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Revealing the effects of aerosol deposition on the substrate-film interface using NaCl coating

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Abstract

Aerosol deposition is a feasible method of fabricating dense ceramic films at room temperature by the impact consolidation of submicron-sized particles on ceramic, metal, glass, and polymer substrates at a rapid rate. Despite the potential usefulness of the aerosol deposition process, there are issues, such as deposition mechanisms and structure of the film-substrate interface, that are not well understood. We have used complementary structural and microstructural analysis to capture the state of the substrate surface after the aerosol deposition process. The results reveal that modification of the substrate surface by the ejected submicron-sized particles is essential for the formation of anchoring layer, thereby, a change in internal residual stress state and surface free energy of the substrate is required to deposit film using AD process. Our analysis also suggests that the adhesion between the metal substrate and ceramic particles is possibly contributed by both physical bonding and mechanical interlocking.

Introduction

Aerosol deposition (AD) is a room-temperature film deposition method that uses high kinetic energy of ejected aerosol of a mixture of submicron sized solid particles and a carrier gas, *e.g.*, O₂, air, He, N₂, *etc.*, from a nozzle.(1-6) It is a kind of spray coating method that works under vacuum condition without utilizing any thermal treatment of the carrier gas, sprayed particle, or substrate. The rapid fabrication of high-density films of functional ceramics on different substrate materials, such as metals, ceramics, glass, and polymers as well as the cost-effectiveness, has established AD as a potential method in numerous industrial applications. Among others, the development of microelectronics, sensors, biological implants, solid-oxide fuel cells, solar cells, corrosion-resistive coatings are the potential fields of application. Research on the AD method is expanding in various fields, as evidenced by the increasing number of academic publications(7, 8) and patents.(9-13)

Depending on the carrier gas and nozzle design, it is considered that the particle impact velocity of the ejected particle in AD is generally between 150 – 600 m/s.(1, 7) The range of aerosolized particle velocity makes AD different from the cold-spray (CS) method, a widely used technique for the deposition of metal powders mostly on metallic substrates. In the cold-spraying method, the carrier gas stream is heated to increase the velocity, generally between 500 to 1000 m/s, of the ejected particles through a de-Laval nozzle.(14, 15) Therefore, the required energy for a particle to substrate bonding as well as particle-to-particle bonding in the cold-spraying process is most likely different than in AD.

The deposition mechanism of metallic particles on metallic substrates using the CS method is relatively well understood and is generally considered to be associated with viscoplastic deformation of micron-sized, *e.g.*, 10 – 50 μm , impacted solid particles.(16) It should be noted that the CS method, although highly practical for the deposition of ductile materials, it is not that suitable for brittle materials, *e.g.*, ceramics and glasses.(16) For brittle materials, AD is the best-suited method and the deposition window based on particle impact velocity is generally considered to be between 150-600 m/s.(1) However, the deposition mechanisms, *i.e.*, impacted particle-to-substrate bonding and particle-to-particle bonding in the AD process are not well established. As the AD process works well for the fabrication and integration of dissimilar materials, *i.e.*, ceramics on metal,(8) ceramics on polymer, ceramics on glass,(17) or metals on ceramics,(18, 19) it is most likely that the bonding characteristics and the adhesion strength are related to the difference in film and substrate material properties, such as mechanical, chemical, and thermal properties.

Several theoretical and experimental works have focused on film deposition mechanisms.(1, 6, 20, 21) The possibility of partial or total melting of the ceramic particles at the local scale during collision has been ruled out by simulation and experimental results.(1, 7) However, the local temperature rise at the substrate-film interface should be considered with respect to the thermal conductivity of the substrate materials, *e.g.*, metal can dissipate heat more easily than the glass. Therefore, it is possible that particles with the same mechanical and thermal properties will experience differences in the local heat generated at the substrate-film interface. In addition, the film density has been reported to be related to the plastic deformation ability of the starting particles. A recent study by Exner *et al.*(22) suggested that the densification mechanism requires a fracture of crystallites rather than the impacted particles alone. Nevertheless, comprehensive evidence of the formation mechanisms remains a crucial area of study.(1, 23-27)

Similar to other gas deposition methods, the film formation and physical properties of AD films depend on the mechanical properties of the substrate and impacted solid particles. For example, the surface roughness of an AD-deposited Al_2O_3 film was found to increase from 2 nm to 50 nm for a change of substrate elastic modulus from 117 GPa to 375 GPa.(7) The AD process is considered to be a two-stage deposition process, *i.e.*, formation of anchoring layer by interlocking the particles with substrate surface and then impact consolidation of subsequent particles.(6) The modification of the substrate surface is assumed to be essential for the formation of the anchoring layer. High-resolution TEM has been used to reveal a 100-150 nm thick damaged layer in the $\text{Pb}(\text{Zr,Ti})\text{O}_3$ ceramic film-Si substrate interface.(2) It is known that the film-substrate interface influences the electrical properties of dielectric ceramic films. From these works and considering the principles of spray coating, it is evident that for successful film deposition the modification of substrate surface during the deposition by the impacted particles is crucial.

