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Preparation of composite spherical Al–Y_2O_3 powders by the mechanical shock method and their plasma spray application

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Composite spherical powders made of aluminium and yttria were prepared by the mechanical shock method. This unique idea for composite powder products, without the use of heat, and making use of the ductility of the metal and the brittleness of ceramics, has a preparation time of only 3 min. For analysis of the cross-sections of an Al–Y_2O_3 composite particle, electron probe microanalysis was used. In the composite particle, a diffused layer is formed at the interface between Al and Y_2O_3; and the boundary between these components is thought to be quite strong. These composite spherical powders are excellent regarding fluidity and their spray capability. The Al–Y_2O_3 composite powder was initially plasma sprayed on to an aluminium alloy substrate, and then coated with partially stabilized zirconia. The tensile strength of these test pieces was studied as a function of compositional changes in the Al–Y_2O_3 system. For 0.2 to 1 mol% Y_2O_3, the tensile strength is much higher, values up to 75.5 MPa being attainable. The heat-cycle number of test pieces with the above composition range at the present time has been measured at over 2000.

1. Introduction

Much attention has been concentrated on the techniques of spraying and powder metallurgy. To apply these techniques, powders are required, so as to be available under a wide variety of conditions; thus, production by the mechanical alloying method [1–3], through surface modification of a polymer powder by the mechanical shock method, has recently become of interest [4]. The mechanical shock method uses no heat energy, bowls or water. Several years ago, we reported the results on the interface chemistry and tensile strength of an iron and alumina bonding pair using a composite interlayer of iron–wüstite [5–8]. The idea of a composite interlayer was also used for metal/ceramic bonded materials being applied for plasma spray coating. However, differences in specific gravity, particle size and shape between the metal and the ceramic powder in the mixture tended to disrupt powder transportation.

In the present study, mechanical impact was applied to produce small particles of metal-ceramics. This method is based on a unique idea for composite powder production, without the use of heat, and making use of the ductility of the metal and the brittleness of the ceramics. In this case aluminium (Al) metal and yttria (Y_2O_3) ceramic were used. The bonding behaviour of the boundary region at the Al/Y_2O_3 composite particle was analysed by electron probe microanalysis (EPMA).

These composite powders were used for plasma spray coating. The tensile strength and heat-cycle of these test pieces was studied as a function of compositional changes in the Al–Y_2O_3 system.

2. Experimental procedure

2.1. Methods

To prepare composite particles of metal and ceramics, the following conditions are required.

1. Mechanical energy rather than heat energy must be used.
2. The ductility of the major component metal and the brittleness of minor component ceramics are utilized.
3. The composition ratio of both materials must be controlled. The Nara Hybridization System was used for sample preparation, which uses a mechanical shock process.

The apparatus shown in Fig. 1 was used to provide mechanical impact to the powder. The rotor (3) to which the impact blades (4) are fitted radially, rotates at a high speed in the stator (1). The circulation circuit (5), opens at one end, in a portion of the inside wall of the stator, and at the other end, in the vicinity of the centre of rotor. The discharge port (7) was equipped with a discharge valve (8). The powders were fed into the apparatus through a hopper (6) and carried in the direction of the outer circumference of the rotor while being dispersed by the action of the rotor rotating at

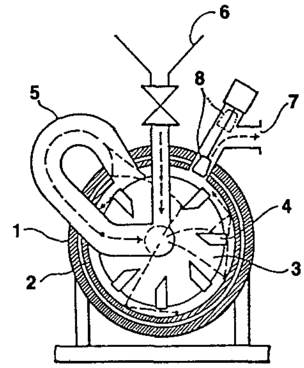


Figure 1 The apparatus used to provide mechanical impact to the powder: 1, stator; 2, heating or cooling jacket; 3, rotor; 4, blades; 5, circulation circuit; 6, hopper; 7, discharge port; 8, discharge valve.

high speed. They repeatedly underwent impact by the impact blades and the inside surface of the stator, again passing through the circulation circuit, before returning to the apparatus. Thus, impact occurred repeatedly. After a predetermined period of time, the discharge valve was opened, and the treated powder was recovered from the discharge port.

2.2. Materials

Atomized powder of aluminium with an average diameter of 40 μm and yttria powder of 99.99% purity with an average diameter of 3.5 μm were used as the starting materials.

