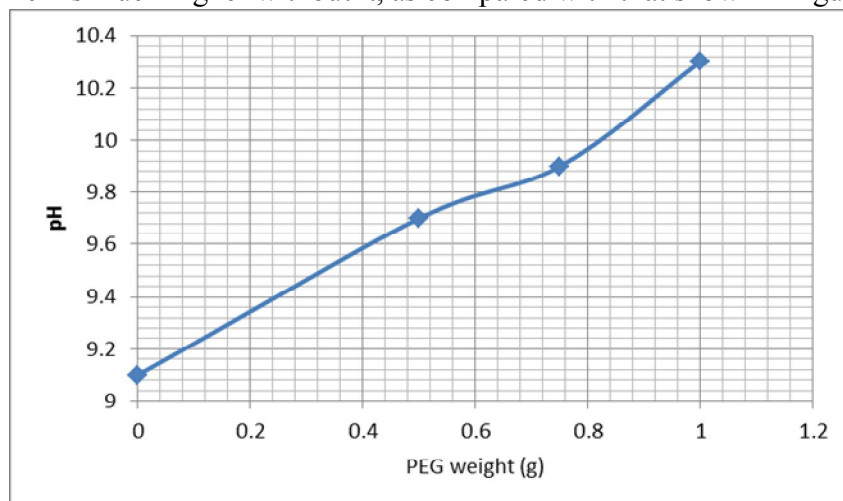
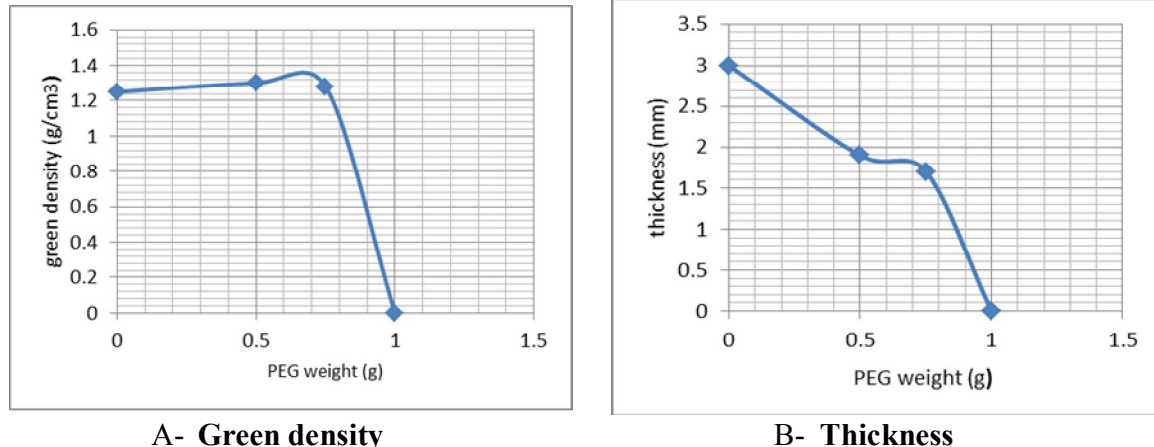


Figure (6): Effect of PEG addition on the pH of ethanol-30% toluene suspensions.

The green density of deposited parted were increased with the ethanol-30% toluene suspension of alumina and reached 1.25 g/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 PEG is increased from 0.75 g to 1.0 g, it seems that the viscosity of the suspension is increased beyond the optimum and led to slowing down the particles mobility and accordingly, drops in the deposit density.

The thickness of deposit start at high value (3mm) at zero PEG content compared with that of ethanol suspension alone (1.92mm). Again, the increase of the particle surface charge enhances the deposition rate. The later drop in thickness for higher values of the PEG (0.5 and 0.75 g) is due to increase of the density, i.e. due to the increase of packing of the deposit. Once more, the unhelpful increase of the viscosity of the

suspension at PEG content of 1.0 g led to depress the deposition rate and the drop of both the density and the thickness of the deposit.



A- Green density

B- Thickness

Figure (7): Effect of PEG on thickness and green density of alumina deposited from ethanol -30% toluene suspensions.

Conclusions

- 1- The pH of the suspension increase with the increase of PEG content, alumina powder loading, and with the addition of toluene. This increase of pH enhances the particle surface charge and enhances the deposition rate, which led to the increase of density of the deposit.
- 2- The exaggerated content of PEG (beyond 0.75 g) led to an increase of the viscosity of the suspension with negative effect on the deposition rate. Also, the exaggerated amount of alumina led to decrease of the electrical conductivity of the suspension that counterparts 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.

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