

Effect of Distance of the Flame's Inner Cone from Substrate Surface on Diamond Films Synthesized on Ti Substrate for Dental Implants by Flame Combustion

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Abstract: Flame combustion enables the synthesis of diamonds via acetylene-oxygen gas flame combustion in ambient air. Recently, titanium has been used for dental implants in the dental industry. In this study, diamond films were synthesized on a Ti substrate using the flame combustion method to improve the strength, wear resistance, and biocompatibility of dental implant surfaces. Moreover, to obtain high-quality diamond films and various synthesized diamond crystallites sizes and to achieve good adhesion on the Ti substrate, as a condition of the synthesis to prevent delamination, the distance of the flame's inner cone from the substrate surface was varied by 1.0, 1.5, 2.0, 3.0, 4.0, and 5.0 mm. Based on the results, diamond films were synthesized on a Ti substrate surface via flame combustion. The surface morphology of the synthesized films was altered by varying the distance between the inner cone of the flame and the substrate surface. This distance affected the surface morphology of the synthesized diamond films. The delamination of the synthesized films was prevented completely. To investigate the factors causing this result, nuclei were generated on the substrate during the initial stages of synthesis. The effect of the distance between the inner cone of the flame and the substrate surface on the diamond films synthesized by flame combustion was investigated. The relationship between this distance and the state of the diamond nuclei on the substrate in the initial stages of synthesis was verified.

Keywords: Diamond films; Flame combustion; Titanium substrate; Distance in flame; Diamond nuclei; Delamination.

1. Introduction

Owing to its remarkable properties such as high thermal conductivity, hardness, and wear resistance, diamond is utilized in the industry for manufacturing cutting and polishing tools. Recently, diamond has been studied extensively as a coating material for medical devices [1-6]. Diamond films have also been considered as coating materials for dental cutting tools [7-9].

In the medical community, owing to the remarkable properties of diamond, further applications of diamond films in medical devices can be developed. Currently, the demand for dental implants is increasing owing to accidents, diseases, and the aging of the population [10,11]. Metals are typically used for dental implants. Recently, titanium has been used for dental implants in the dental industry. However, dental implants can corrode during use and can fracture under various loads. The failed implants are discarded. In addition, implant materials have been reported to cause metal allergies, and the compatibility of metallic materials with the human body is an issue. Recently, diamond films have been studied for use as a Ti substrate [5,12,13]. To address these challenges, the study of diamond films on metals for dental implants has attracted significant attention [14]. However, the experimental equipment utilized in the CVD method is significantly large, and the synthesis speed is significantly low. We focused on the synthesis of diamond films on Ti substrates using the flame combustion method [15].

The flame combustion method enables the synthesis of diamond using acetylene-oxygen gas (C_2H_2/O_2) flame combustion in ambient air [16,17]. It has various advantages over other methods, such as high synthesis speed, safety, and low equipment cost. There are desirable features in the industry. However, the factors affecting diamond synthesis are unknown, and no means of precisely controlling this method have been established. Moreover, during cooling, most diamond films synthesized using the flame combustion method delaminate

because of thermal stress. We previously synthesized diamond films on molybdenum and tungsten carbide substrates using the flame combustion method [18-24]. Therefore, we focused on the synthesis of diamond films for medical devices using this method. It has a similar advantageous feature. We also synthesized diamond films on Ti substrates for dental implants using the flame combustion method [25]. In a previous study, to synthesize diamond films and prevent film delamination, conditions for synthesis on Ti substrates were proposed. The hardness and toughness of films synthesized using the flame combustion method may be higher than those of films synthesized using common synthetic methods.

This study considered that if diamond films can be synthesized directly on a Ti surface by flame combustion and good adhesion can be achieved, surfaces can be improved in terms of hardness and corrosion resistance and ultimately, in terms of the compatibility of metal materials with the human body. Therefore, to improve the strength, corrosion resistance, and biocompatibility of dental implant surfaces, diamond films were synthesized on a Ti substrate (a dental implant material) using the flame combustion method with a mixture of acetylene and oxygen gas. The feasibility of synthesizing a diamond film on a Ti substrate was examined. In addition, to obtain high-quality diamond films and achieve good adhesion on the Ti substrate, we focused on the effect of the distance of the flame's inner cone from the substrate surface. As a condition for the synthesis to obtain high-quality diamond films and various synthesized diamond crystallites size and to prevent delamination, this distance was varied by 1.0, 1.5, 2.0, 3.0, 4.0, and 5.0 mm. Based on the results, diamond films were synthesized on a Ti substrate surface via flame combustion. The surface morphologies of the synthesized films were altered by varying this distance. Therefore, when diamond films are synthesized on a Ti substrate via flame combustion, the distance affects the surface morphology. The delamination of the synthesized films was prevented completely in all the cases. To investigate the factor causing this result, the generation of nuclei on the substrate in the initial stages of synthesis was observed by scanning electron microscopy (SEM). The effect of the distance on diamond films synthesized by flame combustion was studied. The relationship between the distance of the inner cone of the flame from the substrate surface and the density of the diamond nuclei on the substrate in the initial stages of synthesis was verified.

2. Experimental details

2.1 Substrate

Recently, titanium and its alloys have been used in the dental industry as dental implants [10,14,25]. In this study, Ti was used as the substrate for dental implant materials. Ti with 99.5% purity for dental implants was used as the substrate for the diamond synthesis. The substrate was a disk with a diameter of 10 mm and thickness of 3 mm. As a pretreatment to prevent delamination, scratch processing (in which the substrate surface is ground with emery paper in one direction) was used to roughen the surface. Furthermore, as growth nuclei for the diamond synthesis, diamond seed particles with a diameter of approximately 0.25 μm were dispersed in acetone, the Ti substrate was added, and seed attachment processing was performed for 30 min with an ultrasonic syringe. Many diamond seed particles were attached to the substrate surface via the pretreatment.

2.2 Experimental equipment

The experimental setup is illustrated in Figure 1. A 100 \times 100 \times 55 mm rectangular copper box was used for cooling. Cooling water was poured into the box, and the film surface temperature was maintained at 1203 K. A noncontact infrared radiation thermometer was used to measure the film surface temperature during the synthesis. To support the cooling, a copper rod with a diameter of 10 mm was set vertically at the center of the box and fixed to a table using a flange. The Ti substrate was attached to a rod. For efficient cooling, a thermally conductive Ag paste was applied between the Ti substrate and rod. Subsequently, the samples were glued to a furnace at 473 K.

A cooling box was then placed on the stage. Because it was capable of moving vertically, the distance from the cooling water side to the film surface was varied, and the film surface temperature was controlled. A stepping motor was set on the stage and controlled using a stage controller.

Acetylene and oxygen were used as fuels for the synthesis. A burner was used for the welding. The mixed gas was introduced into the burner and combusted. The burner exit diameter was 1 mm. In addition, a mass flow controller that precisely controlled the gas flow rate and digitally displayed the flow quantity was used as a gas flow meter.

2.3 Synthesis conditions

The synthesis conditions are listed in Table 1. The conditions have been determined and reported previously [16-19]. There are considered to be optimum for preventing delamination during the synthesis of diamond films. The ratio of the oxygen flow rate ($F_o = 63.8 \text{ cm}^3/\text{s}$) to the acetylene flow rate ($F_a = 70.9 \text{ cm}^3/\text{s}$) was set to $R_f = F_o / F_a = 0.90$. This was because delamination-free crystallite growth could be realized at $R_f = 0.90$ [19].

