

Title	Effect of Processing Parameters on Microstructure and Mechanical Properties of Zirconia/Alumina Composite Coatings by Gas Tunnel Type Plasma Spraying
Author(s)	Shanmugavelayutham, Gurusamy; Kobayashi, Akira
Citation	Transactions of JWRI. 34(1) P.43-P.47
Issue Date	2005-07
Text Version	publisher
URL	<a href="http://hdl.handle.net/11094/4162">http://hdl.handle.net/11094/4162</a>
DOI	
rights	

***Osaka University Knowledge Archive : OUKA***

<https://ir.library.osaka-u.ac.jp/repo/ouka/all/>

# Effect of Processing Parameters on Microstructure and Mechanical Properties of Zirconia/Alumina Composite Coatings by Gas Tunnel Type Plasma Spraying

SHANMUGAVELAYUTHAM Gurusamy\* and KOBAYASHI Akira\*\*

## Abstract

*Plasma power and substrate traverse number during deposition play an important role in the plasma spray coating process and affect the final properties of the coatings. Zirconia, alumina, zirconia/alumina composite coatings on stainless substrate were prepared by a gas tunnel type plasma spray system to investigate the effects of these parameters on the coating. The results indicated that those parameters such as alumina mixing ratio, plasma power and traverse number over the substrate did affect the hardness, porosity and wear resistance of composite coatings. The Vickers hardness of the coating becomes large with an increase in alumina mixing ratio, the porosity decreased from 33.7 to 7.65%. The lowest porosity and the highest hardness of composite coatings were obtained at a substrate traverse number,  $N=20$  times with a spraying distance,  $L=40$  mm. The adhesive strength of the composite coating decreased with an increase in alumina mixing ratio and at the same time, wear weight of the coating also decreased.*

**KEY WORDS:** (Plasma spray), (Zirconia/alumina composite coating), (Adhesion strength), (Vickers hardness), (Wear resistance).

## 1. Introduction

Alumina ceramic coatings are widely used as wear-resistant and insulating coatings. Alumina/zirconia plasma sprayed coatings are used in a wide range of industrial applications, primarily for wear resistance, thermal barrier and corrosive environment<sup>1,2</sup>. A thermal spray coating is built up and the microstructure is formed, when individual, fully or partially molten particles, travelling at a particular velocity, flatten and solidify on impact with the substrate<sup>3,4</sup>.

Thermal spraying is a highly complex deposition process with a large number of interrelated variables. Due to the high velocity and temperature gradients in the plume, even small changes in the controllable or uncontrollable parameters can result in significant changes in the particle properties and thus in the microstructure of the coatings. The Atmospheric Plasma Spraying is one of the processes based on the creation of a plasma jet to melt a feedstock powder. The powder particles are injected with the aid of a carrier gas and gain their velocity and temperature by thermal and kinematic transfers from the plasma jet. At the surface of the part to be covered, such particles flatten and solidify rapidly forming a stacking of lamellas. The coating microstructure is characterized by a heterogeneous phase configuration with a porosity content due to the voids left by the stacking process.

Gas tunnel type plasma spraying, developed by the author<sup>5,6</sup> for production of new thermal barrier coatings, has superior properties as compared with the conventional type plasma spray method. High hardness ceramic coatings were obtained which were investigated under development with engineered structures. Multilayered graded surface treatments that offer optimized performance and novel coating processing technologies as being used to manufacture numerous coating systems that embody smart concepts.

In order to achieve consistent high quality coatings to meet the more demanding performance requirements of today's applications, there is a need to put more effort into improved control of the plasma spray process. The control of the ceramic in-service properties and especially wear behaviour is sensitive to the large number of the processing parameters and their interdependencies. Such control is obviously complex to establish and most models consider a smaller number of control factors having direct correlations with the processing parameters. These control factors are the velocity and temperature of the feedstock powder particles during their flight, i.e., the in-flight particle characteristics. If the studied in-service property is not sensitive, in a first approximation, to the physical properties of the coated part, control based on the in-flight particle characteristics is then efficient. In such a way, considering the effect of the energetic and injection parameters on the wear behaviour of plasma sprayed coatings becomes a very important factor.

In this study coatings of zirconia/alumina were deposited on a stainless steel substrate. The influence of the coating parameters such as hardness, porosity and wear resistance was investigated at different input powers and substrate traverse times.

