Neutron diffraction residual strain measurements in alumina coatings deposited via APS and HVOF techniques

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Abstract. Residual strains in air plasma sprayed (APS) and high velocity oxy-fuel (HVOF) alumina (Al2O3) coatings were investigated by means of neutron diffraction technique. Results of this investigation conclude that through thickness residual strain in the APS coating was mainly tensile whereas the HVOF coating had both compressive and tensile residual strain. Further analysis of Vickers indentation fracture behaviour using acoustic emission (AE) technique concluded that the nature and magnitude of these residual strain fields had a direct effect on the fracture response of two coatings during the indentation process.

1. Introduction
Thermally sprayed alumina (Al2O3) coatings are used in industry for thermal and electrical resistance applications. Measurement of residual strains in these coatings is critical for modeling and designing improved components. Non-destructive through thickness residual strain measurements in thermal spray coatings is possible via the high penetration depth achieved by the neutron diffraction technique [1-2]. The influence of coating process parameters on the residual stress fields of Al2O3 coatings has been a topic of research of previous investigations [3]. Recent advancements in high velocity oxy-fuel (HVOF) technique has made it possible to deposit much finer alumina powders at relatively lower temperatures than those achieved by air plasma spraying (APS) process [4]. This has the effect of limiting phase transformations during coating deposition, which along with the relatively higher velocity and lower temperature of HVOF process can influence residual stress behavior. This paper therefore aims to investigate the through thickness changes in the residual strain profile of finer powder and conventional Al2O3 coatings deposited by the HVOF and APS processes, respectively. Further analysis interlinking fracture behavior and residual strain is also presented.

2. Experimental procedures
Industrially optimized coating process parameters [5] were used to deposit 250 µm thick HVOF and APS coatings on the surface of 31 mm diameter and 8 mm thick AISI 440 steel substrate discs. HVOF (0-gun) coatings were deposited using finer powder (1-5 µm). APS (9 MB) coatings were deposited using conventional coating powder in the size range of 10-45 µm. Both coating powders (98% purity)
had angular and irregular morphology. Coating microstructure was investigated via cryogenic fracture in liquid nitrogen to reveal coating porosity and splat morphology. X-ray diffraction (XRD) measurements were made to reveal phase composition using Cu-Kα radiation ($\lambda = 0.1542$ nm). Neutron diffraction measurements (ENGIN-X) [1-2] were conducted in vertical scan mode to measure through thickness residual strain profile of the coating substrate system. To achieve a through thickness residual strain profile a partial submerged beam was used for measurements near the coating surface, and a beam submerged in the coating and substrate materials near the coating substrate interface. By using the fast vertical scan and careful leveling of specimen using theodolites, the coating surface and the coating substrate interface was located up to a resolution of about 50 µm. Residual strain was calculated via the shift in individual neutron diffraction peaks. Vickers indentation fracture tests and its AE measurement [6] were conducted at nine different loads between 98 and 490 N (five indentations at each load) with an AE sensor on the surface. The data were sampled at 2.5 MHz so that the full band-width of the AE (0.1-1 MHz) could be captured.

3. Results and Discussion

3.1. Coating microstructure
The cryogenic fracture images (Figure 1) reveal the splats morphology, porosity and linear cracks in both coatings. Unmolten Al$_2$O$_3$ particles can also be observed in HVOF coating. This figure also shows relatively higher porosity for APS coatings compared to the HVOF coatings. The coating surface was dense and exhibited a similar structure throughout with no surface connected porosity.

