

Fabrication of Y_2O_3 coatings by cold-spray

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Abstract

As a candidate material against plasma etching, yttrium oxide has been coated onto etching chamber by plasma spray technique. However, the plasma spray technique introduces undesirable coating properties such as porous structure and deleterious thermal effects. To reduce the disadvantage of thermal impact, cold spray was used as an alternative technology to deposit thick and dense yttrium oxide coatings. Many studies have shown that the powders suitable for cold spray process should be with a size around $20\mu\text{m}$. However, the Y_2O_3 are ceramics, it is difficult to form coatings by cold spray due to the lack of ductility when using the powders with a size about $20\mu\text{m}$. It is also difficult for nano-particles to get through the bow shock of cold spray process, which may cause deceleration or even deflection of lighter particles away from the surface, and therefore fail to be cold sprayed. In this paper, we use Y_2O_3 with an original average size of 30 nm to form agglomerated Y_2O_3 particles by hydrothermal treatment. After the hydrothermal treatment, the nano-size Y_2O_3 agglomerated together to a size around $20\mu\text{m}$ and then deposited by cold spray. Y_2O_3 coatings were forming in this way. In this research, Y_2O_3 nanoscale powder was tailored into a loose agglomerated structure by hydrothermal treatment, and it was found that the addition of inorganic salt promotes the agglomeration process. Cold spray experiments verified the cold spray suitability of the as-modified particle. Gas temperature greatly affects the coating thickness and microstructure, and optimal spraying parameter was fixed at $600\text{ }^\circ\text{C}$. An excellent yttria coating was successfully fabricated on aluminum alloy 6061 with a maximum thickness of $200\mu\text{m}$ and a low porosity less than 1% using compressed air as propellant gas. The loose aggregated feedstock fractured during impact instead of deformation. It is demonstrated that particle structure is key factor for ceramic deposition by cold spray technique. Copyright © 2019 VBRI Press.

Keywords: Cold spray, ceramic coating, yttrium oxide (Y_2O_3).

Introduction

Plasma etching is used for micro fabrication and semiconductor processing. The environment inside the plasma chamber may expose to the plasma etchant gasses and undergo thermal cycling, and thus lead to failure of chamber materials and particles contamination [1]. The use of coatings is one of the most effective method to reduce the cost of consumables and maintain excellent etching quality. Due to its specific chemical property, high purity yttria coating shows better performance than conventional coatings such as aluminum film and alumina coatings [2, 3]. Plasma spray has been widely used for depositing ceramic coatings. However, the porous structure which is related to the in-flight interaction between gas and molten drop, as well as the cracked morphology caused by residual thermal stress, will degrade the protective performance of coatings prepared by thermal spray [4]. To reduce the disadvantage of thermal impact, cold spray was used as an alternative technology to deposit thick and dense yttrium oxide coatings. Many studies have shown that the powders suitable for cold spray process should be with a size around $20\mu\text{m}$ [5-7]. However, the Y_2O_3 are

ceramics, it is difficult to form coatings by cold spray due to the lack of ductility when using the powders with a size about $20\mu\text{m}$ [8-10]. Thus, we investigated the possibility of preparing Y_2O_3 with nano-particles by cold spray. As we know, it is also difficult for nano-particles to get through the bow-shock of cold spray process, which may cause deceleration or even deflection of lighter particles away from the surface, and therefore fail to be cold sprayed. In this paper, we use Y_2O_3 with an original average size of 30 nm to form agglomerated Y_2O_3 particles by hydrothermal treatment. After the hydrothermal treatment, the nano-size Y_2O_3 agglomerated together to an average size of $20\mu\text{m}$ and then deposited by cold spray.

Experimental

Commercial yttrium oxide powder (Beijing DK Nano technology Co., Ltd.) with a nominal grain size of 30 nm was used as raw material to prepare agglomerated powders by hydrothermal treatment. To obtain feedstock for cold-spray, Y_2O_3 particles were prepared by hydrothermal treatment based on the previous work [11]. The hydrothermal treatment was performed by soaking the Y_2O_3 in distilled water with

addition of ammonium sulfate ((NH₄)₂SO₄). The solution was stirred in an oil bath to keep the temperature at 150 °C for 4 h

Suitability of as-modified particle for cold spray were tested using a custom-made cold spray equipment equipped with a standard De Laval of rectangular cross-section, an aperture of 2 mm × 10 mm, and a throat of 2 mm × 2 mm. The feedstock was propelled by compressed air with varied preheating temperature from 500 °C and gas pressure from 1.5 MPa, and spraying standoff was fixed at 20 mm. The AA6061 substrate coupons were ground with SiC paper up to 800 grit, cleaned ultrasonically in acetone, dried and then sandblasted to remove surface impurities before cold spray.