The combination of the metal and ceramic in the composite powder has a large degree of freedom. However, the particle size ratio of the major constituent to that of the minor should be preferably more than 1 to 5.

2.2.1. Production of an Al–Y_2O_3 composite spherical powder

The composite spherical powder of aluminium metal and Y_2O_3 ceramics, were prepared by two methods; Figs 2a and b show schematic illustrations of the preparation process. In (a), an irregularly-shaped atomized aluminium powder is spherodized by mechanical impact. Ductile aluminium metal is easily spherodized using the apparatus shown in Fig. 1, at a rotor peripheral speed of 80 m sec^{-1} for 3 min. Then, the spherical aluminium powder and raw fine Y_2O_3 powder are mixed. Finally, they are prepared into composite powder of aluminium and Y_2O_3 using this apparatus for 3 min.

In (b), an irregularly shaped atomized powder of Al and a fine Y_2O_3 powder were intimately mixed. Then, mechanical impact was applied to the resulting mixture using the apparatus at a peripheral rotor speed of 80 m sec^{-1} for 3 min. The spherical composite powder had an average diameter of about 40 μm. Case (b) has two merits: first, the duration of the preparation is only 3 min; second, the surface area of irregularly shaped atomized aluminium powders is essentially greater than that of the spherical aluminium powder. Thus the maximum composition of about 5 mol% Y_2O_3 compared to the aluminium was much greater than the 1.7 mol% in case (a).

2.2.2. A test piece using a composite spherical powder for the plasma spray

Test pieces were prepared for tensile strength and for heat-cycles. For tensile testing, two rods of an aluminium alloy metal, 100 mm long and 18 mm diameter, were used. An interlayer composed of a composite powder Al–Y_2O_3 was plasma sprayed on to the top plane of the aluminium alloy rod, and then ceramic partially stabilized zirconia (PSZ) powder was plasma sprayed. By using a jig, the axes of the two pieces of the aluminium alloy rods were aligned in a straight line. A one-package heat-curable epoxy adhesive was coated on the abutting surfaces, and they were pressed and heated.

An aluminium alloy substrate metal of dimensions of 6 mm height $\times$ 100 mm length $\times$ 50 mm width was used for the heat-cycle test. Half of portion A was plasma sprayed and half of portion B was used for grasping. A thermocouple was inserted in the centre of the aluminium alloy side of portion A. Therefore, the heat-cycle test piece rotates around the rotating shaft. A burner for heating the plasma spraying portion was installed in a certain part of the rotation path of plasma spraying portion A. When the heat-cycle test piece was rotated at a predetermined speed, the spraying portion A repeatedly underwent heating and cooling.

3. Results and discussion

3.1. Interface chemistry

3.1.1. Al–Y_2O_3 system

The composite spherical Al–Y_2O_3 powder was prepared by the two processes shown in Figs 2a and b. In process (b), the duration of the preparation was only 3 min. Figs 3a and b show scanning electron

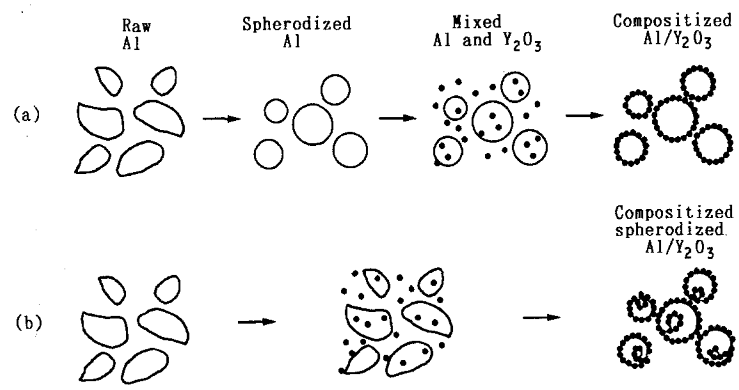
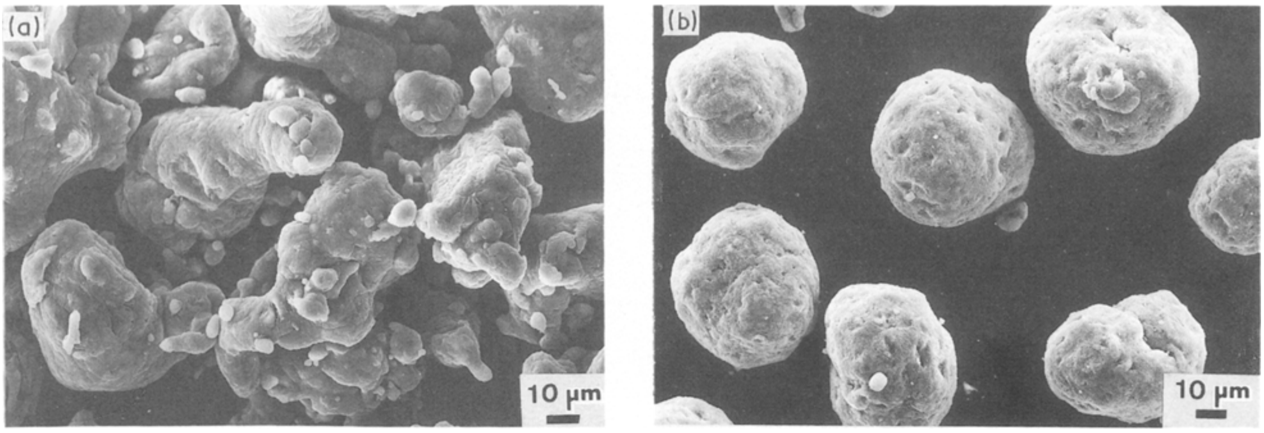