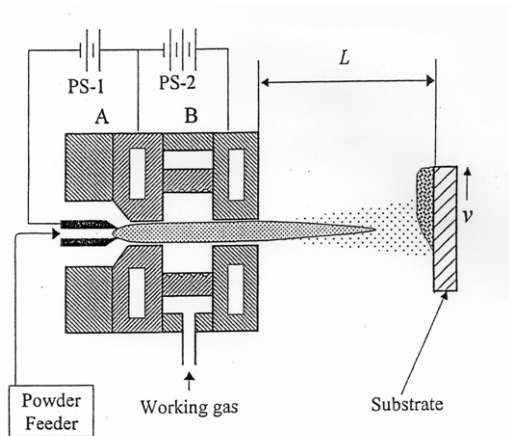
## 2. Experimental Details

Commercially available alumina-zirconia powders were thermally sprayed onto a stainless steel substrate using a gas tunnel type plasma spray system under atmospheric plasma spraying conditions, shown in Fig.1. The plasma torch was operated at power levels upto 21kW and was chosen to carry out all the experiments. The plasma jet was generated with the aid of argon gas fixed at 180 lpm. The torch was maintained at a spray distance of 40 mm from the substrate plane. The powder

† Received on July 1st, 2005

\* Foreign Invited Researcher

\*\* Associate Professor



**Fig.1** Schematic diagram of the gas tunnel type plasma spraying torch system.

injection was external to the torch and directed parallel to the plasma flow and parallel to the torch trajectory. The maximum feed rate was 45 g/min. The spray configuration was a combination of a rotating sample holder and fixed torch uniformly. The defined substrate traverse times are 4, 10 and 20 times. To avoid adhesion problems due to different thermal expansion coefficients between the coating and the substrate and to limit the stress level, nitrogen gas was added to the spray configuration to lower the coating temperature during deposition.

Experiments were carried out considering four processing parameters: the arc current ( $I$ ), gas flow rate, the powder carrier gas flow rate and the injection distance. The first two parameters are known to influence significantly the plasma jet properties (enthalpy, velocity, etc..) and the last one to influence the particle trajectories in the plasma jet.

The grit blasted substrates were used for plasma spraying of 100%  $ZrO_2$ , 80%  $ZrO_2$ -20%  $Al_2O_3$ , 50%  $ZrO_2$ -50%  $Al_2O_3$ , 20%  $ZrO_2$ -80%  $Al_2O_3$ , 100%  $Al_2O_3$ . Operating conditions are given in Table 1. The particle size and chemical composition of powders are given in Table 2.

Microscopic observation of the coatings was performed using an optical microscope and porosity was measured by Image analysis software. Each specimen was mounted in a conductive resin, ground with SiC paper and finally polished with 0.05 micron alumina slurry. The investigations of the amount of porosity and thickness of the sprayed coatings were observed by optical microscope. Average thickness and porosity of the coatings were derived from 5 measurements per sample using an optical microscope.

**Table 1** Experimental conditions.

Powder:	$ZrO_2 + Al_2O_3$ Mixture
Traverse number, $N$ :	4, 10 & 20
Power input, $P$ (kW):	17-21
Working gas flow rate, $Q$ (l/min):	180
Powder feed gas, $Q_{feed}$ (l/min):	10
Spraying distance, $L$ (mm):	40
Traverse speed, $v$ (cm/min):	50 & 100
Powder feed rate, $w$ (g/min):	25-45
Gas divertor nozzle dia., $d$ (mm):	20

**Table 2** Chemical compositions and sizes of zirconia and alumina powders.

Composition (wt%)						Size( $\mu m$ )
$ZrO_2$	$ZrO_2$	$Y_2O_3$	$Al_2O_3$	$SiO_2$	$Fe_2O_3$	10-45
	91.65	7.72	0.02	0.01	0.14	
$Al_2O_3$	$Al_2O_3$	$Na_2O$	$SiO_2$	$Fe_2O_3$		10-45
	99.8	0.14	0.01	0.01		

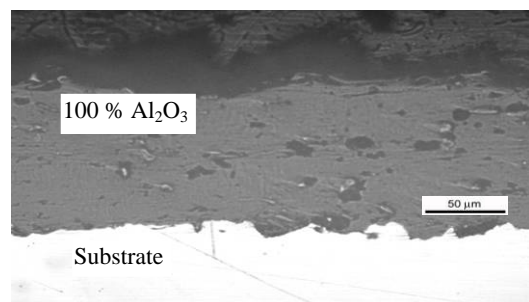
Vickers microhardness was measured on polished sample surfaces using a load of 50g for each material. Indentation parameters were set as 20s loading time and average thickness was derived from five measurements. Wear resistance was calculated by using an abrasive wear resistance method with a 10X10 mm sample holder and 400 mm SiC paper. Relationships between strength and wear resistance were also investigated.

