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Aluminium coating of lead zirconate titanate—A study of cold spray variables

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ABSTRACT

Cold spray was used to deposit conductive aluminium coatings onto lead zirconate titanate (PZT) piezoceramics. The optimisation of processing parameters was explored. Grain removal from the PZT surface due to the impact of Al particles was reduced by increasing the average particle velocity. Surface domain reorientation was detected by X-ray diffraction (XRD). Substrate temperatures during spraying were maintained at a low level by controlling the upstream, cold spray temperature and robot movement. The electrical resistance of the cold sprayed aluminium was 9.9 ± 0.5 μΩ cm. The impedance characteristics of poled specimens were shown to be unchanged by cold spray.

1. Introduction

Since its initial development in the mid-1980s [1], cold spray has proven to be a coating technique suitable for a wide variety of different materials. In cold spray, powder particles (1–50 μm) are accelerated to velocities between 300 and 1200 m/s through a supersonic (de Laval) nozzle. The particles impact onto a substrate whereupon they deform, flatten and bond, forming a coating. Bonding occurs above a critical velocity, v_c which is dependent on the powder material. It is generally found that ductile metals exhibit a low v_c. However v_c is affected by other factors too, such as substrate material, particle temperature and particle size [2].

Inert gas (typically N_2 or He) is used as the carrier in cold spray. The gas is fed to the nozzle entrance at high pressure (up to 4.0 MPa in some systems). Usually the gas is also pre-heated (up to ~800 °C is possible), so as to achieve a higher downstream gas velocity, and consequently greater particle acceleration. In-flight particle temperatures do not approach the melting point, so dense coatings can be formed without the detrimental effects of thermal spray processes, such as oxidation and melting.

Metallic coatings with high electrical conductivity can be made by cold spray, although this depends on the purity of the feedstock material. Various values of resistivity in cold sprayed copper have been reported; 1.7 ± 0.3 μΩ cm [3], 2.4 μΩ cm [4] and 2.2–3.8 μΩ cm [5]. By comparison, the resistivity of bulk copper is 1.7 μΩ cm. After annealing at 600–650 °C, the resistivity of the as-sprayed material may generally be reduced to within 10% that of bulk copper [5–8]. However, this kind of furnace treatment is not appropriate for all substrate materials. The in-plane resistivity of cold sprayed aluminium coatings has been measured at 10 μΩ cm [9].

Metals may also be deposited onto non-metallic substrates such as ceramics [10]. Metallising of insulating substrates such as alumina for electronics applications has been shown to be a viable application [11,12]. Ceramic substrates however, due to their brittle nature, do present more of a challenge to cold spray, since micro-particle impact inevitably entails a certain degree of mechanical shock to the surface. Furthermore, the possibility of thermal shock in ceramics, and the large difference in thermal expansion coefficient between the substrate and metallic coating limit the use of high gas temperatures during spray. Roughening of the substrate surface by grit blasting, while common practice for improving the adhesion of coatings to metals, may not appropriate for certain, brittle materials.

In Ref. [13] it was shown that cold spray could be used to deposit metallic coatings onto lead zirconate titanate (PZT) substrates as an alternative to current coating methods. PZT is a piezoceramic that is used in a large number of diverse applications that require the efficient conversion of mechanical to electrical energy, or vice versa. Metallic electrodes on opposing faces of a PZT element allow a uniform electric field to be applied. The electrodes are typically sputtered or vapour-deposited thin films or coatings of silver paste that are applied to the PZT surface and then fired. They are used for the 'poling' procedure, during which a large voltage applied at an elevated temperature causes favourably-aligned ferroelectric domains to grow at the expense of others. The same electrodes are typically also kept for service, although for some applications a different electrode configuration is required, meaning additional electrode removal and re-deposition steps. Note that the latter case requires an electroding procedure that does not cause depolarisation

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of the element. A cold spray method for electroding PZT will allow coatings to be built up rapidly and without the need for complicated vacuum equipment. The process is flexible with regards to substrate size and shape. In combination with masking techniques, intricate electrode patterns can be produced.

While it has been demonstrated [13] that aluminium and two-layered aluminium / copper coatings can be deposited, this paper presents a more detailed study of the effect of process parameters on the build up of aluminium on the PZT surface. Optimally, the spray conditions are adjusted to minimise impact damage to the substrate. However, in the following work a range of temperatures, pressures, particle sizes and pauses between coating passes was investigated, to explore the effect of these variables on substrate fracture and grain pullout due to micro-particle impact, heating and possible depolarisation by the gas jet. Finally, an assessment of the coating electrical conductivity and impedance characteristics of the coated elements is given.

2. Experimental methods

Commercial PZT elements were supplied by Fuji Ceramics, (Fuji Ceramics Corporation, Tokyo, Japan). For studies of the effect of cold spray particle impact, two different grades of “hard” PZT were used; C-213 in the form of 20 mm-diam., 10 mm-thick discs, and C-203 in the form of 20 mm-diam., 1 mm-thick discs. The properties of both materials are listed in Table 1. Grain size and porosity were determined by image analysis of polished sections. Both grades contained a significant amount of porosity, while C-213 was considerably finer-grained than C-203. There was a notable difference in mechanical hardness.