Figure 2 The composite spherical powder of aluminium metal and Y_2O_3 ceramics produced by two methods: (a) and (b) show schematic illustrations of the preparation process.



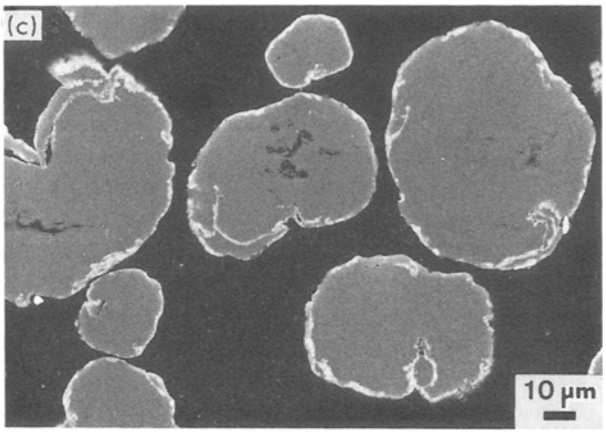


Figure 3 Scanning electron micrographs of the external form and the cross-sections of the particles: (a) atomized aluminium powder used as the raw material; (b) external form of Al–1.7 mol % Y_2O_3 composite spherical powder prepared by the process shown in Fig. 2a, (c) cross-sections of the particles.

micrographs of the external form of the irregularly shaped atomized aluminium powder and of the Al–1.7 mol % Y_2O_3 composite spherical powder prepared by process (a) in Fig. 2. Fig. 3c shows an SEM image of the cross-section of a particle. EPMA micrographs of cross-sections of Al–4 mol % Y_2O_3 composite spherical particles prepared as in Fig. 2b, are shown in Figs 4a to d. In this case, it was observed