It has been indicated [23-26] that the surface roughness generated by the pretreatment of a substrate surface causes delamination. Therefore, diamond films should be synthesized on pretreated substrate surfaces. As a pretreatment to prevent delamination, scratch processing was performed. Herein, the substrate surface was ground with emery paper in one direction. The Ti substrate was scratched to roughen its surface. We verified that the delamination of pretreated Ti substrates during the scratching process with emery paper of grain size # 400 as part of the diamond film synthesis was prevented effectively [25]. Therefore, diamond films were synthesized on pretreated Ti substrates for the scratching process using emery paper with a grain size of # 400. The surface roughness during the scratching process was measured using a scanning white light interferometer (SWLI, Zygo New View 6K). The mean value of the measured surface roughness R_a (arithmetical mean deviation of the assessed profile) for emery paper of grain size # 400 was 0.298 μm [25].

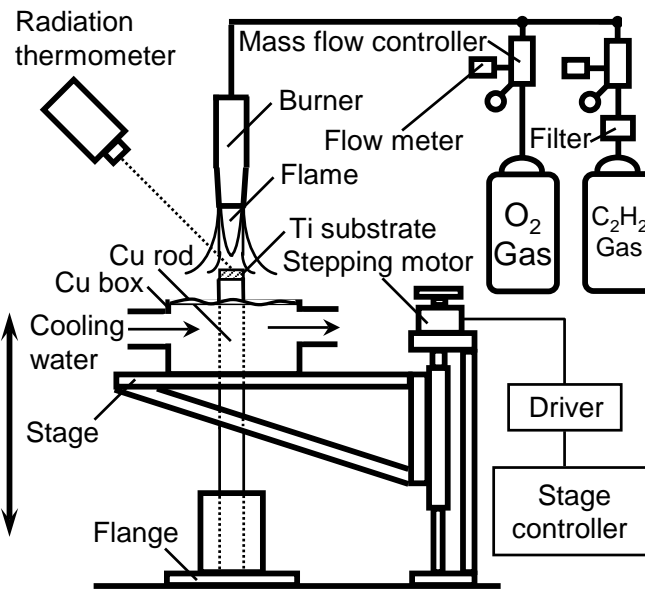


Figure 1. Experimental setup for synthesizing diamond by acetylene-oxygen flame combustion.

Table 1. Conditions for diamond syntheses.

Reaction gas	C ₂ H ₂ + O ₂
Film surface temperature	1203 K
Emery paper grain size level	# 400
Pure C ₂ H ₂ flow rate, F_a	70.9 cm ³ /s
O ₂ flow rate, F_o	63.8 cm ³ /s
Flow ratio, $R_f = F_o / F_a$	0.90

2.4 Distance of the flame's inner cone from the Ti substrate surface

An outline of flame combustion is illustrated in Figure 2. Flame combustion comprises an inner flame cone, acetylene feather, and outer luminous layer. Diamonds are synthesized from acetylene feathers. The distance d between the inner cone of the flame and Ti substrate surface is shown in the figure. We focused on the effect of this distance as a condition for the synthesis of diamond films. To obtain high-quality diamond films and various synthesized diamond crystallite sizes and to achieve a good adhesion on the Ti substrate, the distance of the inner cone of the flame from the substrate surface was varied by 1.0, 1.5, 2.0, 3.0, 4.0, and 5.0 mm. During synthesis, the acetylene feather area varied as the distance did. This is considered to affect the synthesis and delamination of the diamond film. The distances between the inner cone of the flame and the substrate surface are presented in Table 2. The total synthesis time was set to 3600 s [19,20,25]. The average thickness of synthesized films was 30 $\mu\text{m}/\text{h}$. In this method, the film surface temperature was set to 1203 K during the synthesis.

Table 2. Conditions for distance d of the flame's inner cone from the Ti substrate surface.

	Case A	Case B	Case C	Case D	Case E	Case F
Distance d of the flame's inner cone from the Ti substrate surface [mm]	1.0	1.5	2.0	3.0	4.0	5.0

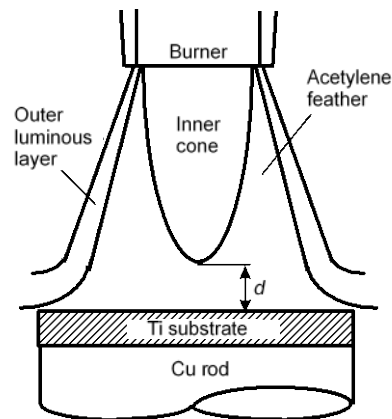


Figure 2. Outline figure of flame combustion.

3. Results and discussion

3.1 Delamination of synthesized films

The delamination of the synthesized films for various values of the distance of inner cone of flame from substrate surface (Cases A, B, C, D, E, and F; $d = 1.0, 1.5, 2.0, 3.0, 4.0,$ and 5.0 mm) was prevented completely. In a previous study [25], the delamination of the synthesized films during the scratching process with emery paper # 400 grain size ($R_a = 0.298 \mu\text{m}$) was prevented completely. In this study, diamond films were synthesized on Ti substrates pretreated by scratching with emery paper of grain size # 400. Therefore, it is considered that the delamination was prevented even when the distance of the flame's inner cone from substrate surface varied.

3.2 Investigation of synthesized films

The films synthesized with various values of the distance from the inner cone of flame to substrate surface were analyzed by SEM (JEOL JSM-7800F, JEOL JSM-5800) and X-ray diffraction (XRD, Rigaku RINT-2200V). The SEM images of the films synthesized in Case A ($d = 1.0$ mm) are shown in Figure 3. The SEM image verifies that the film was synthesized under the conditions of Case A, thereby indicating that film synthesis was feasible. The SEM images show that the size of the individual crystals was large. Moreover, the synthesized crystallites did not indicate a uniform size or high density. Atypically growing crystals were also observed among the crystals. The SEM images of the films synthesized in Case B ($d = 1.5$ mm) are shown in Figure 4. The SEM images show that the synthesized crystallites had nearly uniform sizes and high densities. The sizes of the individual crystals were smaller than those in Case A. The number of crystals with atypical growth decreased. Thus, we considered that the distance of the flame's inner cone from the substrate surface affected the diamond synthesis. The SEM images of the films synthesized in Case C ($d = 2.0$ mm) are shown in Figure 5. In the SEM image, the synthesized crystallites indicate a nearly uniform size and high density similar to Case B. From the image, the size of individual crystals was smaller than that for Cases A and B. SEM images of the films synthesized for Cases D, E, and F are illustrated in Figures 6, 7, and 8, respectively. Each SEM image shows that the synthesized crystallites had a nearly uniform size and high density. From the SEM images of Cases D, E, and F, the size of the individual crystals was smaller than that for Cases A, B, and C. The synthesized crystallites became smaller as the distance of the flame's inner cone from the substrate surface increased.

The XRD patterns for the typical Case A are illustrated in Figure 9. Here, the peaks indicating the presence of diamond and the Ti substrate are verified. Peaks indicating the presence of diamond (111) and (220) surfaces are verified. The peak of the diamond (111) surface exhibited a crystal growth direction at the top of the octahedral morphology. The XRD patterns for the typical Case C are illustrated in Figure 10. Here, the peaks indicating the presence of diamond and the Ti substrate are verified. Peaks indicating the presence of the diamond (111) and (220) surfaces are also verified. Furthermore, the XRD patterns for the typical Case F are illustrated in Figure 11. Here the peaks indicating the presence of diamond and the Ti substrate are verified. Peaks indicating the presence of the diamond (111) and (220) surfaces are also verified.