Naoe *et al.*(28) showed that, for Al_2O_3 particles, the deposition efficiency and film-substrate interface depends on the particle impact velocity. In addition, by using electron energy-loss spectroscopy and previously reported first principles calculation,(29) the authors inferred the existence of ionic and covalent bonding at the interface of Al_2O_3 ceramic films on Cu substrates. Therefore, for the understanding of the deposition mechanisms as well as their effect on film quality, investigation of the post-deposited substrate surface is crucial. However, report on the actual substrate surface after film deposition is scarcely available, possibly due to the difficulties in achieving an unperturbed as-deposited substrate surface.

For example, heat-treatment can be used to remove the film from the substrate by utilizing the thermal expansion mismatch of the film and substrate. However, a heat-treatment step will also likely alter the as-deposited substrate properties due to the thermal expansion and possible damage during film removal. Lee *et al.*(30), however, showed hole-like craters on the Cu substrate after one scan during the deposition of Al₂O₃ ceramic film, although they did not fully remove the ceramic film.

Here, we have developed a simple method to reveal the AD deposited substrate surface and directly observed the effect of the deposition process on the substrate. For this purpose, NaCl was used to form an AD film, which can be easily washed away using a polar solvent, such as water. NaCl films were also deposited on different substrates with varying elastic moduli to observe the changes to the interface during AD deposition. This work demonstrates that to form a successful AD film, it requires *in situ* modification, such as a change in surface free energy and internal stress state of the substrate in addition to the fracture of the impacted brittle ceramic particles.

Experimental Methodology

Commercial NaCl powder was used to coat stainless steel (SUS 304), commercial silica float glass, and c-plane oriented sapphire with the AD process. Substrate materials were chosen to cover a range of materials with different elastic modulus. As-received NaCl particles were roll-milled for 15 h using n-hexane, a non-polar solvent, to obtain a median particle size $d_{50} \approx 0.8 \mu\text{m}$, with $d_{90} \approx 2.7 \mu\text{m}$. The vacuum dried powder (200 °C for 24 h) was deposited using N₂ carrier gas with a flow rate of 16 l/min. Film deposition parameters are listed in Table 2. Details describing the AD process can be found in previous works.(31, 32) The NaCl coating was washed using distilled water to obtain the post-deposited substrate (Post-AD). The use of a NaCl film allowed for direct observation of the post-deposited substrate without any mechanical or thermal alterations. Structural and microstructural investigations were conducted on the pre-deposited (Pre-AD), deposited and Post-AD substrate. Change in surface roughness and surface microstructure of the substrates were analyzed using a laser profilometer (UBM Messtechnik GmbH, Ettlingen, Germany) and a scanning electron microscope (Quanta 200, FEI Co., Hillsboro (OR), USA), respectively. A Bruker D8 Advance diffractometer (Bruker AXS GmbH, Karlsruhe, Germany) with a tilting stage was used to collect x-ray diffraction patterns for the analysis of internal stress state, *i.e.*, variation in $d\text{-}\sin^2\psi$ slope of the pre- and post-deposited substrates. Contact angle measurements with three different solvents (formamide, water, and diiodomethane) were carried out on the pre-

and post-deposited stainless-steel substrates. Such contact angle measurement can be used to investigate the surface free energy according to the Owens, Wendt, Rabel, and Kaelble (OWRK) method.(33-36)

Table 1 NaCl film deposition parameters

Average starting particle size	0.8-1.2 μm
Substrate thickness	1 mm
Substrate material	SUS 304, window glass, sapphire
Carrier gas	N_2
Number of scans	20
Velocity of scan	5 mm/s
Nozzle to substrate distance	3 mm
Size of the nozzle orifice	10 $\times$ 0.5 mm $\times$ mm
Pressure in the deposition chamber	~14 mbar
Carrier gas flow rate	16.0 l/min
Deposition area	10 $\times$ 10 mm $\times$ mm
Film thickness	~2-4 μm

