Electric-Field-Assisted Stagnation-Swirl-Flame Synthesis of Porous Nanostructured Titanium-Dioxide Films

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ABSTRACT:

Mesoporous nanostructured titanium-dioxide (TiO_2) films, with high uniformity and low packing density, are synthesized in a stagnation swirl-flame setup under an applied electric field, with voltage bias on the substrate and the burner grounded. The effects of uniform external electric-field strength and polarity are studied for different substrate temperatures and precursor-loading concentrations. The results manifest considerable differences in film characteristics, for differing electric fields, with more columnar structures and higher porosities produced under low-magnitude substrate voltages up to $|\pm 400| \text{V}$. Interestingly, films have higher packing density at higher-magnitude voltages of $|\pm 800| \text{V}$. At low substrate temperatures, the morphology and structure are more prominent owing to less on-substrate sintering of the nanoparticles. At low-magnitude substrate voltages, oppositely-charged particles are attracted to the substrate, increasing their electrophoretic velocity but decreasing their in-flame growth and agglomeration, resulting in smaller particle size. Along with van der Waals forces and induced dipole-dipole interactions, more columnar-structured growth with lower packing density results. At high-magnitude voltages, the particles seem to acquire the same charge polarity as the voltage bias applied, being repelled and residing in the flame longer, thus increasing in-flame agglomeration and particle size. More branched-structured film growth with high packing density develops.

Key words: porous film; titanium dioxide; electric field; packing density; flame synthesis

1. INTRODUCTION

Titania (TiO_2) nanoparticles have been widely used for UV filtering in sunscreens and as pigments in paints [1]. Since the 1970s, titania has emerged as an excellent photocatalyst for air purification [2], water treatment [3], and water splitting [4]. Recently, mesoporous nanostructured TiO_2 films have been used in dye-sensitized solar cells (DSSCs) [5–8], where the efficiency depends largely on film properties such as porosity, morphology, and thickness [9–11]. Efforts have been made to optimize these properties to improve their efficiency [5,11].

Flame synthesis has been utilized extensively for the production of particles and films [12–14]. Thimsen and Biswas [15] employed a premixed-flame aerosol reactor to make nanostructured TiO_2 films with controlled morphologies and film characteristics. Nikraz et al. [11] utilized a premixed flame stabilized on a rotating surface to obtain thin TiO_2 films suitable for DSSCs. Wang et al. [16] developed a turbulent, premixed stagnation swirl flame (SSF) to produce titania nanoparticles with 5-10nm size at high production rate, as well as to grow nanostructured TiO_2 films with high porosity [17]. The properties and characteristics of the nanostructured films can be controlled by adjusting flame conditions, substrate temperature, and precursor-loading rate [11,15,17,18].

Considerable research has been conducted on the effect of external electric fields on nanoparticle characteristics. Vemury et al.[19] investigated the formation of nanoparticles in a diffusion flame by applying an electric field and found a reduction in the primary-particle size. Katzer et al. [20] noticed that primary-particle size and crystalphase transformation correlated well with changes in flame temperature profile under an electric field. Zhao et al. [21] isolated the electrophoretic transport of nanoparticles, with no ionic wind, synthesized under uniform electric fields in low-pressure premixed stagnation flames. In this work, the effect of an external electric field applied during flame synthesis of nanostructured titania films is studied, for different substrate temperatures and precursor concentrations.