The synthesized crystallites became smaller as the distance between the inner cone of the flame and the substrate surface increased. The surface conditions of the synthesized films were determined by varying their distances. Peaks indicating the presence of the diamond (111) and (220) surfaces were also verified. From the SEM images and XRD patterns, diamond crystallites were synthesized, and diamond films were synthesized on the Ti substrates in each case. In the XRD patterns, the peaks of the diamond (111) surface are highly distinct. The synthesized diamond crystallites exhibited an approximately octahedral morphology and were thus considered to be of good quality. From the SEM images, the synthesized crystallites became smaller as the distance between the inner cone

of the flame and the substrate surface increased. The surface morphology of the synthesized films was altered by varying the distance between the inner cone of the flame and the substrate surface ($d = 1.0, 1.5, 2.0, 3.0, 4.0,$ and 5.0 mm). It was observed that the surface morphology of the synthesized films obtained by modifying the distance varied and that the variation in the distance had a significant effect on the surface of the synthesized films. These variations in the distance are considered to have caused variations in the initial diamond nucleation state.

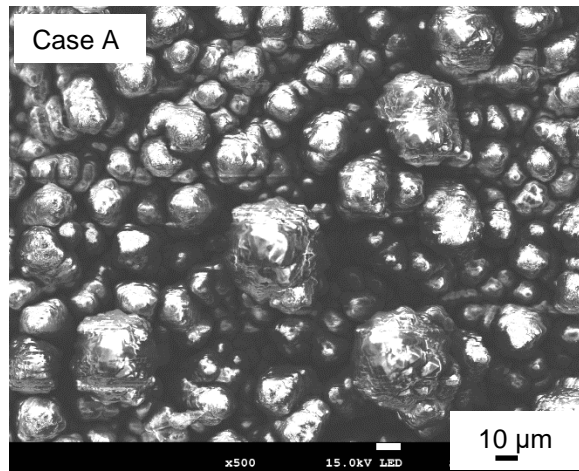


Figure 3. SEM image of the synthesized diamond film for Case A.

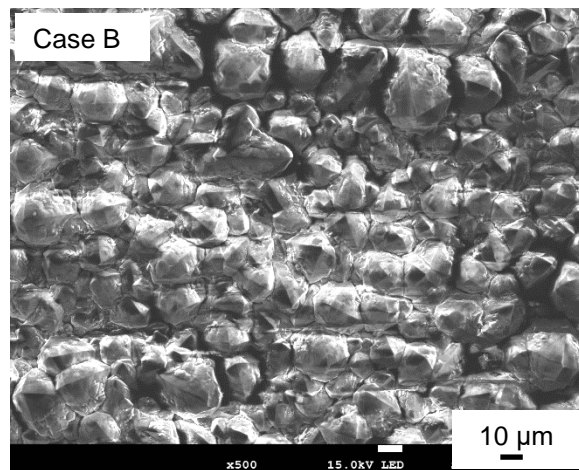


Figure 4. SEM image of the synthesized diamond film for Case B.

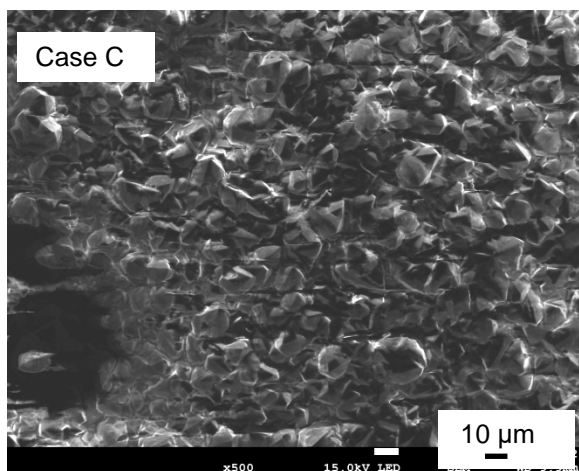


Figure 5. SEM image of the synthesized diamond film for Case C.

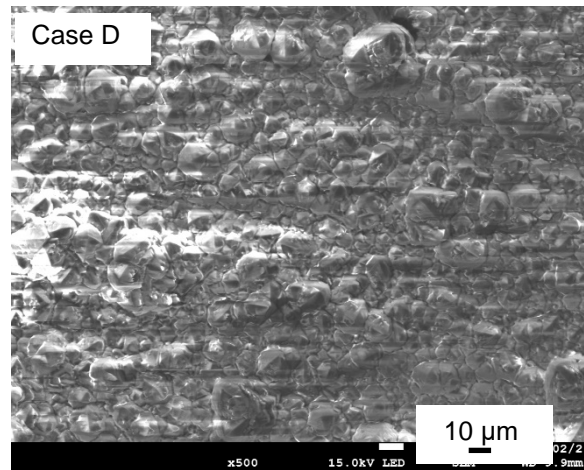


Figure 6. SEM image of the synthesized diamond film for Case D.

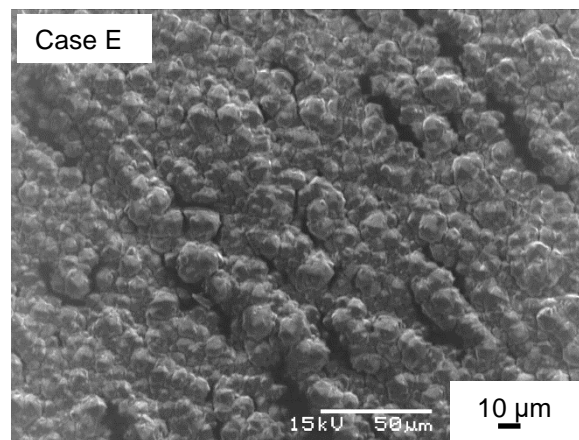


Figure 7. SEM image of the synthesized diamond film for Case E.

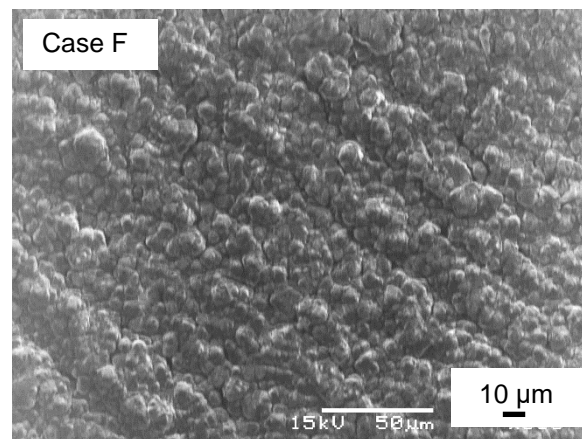


Figure 8. SEM image of the synthesized diamond film for Case F.

In each case, the distance of the inner cone of the flame from the substrate surface was varied to synthesize diamond films on the Ti substrates. Therefore, the synthesized films were analyzed by scanning white light interferometry (SWLI, Zygo New View 6K), and the surface roughness R_a (arithmetical mean deviation of the assessed profile) of the synthesized films was measured. The relationship between the measured R_a and the distance between the inner cone of the flame and the substrate surface is illustrated in Figure 12. The error bars in the figure illustrate a marginal variation in the surface roughness. The standard deviation is indicated. The results verified that the mean value of the measured surface roughness varied in each case and that the measured surface

roughness decreased as the distance increased. The results revealed that the surface roughness of the synthesized diamond films was affected by the variations in distance. This was attributed to the variation in the growth of the generated nuclei and the growth rate of the diamonds as the distance varied. When the distance was short, in Case A, the growth of the generated nuclei and the growth rate of the diamonds were high, the size of the diamond crystallites was large, and the surface roughness was large. In Case B, the distance was longer than that in Case A, the growth of the generated nuclei and growth rate of the diamonds was less than those in Case A, and the size of the diamond crystallites and the surface roughness were smaller. This result may be attributed to the increased distance. In Case C, the distance was longer than that in Case B, the growth of the generated nuclei and growth rate of the diamonds was less than that in Case B, and the size of the diamond crystallites and the surface roughness were smaller. In Cases D, E, and F, the distance was longer. Moreover, as the distance increased, the growth of the generated nuclei and growth rate of the diamonds decreased, and the size of the diamond crystallites decreased. Moreover, the surface roughness was smaller for the same reason as in Case C. In this study, the surface roughness was considered to be the lowest. The results show that it is feasible to alter the surface roughness of the synthesized diamond films by varying the distance between the inner cone of the flame and the substrate surface.