X-ray diffraction analysis was performed on D/max-2500pc (RIGAKU, Japan). Scanning electron microscopy imaging was obtained on SSX-550 (Shimadzu, Japan) to investigate the morphology of particles and as-sprayed coatings. The size distribution of as-modified yttrium oxide particles were measured by a laser particle size analyzer (MS2000, Malvern, UK). The coating porosity was accurately measured by the metallographic image analysis software (SISC IAS V8.0).

Results and discussion

Hydrothermal treatment of the Y₂O₃ powder

The original nanosized Y₂O₃ powders were hydrothermal treated with the addition of (NH₄)₂SO₄. **Table 1** listed the condition of hydrotreatment. Different mix ratio between Y₂O₃ and (NH₄)₂SO₄ were set to study the influence of addition of inorganic salt on particle structure, which were identified as A₀: ammonia sulfate free, A₁: ammonia sulfate 1% addition, A₂: ammonia sulfate 2% addition, A₃: ammonia sulfate 3% addition, A₄: ammonia sulfate 4% addition.

Table 1. Hydrothermal treatment condition.

No.	Y ₂ O ₃ (g): H ₂ O(ml): (NH ₄) ₂ SO ₄ (g)Ratio /g	T/ °C	t/h
A ₀	20:300:0	150	4
A ₁	20:300:1		
A ₂	20:300:2		
A ₃	20:300:3		
A ₄	20:300:4		

Fig. 1 shows the original nanosized Y₂O₃ powders. It can be seen that the particle size was with an average size of 30 nm, which were agglomerated to 2 μm particles driven by surface energy.

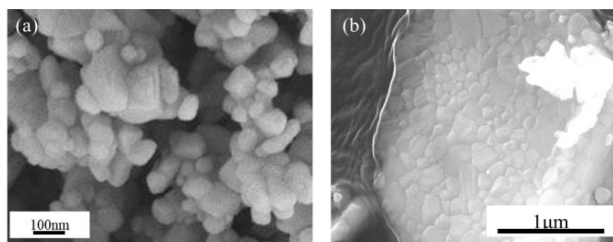


Fig. 1. Morphology of the (a) primary Y₂O₃ particles.

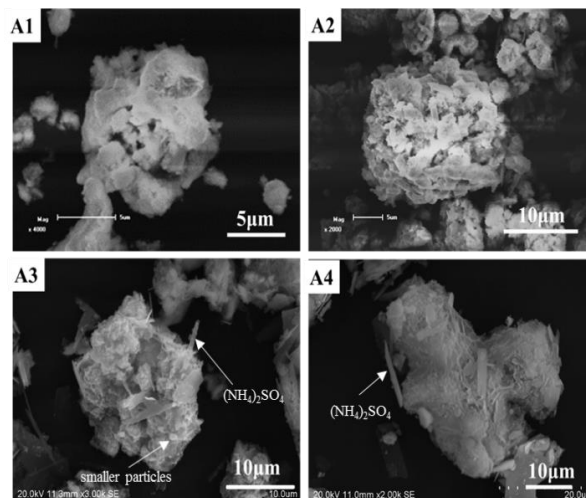


Fig. 2. SEM morphology of hydrothermal treated Y₂O₃ particles with different (NH₄)₂SO₄ addition.

After adding inorganic sulfate (NH₄)₂SO₄, the particles agglomerated into near spherical structure like a snowball. The big particles are clusters of finer powder in a loose agglomeration state with pores between primary particles as shown in **Fig. 2**. After adding little amount of (NH₄)₂SO₄ (**Fig. 2** A₁, A₂), elementary particles agglomerated together loosely and the interval between particles were obvious. When rising the addition of (NH₄)₂SO₄ (**Fig. 2** A₃, A₄), the agglomeration of the particles was getting compact and the morphous of the particle were getting rougher, with small particles adsorbed at the surface of the big agglomerated particles.