2. EXPERIMENTAL METHODS

2.1 Experimental Setup

The premixed-stagnation-swirl-flame setup for the study of TiO₂ film synthesis with applied electric field is shown in Fig. 1. The setup mainly includes an 18-mm stainless steel burner with an internal swirler (the same size as the burner inner diameter) and a stagnation aluminum plate positioned 20mm from the burner exit. Nitrogen is used as carrier gas for the precursor, titanium tetraisopropoxide (TTIP, Sigma Aldrich, 97%). The TTIP is heated to 80°C in a bubbler, and the downstream line is heated to 130°C to hinder condensation of the precursor as it flows to the burner. The main gas lines of CH₄, N₂, and O₂ are preheated and then mixed with the precursor line. The flow rates for N₂ (including both N₂ lines), O₂, and CH₄ are 27, 6.6, and 2.25L/min respectively. A 380μm-thick silicon substrate is placed on the aluminum plate to grow the TiO₂ films. A cooling plate placed below the aluminum plate, separated by quartz blocks, is used to control the substrate temperature. The aluminum plate is electrically isolated from all the other surfaces. The thickness of the quartz blocks and the flow rate of cooling water can be changed to achieve different substrate temperatures. A 125-μm S-type thermocouple between the aluminum plate and silicon substrate monitors the substrate temperature. A high-voltage power supply provides the voltage bias at the aluminum plate, creating the electric field between the grounded burner and the substrate.

2.2 Experimental Procedure

This work focuses on the effects of an electric field on the TiO₂-nanoparticle-film structure, porosity, and morphology over a range of substrate temperatures and precursor concentrations. Cases conducted are listed in Table 1. For each Case, the voltage varies from -800V to +800V in intervals of |200|V. The precursor concentration is varied by adjusting the nitrogen carrier-gas flow rate with the main gas line balancing the total N₂ flow rate, and the synthesis duration is adjusted accordingly to keep the total amount of precursor delivered the same for all Cases. The effect of electric field under different substrate temperatures (Cases 1-3) and different precursor concentrations (Cases 3-6) are investigated.

2.3 Packing Density

The TiO₂-film porosity can be determined by calculating the packing density ε of the film, where the porosity is $(1 - \varepsilon) \times 100\%$; and $\varepsilon = \frac{m_f}{\rho A_f h_f}$, where m_f is the weight of the film; ρ is the density of TiO₂ particles (3,840kg/m³); A_f is the top surface area of the film; and h_f is the thickness of the film (determined by SEM).

3. RESULTS AND DISCUSSIONS

3.1 Effect of electric field for Case 1

Figure 2 shows the SEM images of cross-sections of nanostructured TiO₂ films for Case 1, with 116ppm precursor concentration and 473K substrate temperature. The image for 0V (grounded substrate) shows a tree-like branched fractal film, which is similar to that obtained by Zhang et al.[17], Thimsen and Biswas[15], and Thimsen et al.[22]. However, as the voltage magnitude increases to $|\pm 400|V$, the films exhibit a more well-aligned columnar structure. Noteworthy, the trend is non-monotonic, as the film becomes less columnar as the voltage magnitude is set higher to $|\pm 800|V$. The columnar structure is similar to the simulation results of particle deposition in an electric field reported by Kulkarni and Biswas[23], as well as the experimental results for deposition on a single fiber of polarized and charged particles by Huang et al.[24].

To further explore the effects of electric field on particle growth, particles are collected from the substrate and characterized by XRD and TEM. The phase of the particles are anatase, irrespective of the voltage bias (magnitude or polarity) applied. A typical XRD spectrum is given in Fig. 3, shown with anatase lines from the database.

TEM images of titania nanoparticles taken directly from the TiO₂ films grown on the silicon substrate for Case 1 are shown in Fig. 4. Since the substrate temperature is low, there is insignificant on-substrate sintering resulting in less particle sintering. The average primary-particle sizes are 5nm, 4nm, and 9nm for 0V, -400V, and +400V, respectively. The applied electric field will impose a force on charged particles, affecting their velocity as well as residence time in the flame. Moreover, the effect of

electric field on the average size of particles depends on the particle charge distribution. In the flame, there are both positively- and negatively-charged TiO_2 nanoparticles [20], but not necessarily in equal distributions, since the “hot” flame can induce thermionic emission [21]. Although there exist models to predict the particle charge distribution [25], many experimental results indicate that the charge distribution for small nanoparticles may not coincide well with the computed results [26–28]. In the present case for TiO_2 nanoparticles, the average primary-particle size is much larger under positive voltage and smaller under negative voltage, indicating that more TiO_2 nanoparticles are positively charged.