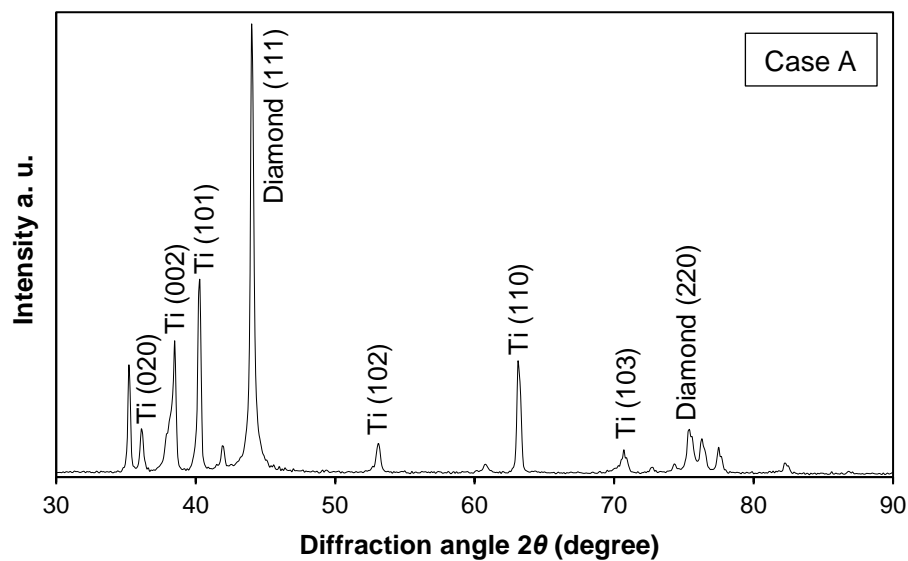


Figure 9. XRD patterns of the synthesized diamond film for Case A.

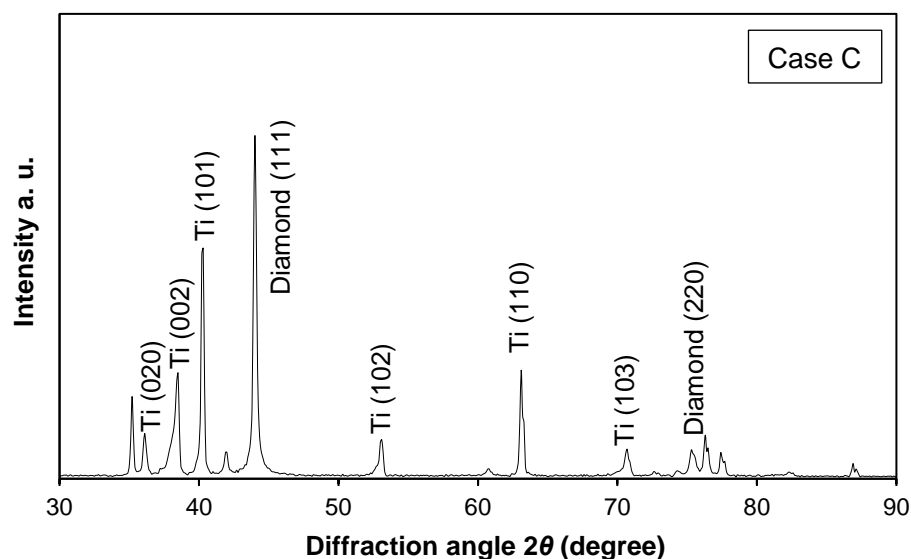


Figure 10. XRD patterns of the synthesized diamond film for Case C.

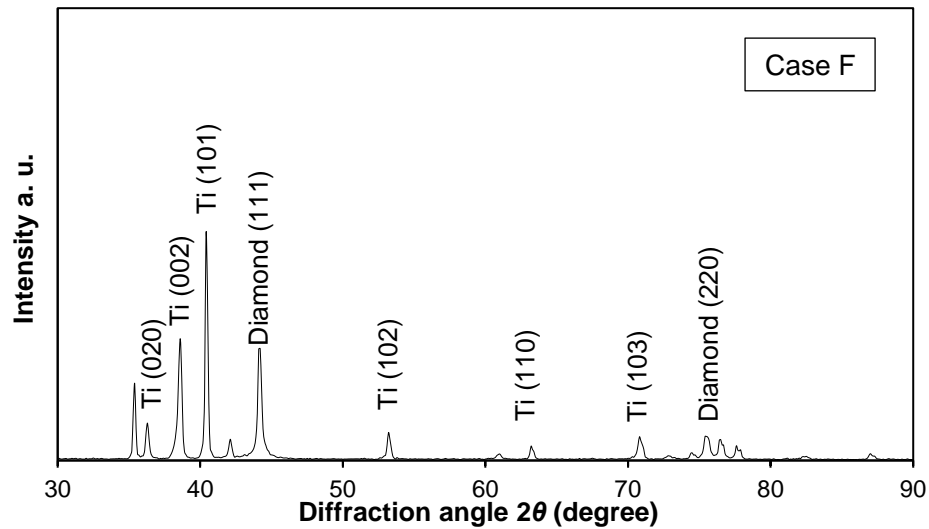


Figure 11. XRD patterns of the synthesized diamond film for Case F.

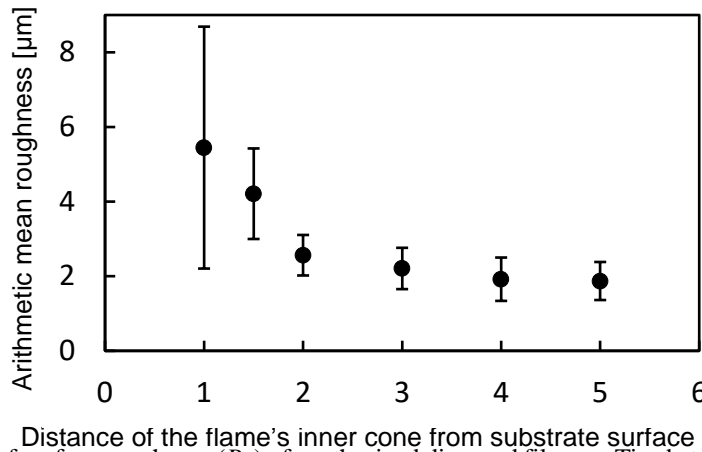


Figure 12. Results of surface roughness (R_a) of synthesized diamond films on Ti substrate surface for each distance.

3.3 Discussion

The SEM images illustrate that the diamond morphology of the synthesized diamond films was affected by the distance of the inner cone of the flame from the substrate surface in Cases A, B, C, D, E, and D ($d = 1.0, 1.5, 2.0, 3.0, 4.0,$ and 5.0 mm). The delamination of the synthesized films at different distances was prevented completely in all cases. In this regard, the following reasons are provided. The delamination owing to thermal stress during the cooling process after film synthesis was suppressed. This is considered to have resulted from the films being synthesized at a constant temperature, which resulted in a uniform coating growth rate, uniform thickness, and moderate dispersion of thermal stress. In a previous study [25], the delamination of synthesized films during the scratching process with emery paper #400 grain size ($R_a = 0.298 \mu\text{m}$) was prevented completely. The substrate surfaces were scratched using emery paper. This effectively prevented delamination. As a pretreatment to prevent delamination, a scratching treatment in which the substrate was scratched with emery paper was used to roughen the surface. The substrate surface conditions were optimal for preventing delamination. In this study, diamond films were synthesized on pretreated Ti substrates by scratching with emery paper of grain size # 400. Therefore, it is considered that the delamination was prevented even when the distance varied.