Results and Discussion

Figure 1 shows the x-ray diffraction patterns of the as-processed NaCl powder, a SUS304 substrate with NaCl coating, and the post-deposited SUS304 substrate. The surface microstructure of the AD-processed NaCl coating on SUS 304 is shown in Figure 1b. The surface microstructure shows typical well-compacted particles with inhomogeneous size, a result that is consistent with previous AD-processed coating.(7) It is worth noting that the NaCl coating as shown in the photograph in Figure 1b is stable in the normal lab environment, *i.e.*, no crack or delamination of the salt film was observed. The x-ray diffraction patterns indicate that the NaCl film has the same crystal structure as the powder NaCl, *i.e.*, no apparent change in the crystallographic phase was evident. It has been reported that the crystallite size reduction is required for the deposition of AD film,(22, 37) which can be detected by a quantitative evaluation of the XRD pattern using the Scherrer equation.(38) A change in NaCl crystallite size was evident due to the deposition process in this work. It was found that the NaCl powder crystallite size decreased from 46 nm to 19 nm in the AD film, indicating particle fracture during the deposition process. This change in crystallite size also highlights that the NaCl deposition process is similar to that in other ceramic material

systems that have been deposited using AD.(6) The diffraction pattern of the Post-AD substrate clearly shows the absence of NaCl diffraction patterns, signifying that the NaCl coating is completely removed with water. Therefore, it is possible to analyze the post-deposited substrate surface using this water-soluble material.

Figure 1 (a) X-ray diffraction patterns of NaCl powder, NaCl coated SUS304 substrate, and post-AD substrate, i.e., NaCl removed SUS304. (b) SEM image of the AD processed NaCl coating. Inset in b shows the photograph of the 10 mm $\times$ 10 mm NaCl coating on the SUS 304 substrate taken 48 hr after AD processing.

Figure 2 shows the surface microstructures of pre- and post-deposited substrates. Pre-AD substrates in Figure 2 (a-c) show homogeneous smooth surfaces, signifying a flat, polished surface, which is typically a requirement for the deposition of AD films. It was

reported that substrate surface roughness influences the AD film quality and substrate surface roughness in the range of 0.04-0.12 μm is suitable for ceramic substrates.(39) Figure 2 (d-f) shows the post-AD substrate microstructure with an apparent change in the surface microstructure, in particular for SUS 304 and glass. It is worth mentioning that the deposition parameters, *e.g.*, carrier gas, carrier gas flow rate, nozzle to substrate distance, were kept constant for all the substrates. Therefore, the difference in the microstructures between the post-AD substrates is solely related to the difference in their mechanical properties. Using the process parameters listed in Table 1, almost no variation in surface microstructure could be observed for the sapphire substrate. This is possibly related to the mechanical properties of sapphire, *i.e.*, its higher hardness. Although a few reports(23, 40, 41) have demonstrated the deposition of ceramic films on sapphire substrates, in our work, it was found that NaCl deposition on the sapphire was not efficient, indicated by a non-continuous film with inhomogeneous film thickness. The formation of an anchoring layer as well as the deposition rate is highly dependent on the hardness of the substrates. For instance, it was shown that higher particle impact velocity was required to deposit Al_2O_3 ceramics on a harder sapphire (3200 Hv) substrate than on a Cu (95 Hv) substrate.(41) Additionally, it should be noted that the particle velocity in the AD process depends on the nozzle-design/type, carrier gas type. Therefore, it is possible that the particle velocity generated in our set-up is different from those reported previously.

Figure 2 Pre-AD and Post-AD substrate surface microstructures of (a),(d) SUS 304, (b),(e) glass, and (c),(f) sapphire.

Analysis of surface roughness using a laser profilometer is shown in Figure 3. Except for the sapphire substrate already discussed in Figure 2, a clear change in the substrate surface roughness of pre-AD and post-AD (the area between the dotted lines) is visible. Estimated R_a values of pre- and post-AD substrates and their percentage changes, as a result of the deposition process, are listed in Table 2. From the profilometer data and calculated roughness values, it is clear that the SUS 304 substrate has the roughest post-AD surface and corresponds well with the microstructure data shown in Figure 2 (a) and (d). It is interesting to note that the substrate with the lowest elastic modulus, *i.e.*, glass (~ 60 GPa), did not exhibit the maximum post-AD substrate surface roughness. Although the carrier gas flow rate, and therefore particle impact velocity, was the same for SUS 304 and glass, the former showed significantly higher surface deformation due to the impact. Therefore, it can be stated that in addition to the elastic modulus, mechanical deformability of the substrate should also be considered for the formation of the anchoring layer. Previous studies indicated that ceramic film deposition on the metallic substrate is easier than that on a ceramic substrate.⁽³⁹⁾ This is possibly related to the ductility of the metal substrate, *i.e.*, the ductile substrate can plastically deform more easily during particle impact.