The packing density of the nanostructured TiO_2 films under substrate voltages from -800V to +800V for Case 1 is presented in Fig. 5. A lower packing density is observed at lower magnitudes of voltages up to $|\pm 400|V$, after which it increases. The packing density is minimum at -400V. However, as the voltage magnitude exceeds $|\pm 400|V$, the particle size increases with voltage magnitude regardless of polarity. Figure 5 evinces that particle deposition and film porosity are strongly affected by an electric field, which presents another degree of freedom for process control.

3.2 Electric field Interactions

The main aspects which govern the deposition of particles are thermophoretic, electrophoretic, and drag forces, along with Brownian motion. The particles are assumed to move vertically toward the substrate until they collide with the nanoparticle film due to their transverse Brownian motion. Once they collide with the film, the particles are captured because of strong surface forces, and any further penetration into the film is terminated.

The mean travel distance of the primary-particle before collision with the film (l_p) can be calculated by basic collision theory in the free molecular regime [29,30] as

$$l_p = \frac{1}{c_f \beta \frac{\pi}{4} (d_{pi} + d_{pf})^2} \quad (1)$$

where, β is the enhancement factor for collision rate (including van der Waals, dipole, and coulombic forces [31,32]); d_{pi} and d_{pf} are the diameters of incident primary particles and deposited particles, respectively; and c_f is the local concentration of primary particles in the film, given by $c_f = \frac{\varepsilon}{(d_{pf})^3}$.

Particle movement towards the substrate depends on the total effects of thermophoretic, electrophoretic, gravitational, and drag forces acting on the particle. Since the gravitational/buoyant force is negligible for small/dense nanoparticles [14], an expression for particle velocity is formulated from the thermophoretic force (F_{th}) [33,34], electrophoretic force (F_e) [33], and drag force (F_d) [30] as given below

$$\frac{-p\lambda d_p^2 \nabla T}{T} + nEe = \frac{3\pi\mu_{gas}d_p}{C}(v_p - v_{gas}) \quad (2)$$

where, ρ_{gas} is the gas density; T is the temperature; ∇T is the temperature gradient; n is the average number of charges on a particle; e is the elementary charge; E is the electric-field strength; d_p is the particle diameter; μ_{gas} is the dynamic gas viscosity; v_p is particle velocity, v_{gas} is the gas velocity, and C is the slip correction factor, given as $C = 1 + \frac{2\lambda}{d_p}(1.257 + 0.4\exp(\frac{-0.55d_p}{\lambda}))$ [30].

The temperature gradient is obtained from the energy equation considering conduction and convection heat transfer in the boundary layer [35], using the velocity profile from stagnation-point flow, as described in Ref. [36]. For particles with a single charge, the electric forces range from 1.6×10^{-15} N to 6.4×10^{-15} N at different voltages, and the thermophoretic forces range from 3.8×10^{-16} to 1.9×10^{-15} N. Thus, the electric force is the main force on the bulk motion of charged particles.

The effect of the applied electric field on particle growth and packing density of the films can be classified in two main regimes: *i*) in-flame growth, and *ii*) film deposition. When the applied voltage is low, the charged particles are attracted to opposite polarity. For example, for a ‘negative’ electric field (i.e. negatively-biased substrate), positively-charged particles will be attracted to the film. This attraction increases the electrophoretic velocity causing the particles to move faster through the flame.

However, negatively-charged particles will be repelled from the substrate, residing longer in the flame. The residence time in the high-temperature regime in the flame will determine the particle-growth time and thus the particle size. If the particles were equally positively- and negatively- charged, then the particle size and the packing density curves should be symmetric about 0V. However, Figure 5 reveals that the packing density is lower at negative voltages of -200V and -400V than at positive voltages of the same magnitudes. This result is consistent with the TEM results in Fig. 4, that particles are more positively charged.