In addition, a discussion on the crystallinity of the synthesized films revealed that differences in the crystallinity of synthesized films occurred under the conditions in each case. This indicated that the differences in the distance of inner cone of flame from substrate surface also affected the crystallinity of the synthesized diamond films. We consider that the distances affected the diamond film synthesis of the synthesized films. In Case A, synthesized with a distance of 1.0 mm, the size of the individual crystals was large, and the synthesized crystallites did not indicate a uniform size or high density. Abnormally growing crystals were also observed among the crystals. This is considered to be because the short distance resulted in a high growth rate. Moreover, only a few crystals grew, whereas others could not grow significantly. This resulted in variations in the crystal size. In Case B, which was synthesized at a distance of 1.5 mm, the synthesized crystallites had a nearly uniform size and high density. The sizes of the individual crystals were smaller than those in Case A. The number of crystals with abnormal growth

decreased. This is considered to be owing to the longer distance than that for Case A, which slowed the growth rate, distributed energy to individual nuclei, and prevented crystal size variations. Thus, we considered that the distance conditions affected the diamond synthesis. In Case C (which was synthesized at a distance of 2.0 mm), compared with Case A, the synthesized crystallites had a nearly uniform size and high density (similar to Case B). The sizes of the individual crystals were smaller than those for Cases A and B. This is considered to be owing to the longer distance compared with Cases A and B, which slowed the growth rate, distributed energy to individual nuclei, and prevented crystal size variations. In Cases D, E, and F (synthesized with distances of 3.0, 4.0, and 5.0 mm), each synthesized crystallite had a nearly uniform size and high density. As the distance increased, the size of the individual crystals became smaller than that of the individual crystals in Cases A, B, and C. This is considered to be owing to the longer distance, which slows the growth rate. This can be attributed to the fact that more energy is lost over the longer distance. Thus, we considered that varying the distance of the flame's inner cone from the substrate surface affected the diamond synthesis.

Although the surface roughness ($R_a = 0.298 \mu\text{m}$) of the substrate surface was identical, the mean measured R_a of the synthesized films varied in each case, and the measured surface roughness of the synthesized films decreased as the distance increased. The results showed that the surface roughness of the synthesized diamond films was affected by the variations in distance. The growth rate depended on the distance between the inner cone of the flame and the substrate surface. This was attributed to the variations in the growth of the generated nuclei and the growth rate of the diamonds as the distance varied.

The results showed that the crystallites of the synthesized diamond film were affected by the distance between the inner cone of the flame and the substrate surface. This effect may be attributed to the initial diamond nucleation during the first synthesis stage. Therefore, we focused on diamond nucleation in the initial synthesis stage to investigate the effect of varying the distance on the initial nucleation. In each case, the distance of the inner cone of the flame from the substrate surface was varied (as shown in Table 2), and the state of diamond nucleation in the initial stages of the synthesis was observed using SEM (JEOL JSM-7800F). From the results obtained, we determined the effect of varying the distance of the inner cone of the flame from the substrate surface during the synthesis.

The substrate surfaces were observed after the first 120 s of synthesis. Herein, the distance of the inner cone of the flame from the substrate surface was varied in each case. The effect of this variation on the initial diamond nucleation behavior during the first synthesis stage was discussed. SEM images of the substrate surfaces after the first 120 s of synthesis for Cases A, C, E, and F are shown in Figures 13, 14, 15, and 16, respectively. These figures show that nucleation occurred on the Ti substrate surface in each case when the distance was varied and diamond synthesis was performed.

From Figure 13, the nuclei in Case A were of a high density, and the size of the nuclei increased. This is attributed to the short distance of the inner cone of the flame from the substrate surface, which resulted in a high rate of nucleation. Thus, the nucleation energy was not distributed adequately. Figure 14 shows that the density of the nucleated nuclei in Case C was lower than that in Case A. The size of the nuclei was smaller than that in Case A, with no large nuclei observed. Rather, each nucleus was smaller. This is considered to be owing to the lower nucleation rate caused by the longer distances. From Figure 15 and 16, in Cases E and F, the density of the nucleated nuclei decreased sequentially compared with that in Case C when the distance was increased. The size of the nuclei became sequentially smaller than that in Case C as the distance increased. These results indicate that the nucleation state on the substrate varied when the distance of the inner cone of the flame from the substrate surface did. This, in turn, affected the synthesized diamond film.

To investigate the effect of varying the distance of the inner cone of the flame from the substrate surface on the initial diamond nucleation during the initial synthesis stage, we observed the substrate surface after the first 120 s of synthesis. The results reveal that the state of nucleation was affected by the variations in distance. This indicates that the state of nucleation is affected when the distance is varied, the density of nucleation on the substrate also decreases as the distance increases, and the size of the nuclei on the substrate becomes small as the distance increases. Thus, the surface roughness of the synthesized film was observed to have been affected. This can be attributed to the difference in the rate of nucleation owing to the variation in distance. The nucleation rate was high when the distance of the inner cone of the flame from the substrate surface was short. The nucleation rate tended to be lower when the distance was long. It can also be observed that the size of the nucleus grew faster, tended to be larger when the distance was shorter, and tended to be smaller as the distance increased. A long distance is considered to reduce the rate of nucleation and the density of the nuclei produced on the substrate. These results indicate that the variations in the distance of the inner cone of the flame from the substrate surface affected the nuclei formed during the initial stages of synthesis. This, in turn, affected the synthesis of the diamond film. This can be attributed to differences in the nucleation rate and nucleus growth owing to the variations in the state of the field in the combustion flame caused by the variations in the distance of the inner cone of the flame from the substrate surface. These differences in nucleation rate and nucleus growth are likely to have been caused by the close relationship between the field of the acetylene feather and the inner cone within the combustion flame. This

is considered to be attributable to the loss of energy that occurs to an extent as the distance increases. As the distance increased, the size of the nuclei decreased further, and the nucleation rate reduced. This can be attributed to the fact that more energy is lost over longer distances. Thus, if the distance is excessively short, the nucleus tends to grow atypically as the synthesis proceeds. If the distance is excessively long, the field in the combustion flame is not optimal. This results in a state of inadequate growth with a nucleus, small nucleus size, and low density. It can also be considered that a longer distance results in inadequate nucleus growth or a decrease in the density of the nuclei. Moreover, the size of the nuclei was reasonably small in the field during the combustion flame owing to energy loss. The field in the combustion flame is considered optimal at a distance where the density of the nuclei increases and the size of the nuclei is reasonably large.