All C-213 and C-203 samples were in the polarised condition, with the exception of two C-213 elements. The polarised elements, as received from the manufacturer, had sputtered nickel electrodes on opposing flat surfaces. Polarisation was in the through-thickness direction. Prior to cold spray, the Ni electrodes were removed by wet grinding with SiC paper. Then the surfaces were polished with 1 μm diamond suspension. It should be noted that in normal service, this level of finish is not necessary, however for the purpose of these experiments, it allowed more meaningful interpretation of the effect of cold spray particle impact on the surface state.

Coating was performed using a CGT Kinetiks™ 3000 cold spray system (Cold Gas Technology GmbH, Ampfing, Germany), which has been described in more detail elsewhere [14]. The nozzle was attached to a robot arm, aimed perpendicularly to the flat surface of the PZT substrate at a standoff distance of 20 mm, and was moved laterally in a raster pattern at 0.05 m/s to cover the entire face of the sample. Nitrogen gas was used as the accelerant. The temperature (T_0) and pressure (P_0) of the gas in the stagnation area immediately upstream from the nozzle were varied within the range; $T_0 = 100–300$ °C and $P_0 = 1.0–2.4$ MPa. A sintered tungsten carbide nozzle was provided by CGT with circular cross-section, expansion ratio 10.7 and total length 121 mm.

Aluminium particles were fed centrally through a tube into the converging section of the nozzle, 35 mm upstream from the throat. The average particle size (d_{50}) of the aluminium powder (99.7 wt.% Al) used in the experiments was determined by laser particle size analysis with a Malvern Mastersizer X (Malvern Instruments Ltd., Malvern, Worcestershire, UK). In order to investigate the effect of particle size, the aluminium powder was sieved using a 20 μm mesh. This resulted in three powder feedstocks for grain pullout experiments; (i) the pass-through fraction (d_{50} of 15.6 μm), (ii) the retained fraction ($d_{50} = 33.5$ μm), and (iii) the original, unsieved powder ($d_{50} = 20.3$ μm). Fig. 1 shows the particle size distribution for each feedstock on a cumulative volume fraction basis. Table 2 lists the complete set of conditions used in the experiments.

The velocity v_p and temperature T_p of particles undergoing acceleration through the nozzle were simulated using a one-dimensional model. Similar methods have been used to predict particle velocities in cold spray [15,16]. The predicted exit velocties v_p (exit) and temperatures T_p(exit) from the model are given alongside each experimental condition in Table 2. For the purpose of establishing the gas flow conditions throughout the nozzle the gas was assumed to be isentropic. It was assumed that the entrained particle stream was dilute enough to have no bearing on the gas. It was also assumed that the gas flow was unaffected by any boundary layer effects at the nozzle wall. The model was able to predict particle velocities up to the exit of the nozzle, but no further: out-of-nozzle effects and the bow shock region at the substrate would have required a more sophisticated, 2- or 3-dimensional approach. For the purpose of describing in broad terms the effect of process variables on particle acceleration this simpler method was justified. It has been shown that the bow shock only significantly affects particles with very low inertia, i.e. those with a diameter of the order of a few micrometres [17–19]. The volume fraction of particles below 7 μm was less than 1% in all powder feedstocks used in the experiments (Fig. 1).

According to isentropic theory, the Mach number, M, can be determined at any location in a shock-free, supersonic nozzle from the ratio of nozzle area at that point, A, to the throat area A^*, as per the area-Mach relation, Eq. (1) [20].

$$\left(\frac{A}{A^*}\right)^2 = \frac{1}{M^2}\left[\frac{2}{\gamma+1}\left(1+\frac{\gamma-1}{2}M^2\right)\right]^{(\gamma+1)/(\gamma-1)} \quad (1)$$

where γ is the specific heat ratio. Eq. (1) was solved numerically for M using the bisection method.

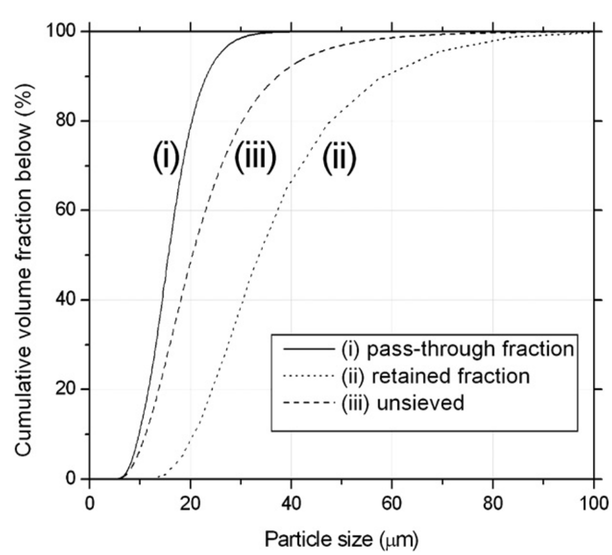

Fig. 1. Particle size distribution of the feedstock powders.

Table 1
Microstructural and mechanical properties of the substrate materials.

PZT grade	C-213	C-203
Grain size (μm)	1.9 ± 0.1	3.9 ± 0.2
Porosity (%)	5.3 ± 0.3	4.3 ± 0.3
Microhardness (HV_{500g})	423 ± 8	281 ± 4
Curie point (°C)[a]	315	350

[a] Manufacturer's data.