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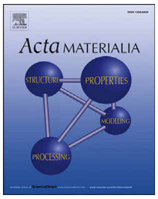

Full length article

Local heteroepitaxy as an adhesion mechanism in aluminium coatings cold gas sprayed on AlN substrates

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ABSTRACT

Cold gas sprayed Al coatings deposited onto wurtzitic AlN substrates show excellent adhesion. As a possible adhesion mechanism, the local heteroepitaxy between Al and AlN was considered and verified experimentally in Al coatings, which were deposited using magnetron sputtering or cold gas spraying on single-crystalline and polycrystalline AlN substrates. Analysis of the local orientation relationships at the Al/AlN interfaces revealed that preferentially such lattice planes of Al align parallel with the upright lattice planes of AlN, which possess similar interplanar distances. The matching lattice planes in the Al coatings grew as continuations of the lattice planes in the AlN substrates. In all samples under study, the parallel alignment of the lattice planes $\{220\}_{Al}$ and $\{110\}_{AlN}$ was found. Additional orientation relationships between Al and AlN arose if parallel lattice planes with similar interplanar spacing could be found in both counterparts via rotation of the lattice planes $\{220\}_{Al}$ around their normal direction. Still, the oriented growth of Al on AlN is only possible if Al atoms in the deposited coatings are mobile enough to rearrange along the AlN surface. Whereas the mobility of Al atoms in a magnetron sputtering process is expected to be sufficiently high, the intrinsic mobility of Al atoms in the cold gas sprayed particles is anticipated to be low. However, the auxiliary microstructure analyses have shown that local recrystallization and partial melting are two phenomena, which can facilitate the rearrangement of Al atoms within the cold gas sprayed coating.

1. Introduction

For many electronic applications, conducting metallic structures deposited on electrically insulating ceramic substrates like Al_2O_3 and AlN are required. In particular AlN is favoured as substrate material for high-power electrical circuits, as it offers a high thermal conductivity, good dielectric properties and a low thermal expansion coefficient. The metallic structures are produced in a metallization process that is usually carried out via cost-intensive and procedurally complex deposition technologies like direct copper bonding or physical and chemical vapour deposition. The cold gas spraying technology is considered as a cost-effective, procedurally flexible and simple alternative deposition technology for the metallization of ceramic substrates [1].

The bonding of kinetically sprayed metal powders on metal substrates is usually explained by metallurgical bonding and mechanical interlocking [2,3], which result from the adiabatic shear instabilities taking place at the particle surface when the impinging particles have a sufficient impact, i.e., when their velocity exceeds a critical value [4,5]. During the impact onto the substrate or already deposited coating, particles with a high kinetic energy experience a high strain rate. As a result of the high strain rate deformation, nearly the complete plastic deformation energy is dissipated as heat [6]. The thermal energy increases the local temperature, which results in softening of the deposited material and even in local melting of the interface region [7–10]. The increase of the local temperature depends on ductility, heat capacity and thermal conductivity of participating materials.

Concurrently, the particle impact leads to a cratering of the substrate and to the formation of a material jet at the impact point between the particle and the substrate. The coating material is then mechanically interlocked with the roughened substrate that improves the resistance of the coating against delamination if no

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cracks are produced in the substrate by the impact of the particles. Additionally, it is expected that native oxide films are broken up or removed from the surfaces of metallic or non-oxide substrates and from the metallic particles by the material jet [3,5,11,12]. The removal of the surface oxides enables a direct contact between the sprayed metal and the substrate which promotes the bonding between the counterparts [5].

For hard ceramic substrates with mirror polished surfaces, the bonding between the sprayed metal particles and the substrate cannot be explained by mechanical interlocking between both counterparts [3], because the hard ceramic substrates do not deform plastically and do not tend to form surface craters upon the impact of the comparatively softer metallic particles. Hence, no undercuts can be formed which would support the interlocking of both counterparts. Nevertheless, a good adhesion of kinetically sprayed metal coatings on ceramic substrates was achieved, which indicates the existence of further adhesion mechanisms in case of Al [1,13,14] or Ti [15] coatings sprayed on ceramic substrates (Al_2O_3 and lead zirconate titanate).

The partial heteroepitaxy, which was found experimentally for titanium coatings that were cold gas sprayed on polycrystalline corundum substrates and sapphire single-crystal substrates [15], is considered as one of the possible mechanisms enhancing the adhesion between the metallic coatings and the ceramic substrates. The local heteroepitaxy between a metal coating and a ceramic substrate was also evidenced for Al coatings that were kinetically sprayed onto atomically smooth (001)-oriented sapphire substrates [16] and that had an average tensile bond strength of (16.3 ± 0.9) MPa [17].

Tensile bond tests performed with the Al coatings that were cold gas sprayed (CGS) onto polycrystalline Al_2O_3 [16], AlN, Si_3N_4 and Si-infiltrated SiC substrates revealed the maximum adhesion strength for the AlN substrates [13]. No correlation of the adhesion strength with the ionicity of the different ceramic substrates was found [18]. Furthermore, the adhesion strength did not correlate with the differences in the coefficients of thermal expansion [18]. For Al deposited on AlN, a superior adhesion was achieved both in the as-sprayed coatings produced at different substrate temperatures (RT, 150 °C and 300 °C) [13] and in the coatings that were annealed at 250 °C, 300 °C and 350 °C [17]. The average adhesion strength of the as-sprayed Al coatings to the AlN substrate was (42.2 ± 4.0) MPa if the substrate was not heated during deposition. The adhesion strength increased to (55.3 ± 6.7) MPa if the substrates were heated to 150 °C and to (65.4 ± 2.0) MPa for a deposition temperature of 300 °C.