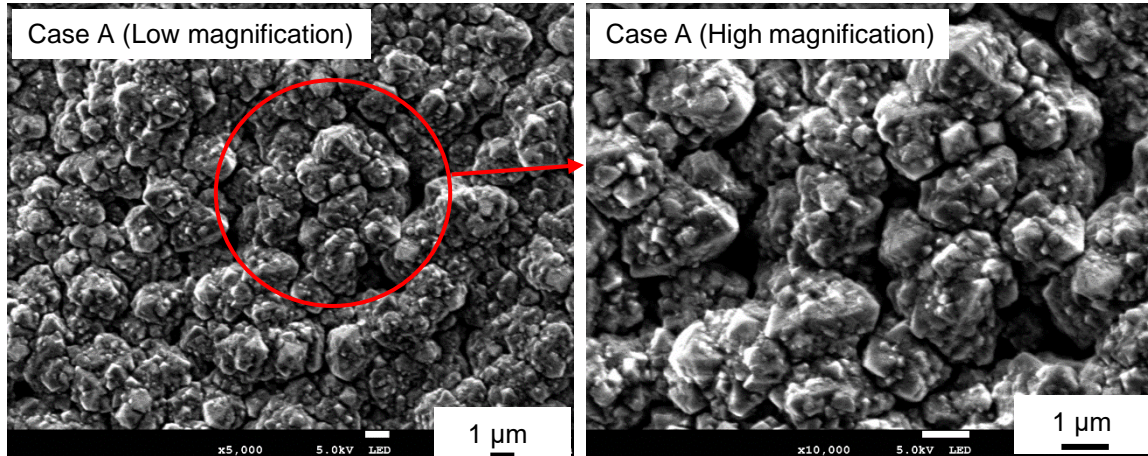


Figure 13. SEM image of the substrate surfaces of the synthesized films after the first 120 s for Case A.

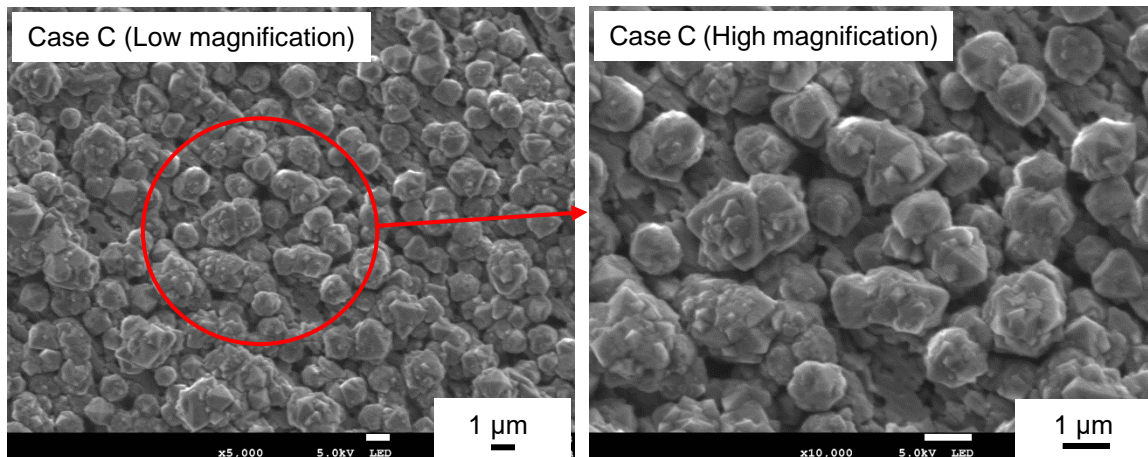


Figure 14. SEM image of the substrate surfaces of the synthesized films after the first 120 s for Case C.

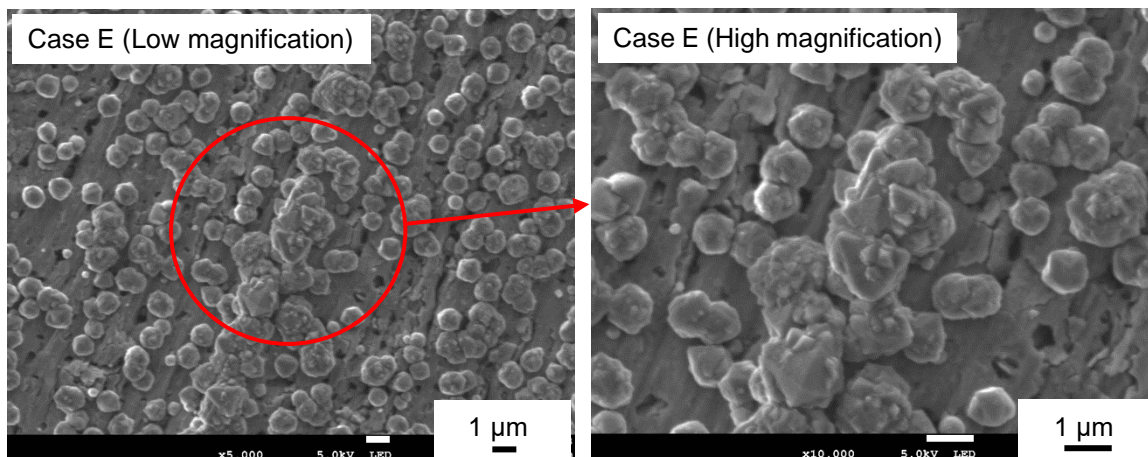


Figure 15. SEM image of the substrate surfaces of the synthesized films after the first 120 s for Case E.

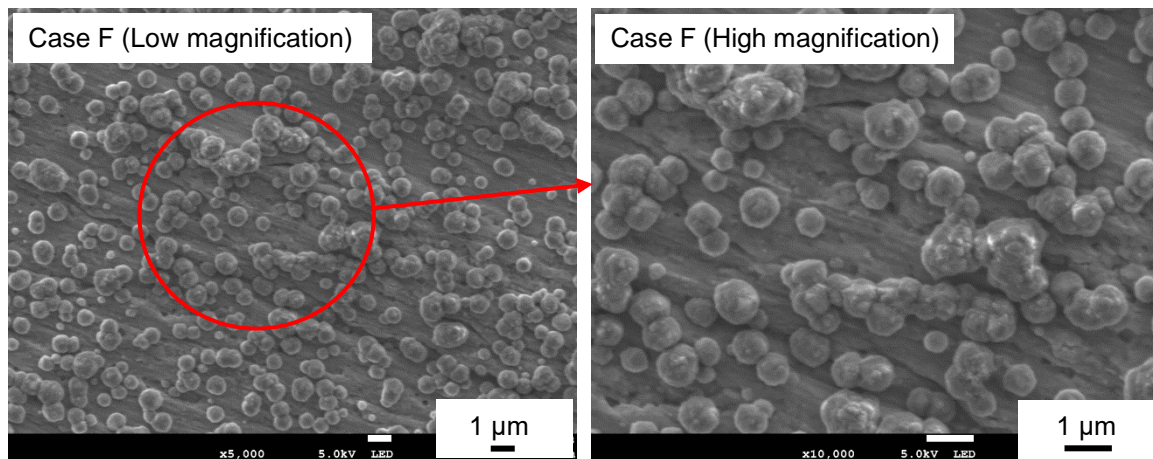


Figure 16. SEM image of the substrate surfaces of the synthesized films after the first 120 s for Case F.

As the distance of the inner cone of the flame from the substrate surface varied, the fields of the acetylene feather and inner cone changed. This altered the number of radical groups in the acetylene feathers required for diamond synthesis. This is considered to affect diamond nucleation and diamond film synthesis.