Figure 3 Surface profiles of (a) SUS 304, (b) glass, and (c) sapphire substrates. The dotted lines (1 mm width) indicate the post-AD area on the substrate.

Table 2: Change in substrate roughness, R_a , due to the AD process.

Substrate	Modulus GPa	Pre-AD R_a (μm)	Post-AD R_a (μm)	Change in R_a (%)
SUS 304	180	0.011	0.090	720
Glass	60	0.014	0.021	50
Sapphire	462	0.010	0.010	0

Figure 4 shows the sample tilt angle, ψ , dependent diffraction patterns of the pre- and post-AD SUS 304 substrate and analysis of internal residual stress state using the $d\text{-}\sin^2\psi$ method. The change in peak position as a function of ψ is more prominent in the post-AD substrate than the pre-AD substrate (Figure 4 a and b). It is known that data obtained by such measurements can be used to plot the $d\text{-}\sin^2\psi$ relationship (Figure 4 c) and the slope is related to the internal stress of the sample.(42, 43) Using the x-ray elastic constant of the material it is possible to calculate the extent of internal stress. For the analysis of internal stress, the (200) reflection was chosen, and the corresponding x-ray elastic constant was taken as $7.63 \times 10^{-6} \text{ MPa}^{-1}$.(44) From the analysis, it can be seen that the post-AD substrate exhibits a significant internal residual in-plane compressive stress of approximately 590 MPa. The estimated value is significantly higher than the pre-AD substrate, which is only approximately 26 MPa. Previous investigations have demonstrated using synchrotron x-ray microdiffraction that residual stress in as-deposited films is compressive and maximum close to the film-substrate interface.(32) The data presented here, however, reveals the extent of the residual stress on the substrate surface after the film has been removed, not directly in the deposited film. This clearly indicates that residual stresses are created in the substrate during film deposition, in addition to an increase in surface roughness.

Figure 4 200 Diffraction patterns as function of sample tilt angle, ψ , for (a) Pre-AD and (b) Post-AD SUS 304 substrates. (c) Variation in internal residual stresses due to the AD processing for a SUS 304 steel substrate estimated using the slope of d_{200} - $\sin^2\psi$ plot and x-ray elastic constant, $\frac{1}{2}S_2^{(200)} = 7.63 \times 10^{-6} \text{ MPa}^{-1}$.(44)

In the present case, it is expected that plastic deformation of the steel substrate results in the residual compressive stress at the substrate surface, similar to that induced by shot-peening. These factors likely help to adhere the impacted particles to the substrate, *i.e.*, the formation of the anchoring layer is associated with the particle fracture and *in situ* modification of the internal stress state of the substrate. Theoretical investigations of particle impact in the AD process were reported by several authors.(21, 23, 45-47) For example, molecular dynamics simulations of a single ceramic particle impacting on a ceramic substrate demonstrated that depending on the particle velocity, (generally $> 500 \text{ m/s}$), in addition to the impacted particle fragmentation, planar defects are also induced in the substrate.(45) Therefore, it can be speculated that effective accommodation of deposition stress by the

substrates through the development of internal residual stress may be crucial for the deposition mechanisms and mechanical stability of the anchoring layer.

It is also expected that the surface free energy plays a role in the deposition mechanisms. However, previously it has not been possible to investigate the change in substrate surface free energy due to the AD process. This is because a post-AD substrate surface without any modification was not achievable. Using the method developed in the present work, it is possible to directly investigate the change in substrate surface free energy in relation to the deposition process since the NaCl film can be removed without inducing mechanical or chemical changes to the substrate. In this work, the pre-AD and post-AD SUS 304 substrate were investigated using contact angle measurement. The images presented in Figure 5 show the morphologies of liquid droplets for solvents having different polarities on the pre-AD and post-AD SUS 304 substrate surface. The contact angle measurements in Figure 5 show a slight decrease in contact angle for water and a significant increase in contact angle for the non-polar diiodomethane, suggesting an overall increase in polarity after the deposition process and subsequent removal of the NaCl film.