This study attempts to clarify the role of the local heteroepitaxy at the Al/AlN interface in the adhesion of the Al coatings deposited on wurtzitic AlN substrates. The discussion is based on a detailed characterization of the interfaces between the CGS Al coatings and different AlN substrates and, in particular, on the description of the orientation relationships (ORs) and the corresponding interplanar spacings of the counterparts. As substrates, (001)-oriented AlN single-crystals and polycrystalline AlN ceramics were employed alternatively. Reference coatings were deposited using DC magnetron sputtering (MS) onto (001)-oriented single-crystalline AlN substrates. The reference samples served for the verification of the OR between face centred cubic (fcc) aluminium and wurtzitic (w) AlN predicted on the basis of the crystallographic symmetry operations and interatomic distances.

2. Experimental details

2.1. Sample preparation

The DC magnetron sputtering was performed in argon atmosphere in a sputter system PLS 500 from Balzers (now Oerlikon Balzers Coating AG, Liechtenstein). Prior to deposition, the chamber was heated to 350 °C to remove the moisture and residual gases from the walls of the deposition apparatus. The base pressure was 3.6×10^{-4} Pa. The deposition was done at room temperature. The argon flow was 32 sccm, the DC power 250 W. An Al target with a purity of 99.999% was used. The MS Al coatings were deposited onto Al-polar (001)-oriented w-AlN single-crystalline substrates with a thickness of 200 μm from CrystAl-N GmbH (Fürth, Germany). The substrates were EPI polished to have a roughness of $R_a < 0.5$ nm. The deposition time was 20 min resulting in a coating thickness of 1.1 μm.

Cold gas sprayed Al coatings were deposited onto Al-polar (001)-oriented single-crystalline w-AlN substrates ($R_a < 0.5$ nm, thickness of 400 μm), onto N-polar (001)-oriented w-AlN substrates ($R_a < 1.7$ nm, thickness of 400 μm), both from CrystAl-N GmbH, and onto polycrystalline w-AlN substrates of the type Alunit® from CeramTech GmbH (Plochingen, Germany). The polycrystalline AlN substrates had a roughness of $R_a = 0.29$ μm and a thickness of 630 μm [13]. According to the phase analysis, they contained 98 vol% w-AlN (space group (SG) $P6_3mc$), 2 vol% of $YAlO_3$ (SG $Pnma$) and $Y_3Al_2(AlO_4)_3$ (SG $Ia\overline{3}d$). $YAlO_3$ and $Y_3Al_2(AlO_4)_3$ are sintering products that originate from the addition of yttrium oxide (Y_2O_3) as sintering additive. All substrates were cleaned with acetone prior to the coating deposition.

Gas atomized aluminium powder with spherical particles (powder fraction 25–45 μm according to the supplier) and a purity of 99.7% from TLS Technik GmbH & Co. Spezialpulver KG (Bitterfeld, Germany) was used as feedstock material for cold gas spraying. The cold gas spraying process was performed with the Kinetics 3000 system from CGT Cold Gas Technology GmbH (Ampfing, Germany), which is equipped with a tungsten carbide Laval type nozzle (Type 27 TC MOC) and a high pressure feeder Praxair 1264. Nitrogen was used as process and powder feeder gas. The gas stagnation pressure was 2.8 MPa, the gas stagnation temperature 350 °C and the nozzle standoff distance 30 mm. For the CGS deposition, a sample holder equipped with a mask, as shown in Ref. [16], was used in order to coat circular sample areas with diameters of 25 mm and 7 mm in case of the polycrystalline and single-crystalline substrates, respectively. During the cold gas spraying deposition process, the substrates were heated to 300 °C by the substrate holder. The nozzle was moved with a traverse speed of 0.3 m/s and a line spacing of 2 mm. The total coating thickness was approximately 140 μm. The adhesion strength of the CGS Al coatings on polycrystalline substrates was determined according to DIN EN 582. Details are given in Ref. [13].

2.2. Sample characterization

The orientation of the Al films that were magnetron sputtered on the (001)-oriented w-AlN single-crystals was determined using pole figure measurements conducted on a PTS diffractometer from Seifert which is equipped with a sealed X-ray tube with Co anode and an Eulerian cradle. The local orientation relationships between the w-AlN substrates and the CGS Al coatings in the vicinity of the Al/AlN interfaces were concluded from the fast Fourier transformations of the high-resolution transmission electron micrographs (FFT/HRTEM) and from the selected area electron diffraction (SAED).

The transmission electron microscopy investigations were done on cross-sectional TEM foils. CGS Al coatings deposited onto polycrystalline AlN substrate and films deposited by MS on (001)-oriented AlN were prepared by polishing dimpled samples with Ar ions. CGS Al coatings deposited on (001)-oriented AlN were prepared by focused ion beam (FIB) technique employing Ga ions. The