Here, as the distance of the inner cone of the flame from the substrate surface decreases, the nucleation rate, size of the nucleus, and density of the nucleus tend to increase. In addition, as the distance of the inner cone of the flame from the substrate surface increases, the nucleation rate, size of the nucleus, and density of the nucleus tend to decrease. The basic concept of the nucleation state in each case is illustrated in Figure 17. In Case A, the synthesized crystallites were large and had a high density. This was because the distance was 1.0 mm, which was the shortest distance used in this experiment. As illustrated in Figure 17 (a), in Case A, because of the short distance, the new nuclei generated at the mountaintop during the growth of the nuclei at the bottom of the valley during synthesis were large in size. In addition, for diamond synthesis, the distance was 1.0 mm, which is the shortest distance. Thus, the growth energy of the nuclei was dispersed moderately over the nuclei. This resulted in large crystallite sizes and a high density. In Case C, the synthesized crystallites were smaller and had a lower density than those in Case A. This was because the distance of the inner cone of the flame from the substrate surface was 2.0 mm, which was longer than the distance used in Cases A and B. As illustrated in Figure 17 (b), in Case C, because of the distance being longer than those in Cases A and B, the newly generated nuclei at the mountaintop during the growth of the nuclei at the bottom of the valley during synthesis were smaller than the newly generated nuclei of Cases A and B. In addition, for diamond synthesis, the distance was 2.0 mm. This was longer than those for Cases A and B. Thus, the growth energy of the nuclei for the nuclei produced was lost moderately over the nuclei. This, in turn, resulted in the synthesized crystallites having a small size and low density compared with the synthesized crystallites in Cases A and B. This was because the distance was 2.0 mm, which was longer than those used in Cases A and B. In Case F, the synthesized crystallites were smaller and had a lower density than those in the other cases. This was because the distance was 5.0 mm, which was longer than the distance used in the other cases. As illustrated in Figure 17 (c), in Case F, because of the longer distance of flame's inner cone from substrate surface than in the other cases, the new nuclei generated at the mountaintop during the growth of the nuclei at the bottom of the valley during synthesis were smaller than those generated in the other cases. In addition, for diamond synthesis, the distance was 5.0 mm, which was longer than those for the other cases. Thus, the growth energy of the nuclei for the nuclei produced was lost over the nuclei. This, in turn, resulted in the synthesis of crystallites with small size and low density compared with the synthesized crystallites in the other cases.

In all the cases, the delamination of the synthesized films was prevented completely when the distance of the inner cone of the flame from the substrate surface was varied. In the previous study [25], the delamination of synthesized films during the scratching process with emery paper # 400 grain size ($R_a = 0.298 \mu\text{m}$) was prevented completely. We considered that the distances affected the diamond film synthesis of the synthesized films. However, in this study, delamination of the synthesized films did not occur when the distance of the inner cone of the flame from the substrate surface varied. For the values of the distance of flame's inner cone from substrate surface of 1.0–5.0 mm in Cases A–F during the scratching process with emery paper # 400 grain size, the distances were good conditions for diamond crystallites for synthesis and non-delamination.

In this study, experiments were conducted to investigate the effects of the distance between the inner cone of the flame and the substrate surface on the crystallite and delamination properties of the synthesized film. The relationship between the distance and state of the diamond generation nuclei on the substrate in the initial stages of synthesis was verified. When the distance was altered, the diamond film synthesis was affected because the

acetylene feather field varied. When the distance was short, the diamond growth rate was high. When the distance was large, the diamond growth rate was low.

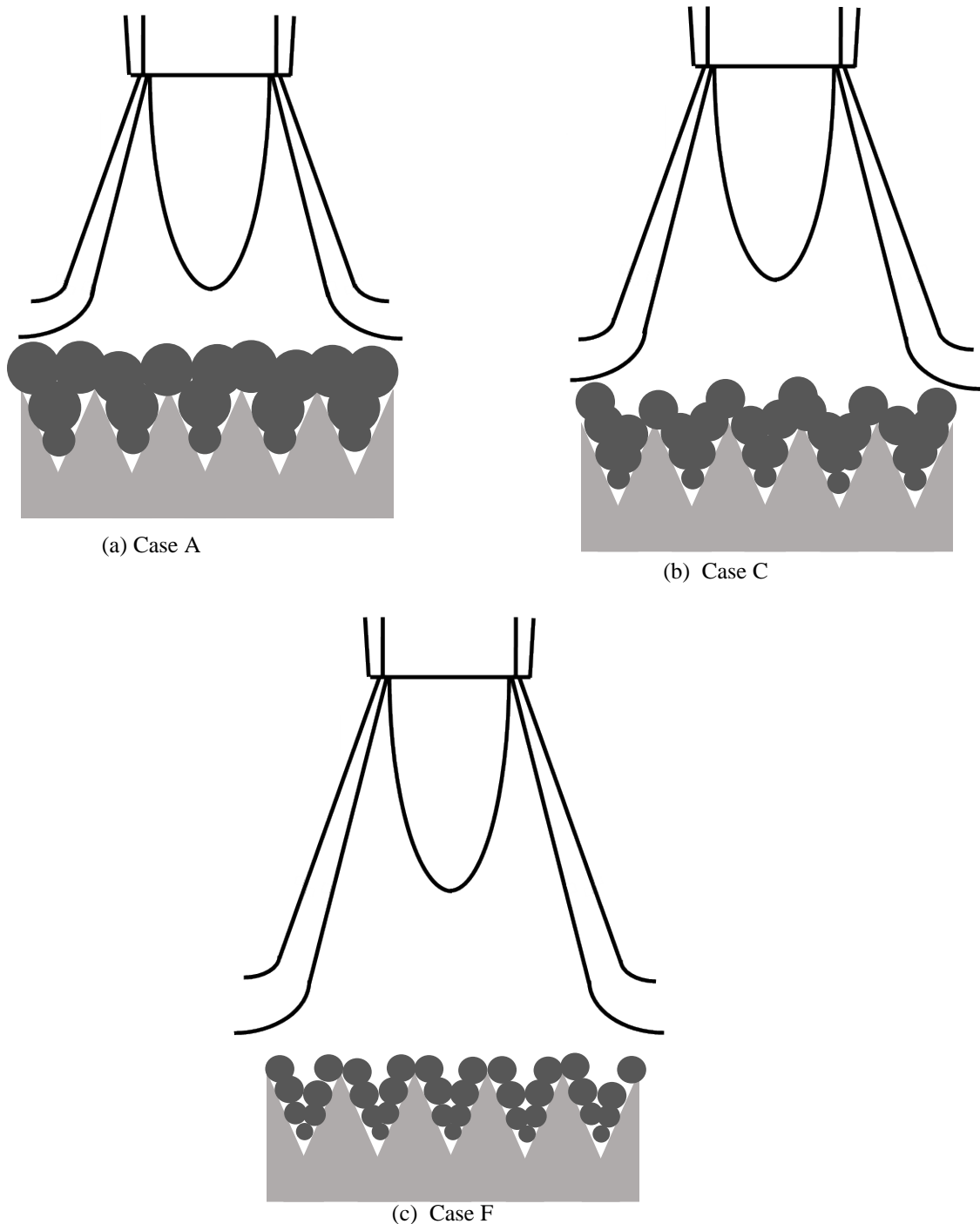


Figure 17. Cross-section morphologies of nucleation of diamond for Cases A, C, and F.

4. Conclusions

In this study, diamond films were synthesized on a Ti substrate (a dental implant material) to improve the strength, wear resistance, and biocompatibility of dental implant surfaces. This was achieved using a flame combustion method with a mixture of acetylene and oxygen. The feasibility of synthesizing a diamond film on a Ti substrate was also examined. The aim was to obtain high-quality diamond films with various synthesized diamond crystallite sizes and achieve good adhesion on Ti substrates for dental implants. The distance of the flame's inner cone from the substrate surface (as a synthesis condition to prevent delamination), the distance of

the flame's inner cone from substrate surface was varied by 1.0, 1.5, 2.0, 3.0, 4.0, and 5.0 mm, respectively. The effect of the distance between the inner cone of the flame and the substrate surface on the diamond films synthesized by flame combustion was investigated.

1) Diamond films were obtained on the Ti substrate by varying the distance of the inner cone of the flame from the substrate surface using a mixture of acetylene and oxygen.

2) The delamination of the synthesized films for various values of the distance of flame's inner cone from substrate surface (1.0, 1.5, 2.0, 3.0, 4.0, and 5.0 mm; Cases A, B, C, D, E, and F) was prevented completely.