![Figure 1. Coatings cross-sections: (a) APS Al$_2$O$_3$ with thick large splats, and (b) HVOF Al$_2$O$_3$ revealing relatively thin small splats and unmolten particles (indicated by arrows).](image)

![Figure 2. XRD pattern of Al$_2$O$_3$ powder, APS and HVOF coatings.](image)

The XRD spectrum (Figure 2) of APS Al$_2$O$_3$ coatings showed relatively higher $\alpha \rightarrow \gamma$ transformation when compared to the HVOF process, due to rapid solidification and relatively higher temperature of the APS process. It can be seen that the HVOF coating predominantly contained the primary alumina phase ($\alpha$-Al$_2$O$_3$) with very little $\gamma$-Al$_2$O$_3$ transformation.
3.2. Neutron diffraction residual strain and indentation fracture behaviour

Figure 3 summarizes the residual strain results for both coatings, plotted on log-scale to highlight the differences in coating strain. These strain values illustrate the combined effect of phase transformations, thermal mismatch, peening effect, quenching of lamellas and differences in the coefficient of thermal expansion (CTE) of the coating substrate system [1-3]. The difference in CTE of Al2O3 (7×10^-6 °C^-1) and steel (12.2×10^-6 °C^-1) had a significant influence on the net residual strain in the coating layer. The average residual strain in the HVOF coating changes from tensile to compressive, whereas the average strain in the APS coating is tensile balanced by a corresponding opposite strain in the substrate. In APS coating, the effect of higher temperature leading to tensile strain is more dominant due to the CTE mismatch between the coating and substrate materials. Similarly, within the substrate material, the strain at the coating-substrate interface in HVOF coating is close to zero, indicative of low substrate heating during HVOF spraying, whereas the strain at the APS coating-substrate interface is compressive indicative of relatively higher substrate heating. Apart from the temperature difference of APS and HVOF processes, there were three other factors responsible for the relative differences in the residual strain of the two coatings i.e. i) peening effect caused by the relatively higher velocity of the partially melted, though relatively smaller (1-5 µm) Al2O3 particles in HVOF spraying, ii) influence of finer powder particle size on splat morphology, porosity and hence residual stress and iii) relatively higher α→γ phase transformation in the APS coatings. The peening effect in HVOF process is understood to cause compressive residual stress in thermal spray coatings. Similarly, investigations relating to the influence of powder particle size on the residual stress of thermal spray coatings have indicated that the decrease in powder particle size increases the residual stress in thermal spray deposits [7-8]. This effect is attributed to the higher surface area to particle volume ratio of smaller particles, resulting in changes in the pore and intersplat morphology, which resists stress relaxation in the deposited layer. In the investigation by Coats et al. [7] on WC-Co coatings, it was concluded that with the increase in powder particle size the tensile stress in the Co and the compressive stress in the WC decreased. In the current investigation, a similar trend can be observed for the compressive residual strain in the HVOF coating, although the tensile residual strain is lower for the finer powder HVOF coating. The shrinkage in APS coating deposit caused by the α→γ phase transformation [5] also contributes to the residual differences between the two coatings. These results indicate that the tensile residual strain in coatings can be substantially reduced using the fine powders deposited by the HVOF process, which can improve the fracture and delamination resistance of Al2O3 coatings.

![Residual microstrain](image)

Figure 3. Neutron diffraction residual strain profile of Al2O3 coatings and steel substrate.

Figure 4 shows influence of this residual stress profile and microstructural morphology on the indentation fracture behavior of both coatings. Figure 4a,b shows typical fracture morphology of both coatings after Vickers indentation. The fracture morphology for APS coating shows spallation with no fully developed cracks from indent corners. This behavior was different to the HVOF coatings. Figure 4c shows the total AE energy of Vickers indentation fracture of both coatings tested under a variety of loads. It can be observed from this figure that the AE energy increases with load until 245 N, followed by a sudden decrease, after which the AE energy broadly remains constant. This indicates that from...
lower to moderate loads the effect of indentation surface tensile stress dominates the surface cracks and eventually at moderate to higher loads the indentation compressive stress dominates the subsurface cracks. Also in most of the cases the AE energy for APS coatings is higher than that for HVOF coatings, which is due to the combined effect of residual tensile stress and microstructural morphology.

4. Conclusions
It is concluded that the through thickness residual strain profile in the APS coating was mainly tensile whereas the HVOF coating had both compressive and tensile residual strain. The nature and magnitude of these residual strain fields and coating morphology had a direct effect on fracture response of two coatings during the indentation process.

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