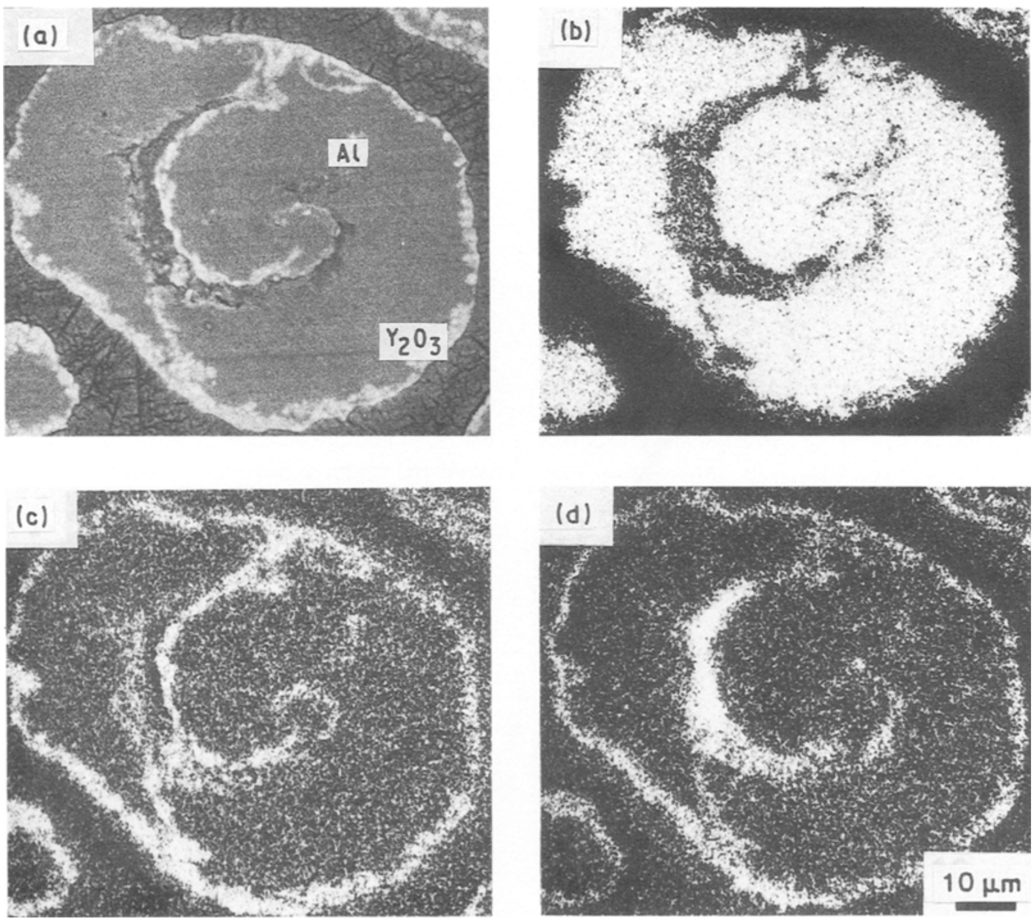


Figure 4 Scanning electron and X-ray image micrographs for cross-sections of a composite spherical Al–Y_2O_3 particle prepared by the process shown in Fig. 2b: (a) SEM, (b) aluminium, (c) yttrium, and (d) oxygen.

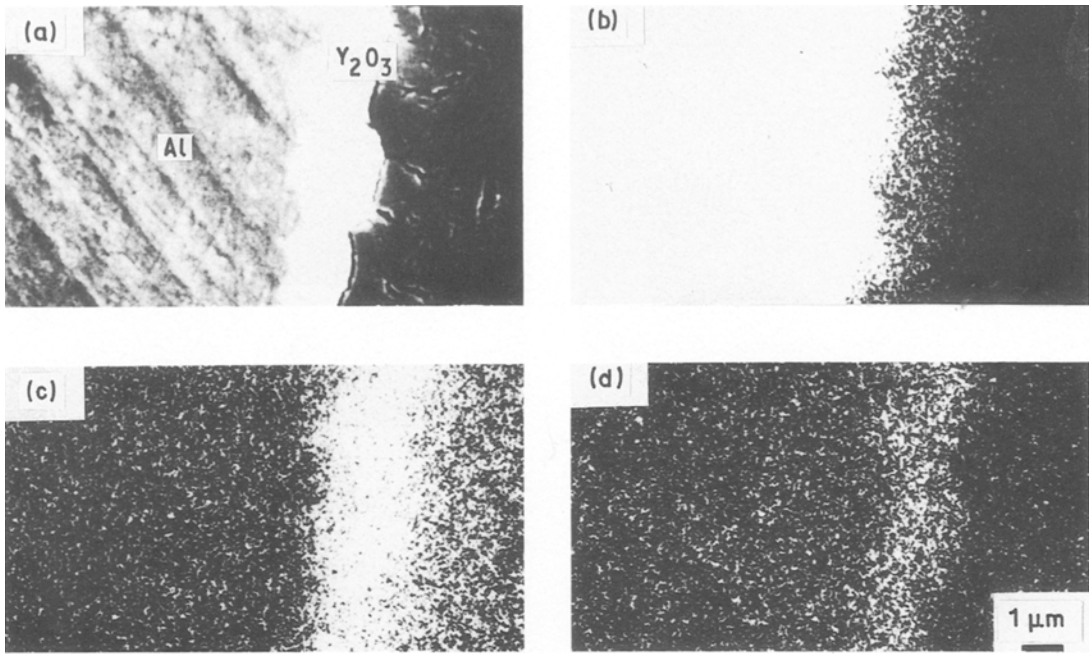


Figure 5 Enlarged scanning electron micrograph and X-ray image micrographs for the boundary region of a composite Al/Y_2O_3 particle: (a) SEM, (b) aluminium, (c) yttrium and (d) oxygen.