Inside the porous film, the Coulombic force works on the particles to alter their velocity, affecting their penetration depth and thus the film porosity. Furthermore, with the electric field, there are induced dipoles both on the nanoparticles in the flame and in the film. As a particle approaches the film, the van der Waals and dipole forces increase rapidly, as shown by Yan et al. [37]. The dipole force enhances the capture of particles by the film, and the enhancement factor is increased from 3.78 with van der Waals forces only to 8.59 with both dipole and van der Waals forces [31]. With a larger enhancement factor, the mean travel distance is reduced according to Eq. (1), which means the particles will be captured more readily, thus reducing the packing density of the film. Moreover, because of the direction of the induced dipoles on the film, the particles tend to deposit on the tip of the particle chain [24], thus forming more columnar structures compared to cases without electric field.

However, when a high enough voltage magnitude is applied to the substrate (at $\pm 600\text{V}$ and $\pm 800\text{V}$), the field accelerates electrons (from thermionic emission) in the ‘hot’ flame and induces stronger collisions between electrons and molecules, forming positive ions and more electrons. Under negative voltage, the positive ions will be attracted to the substrate, while the electrons will be repelled back into the flow field and collide with molecules producing more positive ions and electrons. Some electrons will attach to neutral molecules forming negative ions. The negative ions will stay in the flame for longer times, in turn attaching to particles to give them a negative charge. Then the electrophoretic force will be away from the substrate. Conversely, when a positive voltage is applied to

the substrate, the electrons (from thermionic emission) will be attracted to the substrate, while the positive ions will stay longer in the flame and attach to the particles, giving them a net positive charge. Therefore, the particles will be charged with the same polarity as the substrate regardless of the polarity of the voltage applied on the aluminum plate, which leads to a longer particle residence time in the flame and larger primary-particle size, increasing the packing density. Moreover, in the high voltage magnitude cases, as more particles are charged, the directional growth by the dipole interaction becomes relatively weaker compared to that by the Columbic force between charged particles, and the columnar structure is diminished.

3.3 Effect of electric field for cases with different substrate temperatures

As the temperature of the substrate increases from 473K to 663K to 763K, the packing density without the electric field first decreases from 0.0465 to 0.0426 (at 663K), and then increases to 0.0456 at 763K. This trend is consistent with the results obtained by Zhang et al. [17], for the same conditions, and can be explained by the balance of the thermophoretic-force effect and sintering as the temperature increases.

The electric field continues affecting the film for the entire temperature range. Figure 6 displays the cross-sectional SEM images of the TiO_2 film for Case 3 at the highest temperature of 763K. The film at 0V shows a branched structure such that film growth is more prominent in the horizontal direction. However, the films at $|\pm 400|\text{V}$ still show a columnar growth of the particles in the vertical direction, parallel to the electric field. The particles at the base of the films are sintered. Nevertheless, the electric field still affects the particle size, with TEM revealing that the primary-particle size decreases at -400V (8-9 nm) and increases at +400V (12 nm), as compared to 0V (10 nm).

The packing densities of the TiO_2 films under voltages from -800V to +800V for Case 2 (663 K, 116 ppm) and Case 3 (763K, 116ppm) are shown in Fig. 7. The trends of the packing density for Cases 2 and 3 are similar to that for Case 1, with the packing density being less at moderate voltage magnitudes, thus consistent with the proposed mechanism behind the growth of the films discussed

above. However, the reduced ratio of the minimum packing density to the packing density at 0V becomes smaller at higher substrate temperature.

3.4 Effect of electric field for cases with different precursor concentrations

As the precursor concentration increases from 29.1ppm to 291ppm with a constant substrate temperature at 763K, the packing density increases from 0.042 to 0.060, without the electric field. This result is attributed to the increase of average primary-particle size at higher precursor concentration [16,38,39], which will create a more packed film [17]. Moreover, as the precursor concentration is increased from 116ppm in Case 3 to 232.8ppm in Case 4, and further to 291ppm in Case 5, the film becomes more dense and displays a loss of the branched structure without electric field, as shown in the 0V condition of Fig. 8 for Case 5, which is consistent with the results obtained by Zhang et al.[17]. Nonetheless, the film structure changes under electric field, with more columnar structures at -400V and 400V, as shown in Fig. 8.