3) The surface morphology of the synthesized films was altered by varying the distance between the inner cone of the flame and the substrate surface. A comparison of the films synthesized under all the conditions revealed differences in the crystallites of the synthesized films. The synthesized crystallites became smaller as the distance between the inner cone of the flame and the substrate surface increased. This indicates that when the distance is a parameter, the difference in distance affects the crystallite size of the synthesized diamond film.

4) The mean value of the measured surface roughness (arithmetic mean roughness (R_a)) varied in each case. The measured surface roughness decreased as the distance increased. The results showed that the surface roughness of the synthesized diamond films was affected by variations in distance.

5) A Comparison of the substrate surfaces synthesized for 120 s during the initial synthesis stage under these conditions revealed that the initial diamond nucleation of the synthesized substrate surfaces differed. The state of nucleation was affected when the distance varied. The nucleation density and nuclei size on the substrate decreased as the distance increased. This indicates that when the distance was varied as a parameter, the difference in distance affected the nucleation in the initial stage of synthesis. This, in turn, affected the diamond film synthesis.

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5. References

- [1] Narayan R J, Wei W, Jin C, Andara M, Agarwal A, Gerhardt R A, Shih C C, Shih C M, Lin S J, Su Y Y, Ramamurti R, Singh R N, Microstructural and biological properties of nanocrystalline diamond coatings. *Diamond and Related Materials*. 2006;15(11-12): 1935-1940.
- [2] Lia Y. S, Ye F, Corona J, Taheri M, Zhang C, Sanchez-Pasten M, Yang Q. CVD deposition of nanocrystalline diamond coatings on implant alloy materials with CrN/Al interlayer. *Surface and Coatings Technology*. 2018;353: 364-369.
- [3] Shirani A, Nunn N, Shendeova O, Osawa E, Berman D. Nanodiamonds for improving lubrication of titanium surfaces in simulated body fluid. *Carbon*. 2019;143: 890-896.
- [4] Carcione R, Politi S, Iacob E, Potrich C, Lunelli L, Vanzetti L E, Bartali R, Micheli V, Pepponi G, Terranova M L, Tamburri E. Exploring a new approach for regenerative medicine: Ti-doped polycrystalline diamond layers as bioactive platforms for osteoblast-like cells growth. *Applied Surface Science*. 2021;540: 148334.
- [5] Merker D, Handzhiyski Y, Merz R, Kopnarski M, Peter R J, Popov C, Apostolova M D. Influence of surface termination of ultrananocrystalline diamond films coated on titanium on response of human osteoblast cells: A proteome study. *Materials Science and Engineering C*. 2021;128: 112289.
- [6] Li X, Chen H, Wang C, Chen C, Jiang M, Hu X. Improvement of adhesion under the scratching condition for diamond film on the steel by adding a precarbonization process. *Diamond and Related Materials*. 2023;136: 109927.
- [7] Polini R, Allegri A, Guarino S, Quadrini F, Sein H, Ahmed W. Cutting force and wear evaluation in peripheral milling by CVD diamond dental tools. *Thin Solid Films*. 2004;469-470: 161-166.
- [8] Salgueiredo E, Almeida F A, Amaral M, Fernandes A J S, Costa F M, Silva R F, Oliveria F J. CVD micro/nanocrystalline diamond (MCD/NCD) bilayer coated odontological drill bits. *Diamond and Related Materials*. 2009;18(2-3): 264-270.
- [9] Jetpurwala A M, Dikshit M. Chemical vapor deposition diamond dental burs for high speed air turbine handpieces. *Surface and Coatings Technology*. 2021;418: 127244.
- [10] Takahashi M, Tsutsumi Y. Designing and processing of dental implants. *Materia Japan*. 2016;55(4): 133-136 (in Japanese).
- [11] Hagiwara Y. Future prospects of impact treatment in the viewpoint of super aging society. *Journal of Japanese Society of Oral Implantology*. 2017;30(2): 57-68 (in Japanese).

- [12] Dekkara D, Bénédica F, Falentin-Daudréb C, Rangelb A, Issaouia R, Migonneyb V, Acharda J. Microstructure and biological evaluation of nanocrystalline diamond films deposited on titanium substrates using distributed antenna array microwave system. *Diamond and Related Materials*. 2020;103: 107700.
- [13] Yang B, Li H, Yu B, Huang N, Liu L, Jiang X. Deposition of highly adhesive nanocrystalline diamond films on Ti substrates via diamond/SiC composite interlayers. *Diamond and Related Materials*. 2020;108: 107928.
- [14] Zhang C Z, Li Y S, Tang Y, Sun Y, Tang Q, Hirose A. Nanocrystalline diamond thin films grown on Ti6Al4V alloy. *Thin Solid Films*. 2013;527: 59-64.
- [15] Takahashi M, Yanagi T, Kubo Y, Suzuki R, Pasang T, Kamiya O. Synthesis of diamond films on Ti substrate surface for dental implant by flame combustion, Abstracts of ATEM: International Conference on Advanced Technology in Experimental Mechanics: Asian Conference on Experimental Mechanics, 2019.
- [16] Hirose Y, Okada N, Koike H. Synthesis of diamond using combustion flame in the atmosphere. *Journal of Combustion Society of Japan*. 1989;80: 1-17 (in Japanese).
- [17] Hirose Y. Synthesis of diamond film in combustion flame. *Journal of Japan Institute of Energy*. 1994;73(11): 973-979 (in Japanese).
- [18] Takahashi M, Ito S, Kamiya O, Ohyoshi T. Synthesis of diamond film on molybdenum substrate surface by combustion flame considering the delamination of the interface. *Journal of Solid Mechanics and Materials Engineering*. 2007;1(2): 223-231.
- [19] Takahashi M, Kamiya O, Ohyoshi T. Effects of flow ratio on the delamination of diamond films synthesized by the three-step method using combustion flame on molybdenum substrate surface. *Transactions of the Japan Society of Mechanical Engineers. Series A*. 2007;73(725): 125-130 (in Japanese).
- [20] Takahashi M, Harada Y, Kamiya O, Ohyoshi T. A new method to prevent delamination of diamond films synthesized by the three-step method using combustion flame. *Journal of Solid Mechanics and Materials Engineering*. 2009;3(6): 853-864.
- [21] Takahashi M, Sugawara M, Kamiya O, Ohyoshi T. Synthesis of nanocrystalline diamond films on molybdenum substrate by flame combustion method, *International Journal of Modern Physics: Conference Series*. 2012; 6(1):485- 490.
- [22] Takahashi M, Saito S, Sasaki Y, Saito G, Kamiya O. Effect of nitrogen addition flow rate on bonding strength of diamond films synthesized by flame combustion using high-purity acetylene gas, *Transactions of the Japan Society of Mechanical Engineers. Series A*. 2013;79(806): 1422-1433 (in Japanese).
- [23] Takahashi M, Kamiya O. Effect of nitrogen addition on synthesis of nanocrystalline diamond films on tungsten carbide substrate by flame combustion method using high-purity acetylene. *Advanced Materials Research*. 2015;1110: 277-283.
- [24] Takahashi M, Kamiya O, Pasang T. Effect of pretreatment of substrate on synthesized diamond films on tungsten carbide substrate by flame combustion. *Procedia Manufacturing*. 2017;13: 21-28.
- [25] Takahashi M, Fujita T, Yanagi T, Suzuki R, Kamiya O. Influence of substrate surface roughness on synthesized diamond films by flame combustion on Ti substrate for dental implants. *Journal of Materials and Applications*. 2022;11(1):17-26.
- [26] Takahashi M, Kamiya O. Influence of diamond seed attachment processing on diamond films synthesized on tungsten carbide substrate by flame combustion. *Journal of Materials and Applications*. 2023;12(1):11-20.



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