that Y_2O_3 adhered to the surface of the irregular aluminium powder and changed shape during the process into a spherical particle. Thus it is possible to increase the composition range of the ratio of Y_2O_3 ceramic to aluminium metal by increasing the surface area of the irregular aluminium powder. The maximum composition ratio also depends on the particle diameter ratio between both materials. Enlarged scanning electron micrographs of the Al/Y_2O_3 boundary region of composite spherical particles are shown in Figs 5a to d. An inter-diffusion like phenomenon is observed between aluminium, yttrium and oxygen through the interface between the aluminium and Y_2O_3.

3.1.2. Fluidities

The starting material and composite spherical Al–Y_2O_3 powder exhibited fluidity; this was measured according to the JIS Z2502 "Measuring method of fluidity of metals". This JIS fluidity test is based upon the time (sec) within which 50 g of the powder flows. The starting aluminium and Y_2O_3 powders were not measured, but aluminium powder and composite spherical Al–Y_2O_3 powders were shown to have good fluidity. The resulting spherical aluminium powder and Al–Y_2O_3 composite powder had fluidities of 58 and 56, respectively.

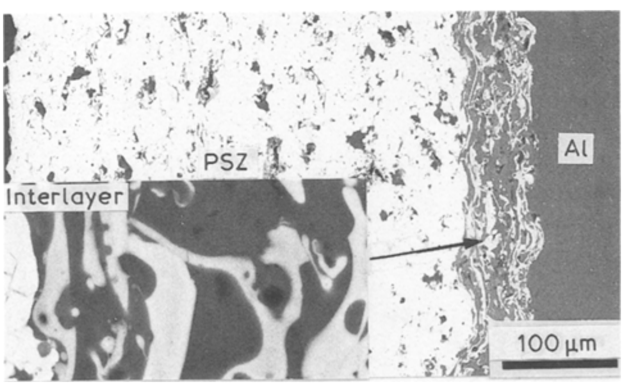


Figure 6 Scanning electron (back-scattering image) micrographs of the cross-sections of the Al–Y_2O_3 composite powder initially plasma sprayed on to an aluminium alloy substrate, and then coated with PSZ.

3.2. Application to plasma spraying

These powders are suitable for plasma spraying. Both the fluidity and the spraying capability of these powders are superior to other commercial powders. The Al–Y_2O_3 composite powder was initially plasma splayed on an aluminium alloy substrate and then coated with PSZ. The coating thicknesses were about 0.1 and 0.3 mm, respectively. Fig. 6 shows scanning electron micrographs of the cross-section in this case.

3.2.1. Tensile strength

The resulting test piece was subjected to tensile testing at a speed of 0.5 mm min^{-1} using an Instron tensile tester. When the structure obtained by bonding metals of Al–Al under optimum conditions was subjected to the tensile test, fracture occurred inside the adhesive and its tensile strength at this time was 75.5 MPa. The tensile strength of these test pieces was studied as a function of compositional changes in the Al–Y_2O_3 system. Fig. 7 shows the composition dependence of the tensile strength. In the case of 0.2 to 1 mol % Y_2O_3, the strength was found to be much higher, and rupture took place within the synthetic resin. The strength was compared with METECO461NS, in case rupture took place between aluminium alloy and METECO461NS; the strength in this system was only 37 MPa.

3.2.2. Heat-cycle

The heat-cycle testing with a cycle time of 30 sec heating (750 K) and cooling (415 K), was repeated 2000