Fig. 3 shows the particle size distribution of the modified Y₂O₃ powders. Seen from volume fraction of particles, the maximum proportion of powders without addition (NH₄)₂SO₄ (A₀) is around 1~10 μm, while for those powder hydrothermal treated with the addition of (NH₄)₂SO₄ (A₁-A₄), the majority particle sizes were ranges from 8 to 30 μm. it is almost twice larger than that without the addition of (NH₄)₂SO₄ (A₀). Compared to control test of A₀, it is considered that the existence

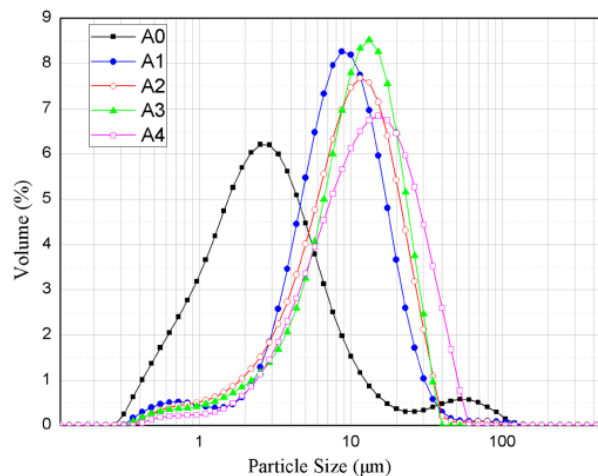


Fig. 3. Particle size distribution of powders after hydrothermal treatment.

of $(\text{NH}_4)_2\text{SO}_4$ plays important role in enhancing the agglomeration process due to the help of electrostatic attraction between the NH_4^+ and SO_4^{2-} ions adsorbed on the surface of Y_2O_3 powder. On account of random sampling, the distribution analysis results basically consisted with that showed in SEM images, from which it can be seen that the mean particle size of powders are approximately 8 μm , 10 μm , 18 μm and 25 μm for the A₁-A₄ powders.

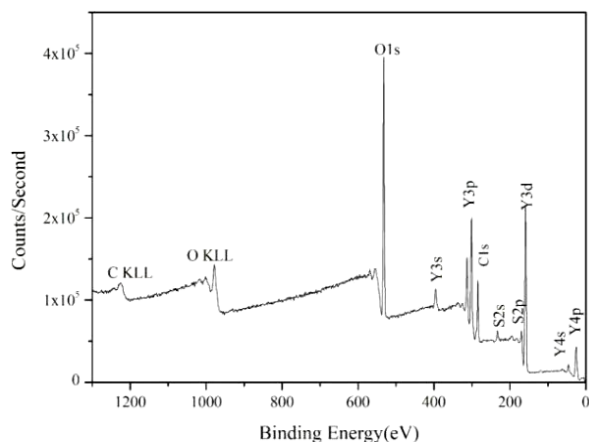


Fig. 4. The XPS wide scan spectra of Y_2O_3 powders after hydrothermal treatment.

Fig. 4 shows the XPS wide scan spectra of the Y_2O_3 after hydrothermal treatment. It can be seen that the weak peaks of S and C element were observed in the spectra, except those peaks of Y and O elements of Y_2O_3 powders. The C elements was probably from the adsorbed CO_2 , while the S element was the residue of additive $(\text{NH}_4)_2\text{SO}_4$ in hydro treatment.

Fig. 5 shows the narrow scan spectra of the treated powders. The spectra of Y3d, O1s, S2p, N1s were analyzed separately. **Fig. 5a** shows two peaks at $3d_{3/2}$ and $3d_{5/2}$ for Y3d. The bonding energy for the two peaks were 158.9 eV and 156.8 eV, which were consistent with that for Y_2O_3 . This result shows that the Y was only bonded with O. **Fig. 5b** shows the narrow scan spectra of O1s, a light shoulder peak was observed. The bonding energy for the two peaks were 529.3 eV and 532.4 eV. The peak with a bonding energy of 529.3 eV was attribute to O1s of Y_2O_3 powders. The peak with a bonding energy of 532.4 eV was attribute to SO_4^{2-} , for the slight adsorption of SO_4^{2-} on the surface of the Y_2O_3 powders. This was consistent with the analysis of S2p with a bonding energy of 169 eV (**Fig. 5c**). **Fig. 5d** shows the narrow scan of N1s, peak at 402 eV was observed, which was attribute to NH_4^+ . It can be concluded that the S and N were only exist in the SO_4^{2-} and NH_4^+ , which was adsorbed on the surface of Y_2O_3 and no chemical bonding were occurred with Y_2O_3 particles. This can be explained that the NH_4^+ and SO_4^{2-} ions were distributed on the surface of the Y_2O_3 powders and then promoting the agglomeration of particles through electrostatic attraction between negative and positive ions [11, 12].