Contrary to the solvents described above, the contact angle measurements with polar formamide exhibited no significant variation in contact angle, a finding that might be attributed to the sensitivity and hence the deviation of the contact angle measurements. Measurement of contact angle using multiple solvents of different polarity is suitable to calculate the surface free energy according to the OWRK method.(33-36) It is required to measure contact angles with a liquid with dominant polar component, *i.e.*, water, and one with dominant dispersive component, *i.e.*, diiodomethane, to calculate the surface free energy using OWRK method. The estimated surface energies based on the contact angle measurements for the pre- and post-AD samples, presented in Table 3, show a change in surface energy for the SUS 304 substrate due to the AD processing, *i.e.*, the total surface energy changes from 37 mN/m to 31 mN/m. In addition, the total surface energy in the metallic SUS 304 substrate is dominated by the dispersive component. The analysis of the substrate surface energy indicates that the AD processing decreases the total surface free energy and the relative contribution of the polar and dispersive component to the total surface energy is also changed. It is acknowledged that the contact angle measurement and consequently the calculation of surface free energy is related to the surface roughness as well as chemical inhomogeneity of the solid surface.(48) Although the difference in contact angle measurement between a smooth and a rough surface may not be conclusive. Nevertheless, the

calculated values are empirical and the interpretation of surface free energy change in pre-AD and post-AD SUS 304 substrate in this work is still valid.(36, 48)

These changes in surface free energy can be an indication of the energy provided by the AD process to change the substrate surface chemistry. This demonstrates that in the AD process, a fraction of the particle kinetic energy is used to modify the substrate surface free energy. Interestingly, the AD process increases the polar contribution to the substrate surface energy, which means that there is the formation of a dipole moment at the substrate surface. Hence, these dipoles on the substrate surface are a consequence of the AD process, where impacted particles partially physically bond to the substrate at the substrate-film interface. Previously, Naoe *et al.*(49) reported both ionic and covalent bonding at the substrate/film interface of Al₂O₃ deposited with AD on a Cu-substrate, which was suggested to be due to the Cu-O interactions.

	Formamide	Water	Diidomethane
Pre-AD	80.24 ± 1.29 °	102.70 ± 1.00 °	45.39 ± 0.35 °
Post-AD	80.24 ± 0.34 °	99.96 ± 0.50 °	57.11 ± 0.95 °

Figure 5 Contact angle measurements for pre-AD and post-AD SUS 304 substrates using three different solvents.

Table 3 Variation in surface energy estimated from contact angle measurement using OWRK method

Sample	Surface energy, σ_s mN/m	Dispersive component mN/m	Polar component mN/m
Pre-AD SUS 304	37.5±0.3	37.4±0.3	0.1±0.05
Post-AD SUS 304	31.0±0.4	29.9±0.5	1.1±0.1
Glass reference	56.5±0.4	18.8±0.1	37.7±0.4

Considering the post-AD substrate surface structure, chemistry, and mechanical state it can be speculated that the adhesion process in AD is a complex combination of mechanical and chemical interactions, where the impacted particle fracture and *in situ* modification of the substrate surface is essential. It is also important to consider the type of substrate/film pairs, *e.g.* ceramic/ceramic, ceramic/metal or metal/ceramic in order to develop an improved understanding of the substrate-film interface structure in AD.

Conclusions

In this work, the substrate surface after the aerosol deposition process was directly investigated by utilizing NaCl, a water soluble material that can be consolidated into an AD film but also be fully removed to reveal the surface structure at the film/substrate interface without artefacts from film removal processes associated with the thermal and/or mechanical treatment. Surface roughness measurements of three substrates with different elastic properties show a strong dependence on mechanical deformability of the substrate for the creation of an anchoring layer and eventual film formation. In agreement with previous studies, the crystallite size of the starting powder decreases during film formation due to particle fracture. Tilt angle-dependent diffraction patterns of the pre- and post-AD stainless steel substrate reveal the development of compressive residual stress at the substrate surface after AD processing. Combined with a change in surface polarity it is proposed that the formation of an anchoring layer involves a combination of a change in surface chemistry and a physical interaction of the particles with the substrate, which can be viewed independently from the particle-to-particle interactions during the densification and film build-up during the aerosol deposition process.

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