Figure 9 shows the packing density of the films for Cases 4 and 5, as the precursor concentration increases. The trends of the packing density for both cases display similar trends, with the packing density decreasing with voltage magnitude to $|\pm 400|V$ and then increasing with voltage magnitude. However, $\pm 600V$ and $\pm 800V$ results are lower than that at 0V, which is different than that for the low concentration cases.

However, as the precursor concentration decreases significantly to 29.1ppm in Case 6, the trend of the packing density is slightly different than that for the earlier cases, as shown in Fig. 10. The packing densities for +200V and +400V are higher than that for 0V, unlike all other cases. As the nucleated size of the primary particles at 0V is smaller due to lower TTIP concentration[33], Eq. 2 indicates that the electric force plays a more important role in particle movement for smaller particles. Since the particles are more positive charged, this effect results in much longer residence time of particles in the flame, and correspondingly much larger primary particle size at +200V and +400V. Thus, the packing densities at these voltages are larger than that at 0V. The change of the packing

density is not remarkable at voltages from -400V to +400V. Nevertheless, the structure of the film is changed under the electric field, as shown in Fig. 11, with more columnar structure at -400V and more dendritic structure at +400V.

4. CONCLUSION

Nanostructured TiO₂ films with high porosity and interesting morphology are grown in a stagnation-swirl-flame setup under an applied electric field. The effects of external electric-field strength and polarity are studied for a series of substrate temperatures and precursor-loading concentrations. The results show that the average diameter of the primary particles is less under a negative substrate voltage than that under a positive voltage, indicating that more synthesized particles are positively charged in the flame. The nanostructure and the porosity of the film can be altered by the applied electric field. With moderate voltage magnitudes up to $|\pm 400|$ V, the induced dipole interaction leads to more columnar-structure growth with higher porosity. The porosity is highest at -400V. However, the porosity decreases as the voltage magnitude becomes larger than $|\pm 400|$ V, probably because of the change in the charge polarity distribution of the nanoparticles. The effect of electric field on the properties of the nanostructured film is encountered over a wide range of substrate temperatures and precursor-loading rates, demonstrating that it can be a useful method for controlling film growth.

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Figure Captions list:

Fig. 1. Experimental Setup of the Stagnation Swirl Flame Burner.

Fig. 2. SEM images of TiO_2 films for case 1 (473K, 116 ppm).

Fig. 3. Typical XRD result of the TiO_2 nanostructured film.

Fig. 4. TEM images of TiO_2 films for Case 1.

Fig. 5. Packing Density of the TiO_2 films for Case 1.

Fig. 6. SEM images of TiO_2 films for Case 3.

Fig. 7. Packing Density of the TiO_2 films for Cases 2 and 3.

Fig. 8. SEM images of TiO_2 films for Case 5 (763K, 291ppm).

Fig. 9. Packing Density of the TiO_2 films for Cases 4 and 5.

Fig. 10. Packing Density of the TiO_2 films for Case 6 (763K, 29.1ppm).

Fig. 11. SEM images of TiO_2 films for Case 6 (763K, 29.1ppm).

Table 1. Set of experimental Cases; PD: Packing Density.

	T (K)	TTIP (ppm)	Time (s)	Analysis
1	473	116	300	SEM, TEM, XRD, PD
2	663	116	300	SEM, PD
3	763	116	300	SEM, TEM, PD
4	763	232.8	150	SEM, PD
5	763	291	120	SEM, PD
6	763	29.1	1200	SEM, PD

Figure 1 color

Figure 1 grey

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7 color

Figure 7 grey

Figure 8

Figure 9 color

Figure 9 grey

Figure 10

Figure 11