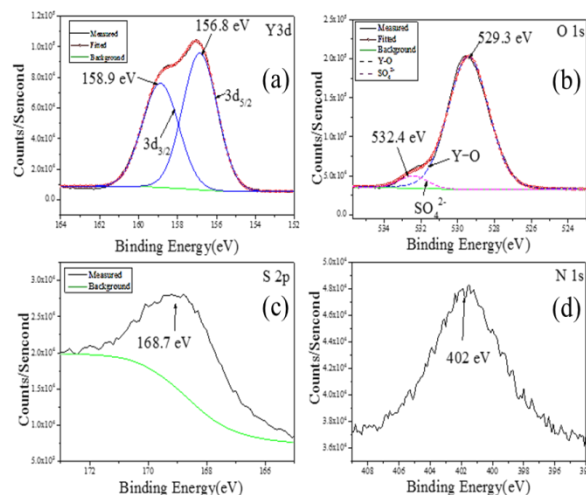


Fig. 5. The XPS narrow scan spectra of hydrothermal treated Y_2O_3 powders for corresponds to (a) Y3d, (b) O1s, (c) S2p and (d) N1s.

Preparation of Y_2O_3 coatings by cold spray

The A₃ powders was used as the feedstock for cold spray to prepare Y_2O_3 . **Fig. 6** shows the SEM images of cold-sprayed yttria coating. It can be seen from the cross-section image that the coating was about 150 μm . The coating was bonded well with the AA6061 substrate. The enlargement image (**Fig. 6b**) showed that the coating was very condense and there were nearly no cracks in the coating. Almost all the size of the particles was no larger than 5 μm , which indicate that the agglomerated structure of A₃ powders were broken during cold spray process.

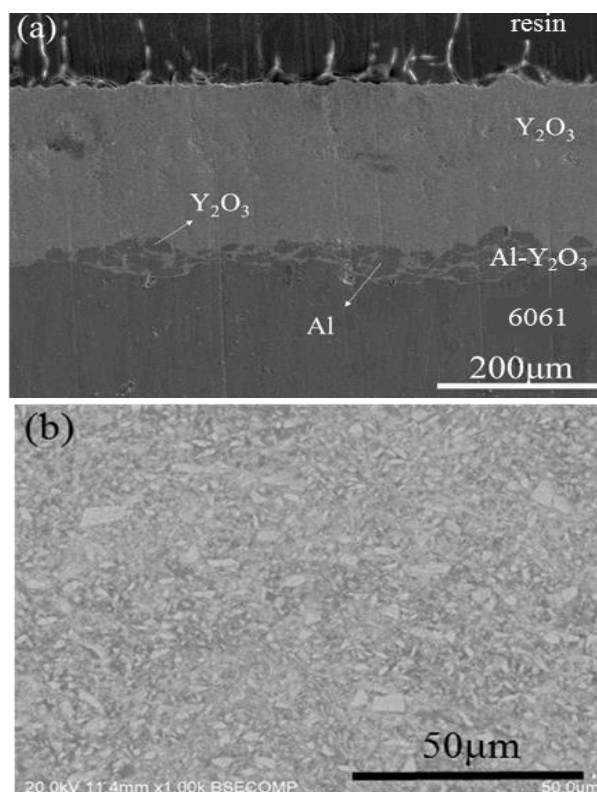


Fig. 6. SEM images of cold-sprayed yttria coating A₃: (a) cross-section and (b) coating.

Conclusion

1. The nano-powder of Y_2O_3 can be agglomerated into larger particles by hydro treatment with the addition of $(NH_4)_2SO_4$ at 150 °C for 4h. $(NH_4)_2SO_4$ can promote the agglomeration of the primary particles effectively. The average size of the agglomerated particles was about 10~30 μm .
2. The powders can be used as feedstock for cold spray process. Very dense and thick Y_2O_3 had been made with the A_3 powders, which was obtained by hydro treatment of the original nano-powder with the addition of 0.9 %wt. $(NH_4)_2SO_4$.

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