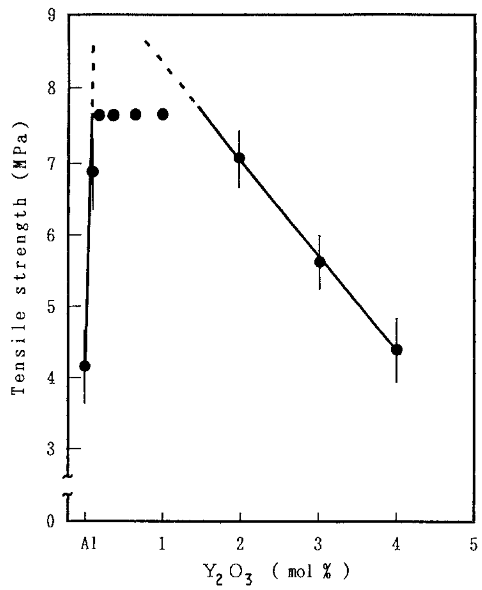


Figure 7 The tensile strength of the plasma-coated test pieces as a function of composition dependence of the Al–Y_2O_3 system. The Al–Y_2O_3 composite powder was initially plasma sprayed on to an aluminium alloy substrate, and then coated with PSZ. (Rupture took place within the synthetic resin at 75.5 MPa.)

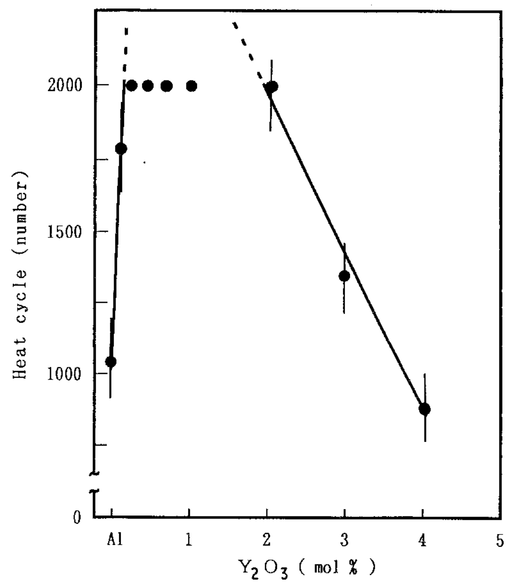


Figure 8 The heat-cycle number of the plasma-coated test pieces as a function of composition dependence of the Al–Y_2O_3 system. The Al–Y_2O_3 composite powder was initially plasma sprayed on to an aluminium alloy substrate, and then coated with PSZ. (When no peeling occurred during 2000 cycles, the heat-cycle number was expressed as 2000.)

times. During this time, the degree of peeling of the plasma sprayed portion was observed under a microscope. The heat-cycle characteristic was evaluated by the number of cycles at which peeling occurred. When no peeling occurred over 2000 cycles, the heat-cycle number was expressed as 2000. Fig. 8 shows the composition dependence curve of the heat-cycle for Y_2O_3 in Al–Y_2O_3 powder using plasma spraying. For 0.2 to 1 mol % Y_2O_3, the heat-cycle number was much higher, being over 2000. The heat-cycle was compared with METECO461NS, in case peeling occurred between the aluminium alloy and METECO461NS; in this system the cycle number was only 295.

This method is based on a unique idea for composite powder production, without the need for heat energy, and making use of the ductility of the metal and the brittleness of the ceramics. In the present case, aluminium metal and Y_2O_3 ceramic were used. However, this method can be applied to many cases, for example those having different specific gravities, particle sizes and shapes, between the metal and ceramics, and also, in such cases as those with a high melting temperature metal and low temperature metal powder. These composite powders can be used not only for plasma spraying but also for sintering.

4. Conclusion

Based on a unique idea, composite spherical powders of aluminium and Y_2O_3 prepared by the mechanical shock method can be obtained using high-speed rotors. This method requires no heat energy, and thus can make the best use of the ductility of the metal and the brittleness of the ceramics. The preparation time is only 3 min. In the present work, aluminium metal and Y_2O_3 ceramic were used. However, this method can be applied to many cases, for example those having different specific gravities, particle sizes and shapes, between the metal and ceramics. At the boundary region, a diffuse layer is formed at the interface between aluminium and Y_2O_3 and thus, the boundary between these components is thought to be quite strong. These composite spherical powders are excellent regarding fluidity and spray capability. The Al–Y_2O_3 composite powder was initially plasma sprayed on to an aluminium alloy and then coated with PSZ. The tensile strength of these test pieces was studied as a function of compositional changes in the Al–Y_2O_3 system. For 0.2 to 1.0 mol % Y_2O_3 the strength attained is much higher than normal being up to 75.5 MPa. The heat-cycle number of the test pieces with the above composition range at the present time has been measured at over 2000.

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