### 3. Results and Discussions

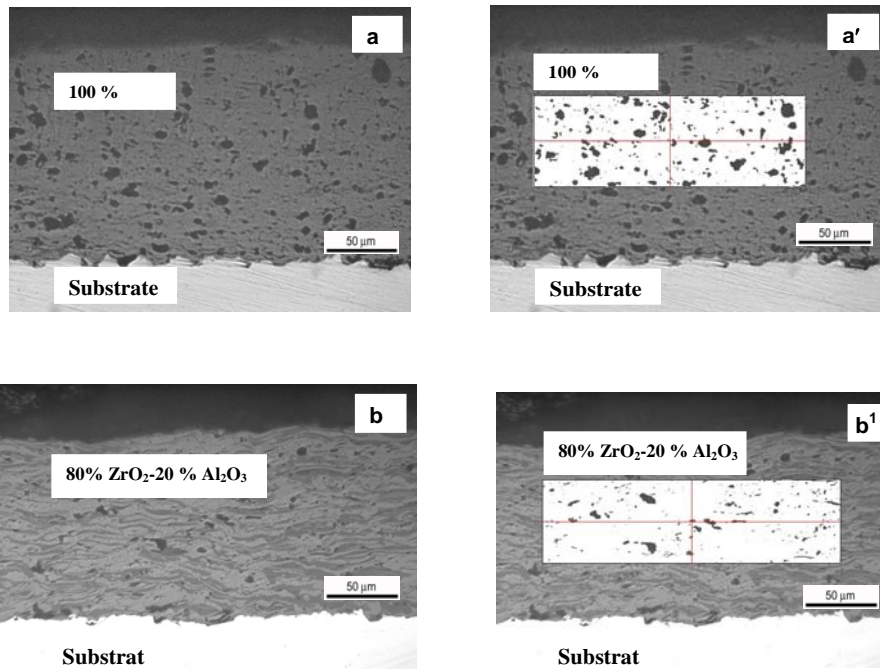
#### 3.1. Microstructure of the coatings

**Figure 2** shows a typical microstructure of the plasma sprayed  $Al_2O_3$  coating prepared under these conditions (substrate roughness, spray distance of 40mm, substrate temperature and coating thickness of 180micron) by the plasma spray system. The micrograph shows a dense coating with homogeneously dispersed porosity. No macrocracking was evident. Pores indicated in the alumina coating are represented by the black areas in the microstructure. It has been reported that pore content is connected with the surface roughness of the substrate, spray distance, substrate temperature and coating thickness<sup>7</sup>.

**Figure 3a and Figure 3b** shows the optical cross sectional micrographs of the pure zirconia and composite coatings. It can be seen that the microstructure was apparently related to the investigated spraying parameters. The thickness of all coatings and the distribution of porosity along the cross section were reported. The coatings present a porous and lamellar structure which is characteristic for this kind of coatings<sup>8</sup>). The splats are separated by interlamellar pores resulting from rapid solidification of the lamellae, very fine voids formed by incomplete intersplat contact or around unmelted particles, and cracks due to thermal stresses and tensile quenching relaxation stresses. The presence of cracks also increases the strain tolerance and enhances the thermal shock resistance for TBCs in service.

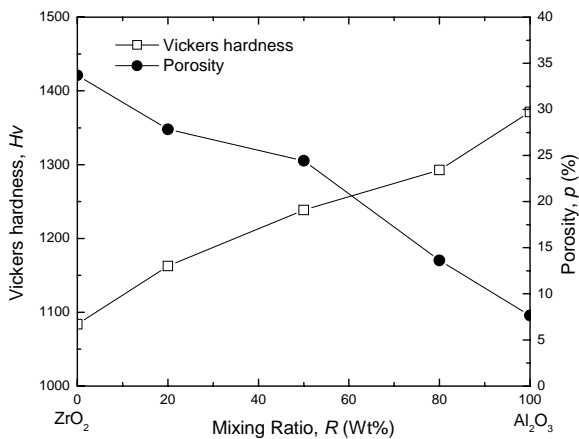


**Fig.2** Optical micrographs of microstructure and porosity on the cross section of 100%alumina coating produced by gas tunnel type plasma spraying.



**Fig.3(a), (b)** Optical micrographs of microstructure and porosity on the cross section of zirconia/alumina composite coatings produced by gas tunnel type plasma spraying. a and b are 100 % ZrO<sub>2</sub> and 80% ZrO<sub>2</sub>-20% Al<sub>2</sub>O<sub>3</sub> composite coatings respectively and a', b' are plasma coatings after image processing.

From the optical micrographs it is possible to observe the variation of the porosity for different coatings and also its variation from the interface with bond to the surface demonstrated in Fig. 3 (b). The quantification of this variation along a cross section has been measured by image analysis is shown in Fig.3 (a')&(b'). Additionally, small microcracks are observed. These cracks originate from the thermal stresses which arise from the rapid cooling during the spray process. Small microcracks with diameters of about 200 nm have been reported<sup>9)</sup> and each coatings show an excellent thermal shock resistance. In our opinion, the gradation in porosity improves this resistance because the coatings have better accommodation of thermal stresses during the quenching period

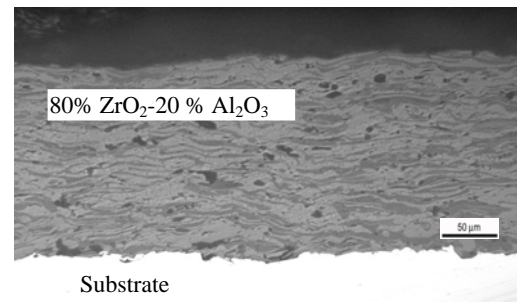


**Fig.4** Dependence of hardness and porosity of zirconia composite coatings on the alumina mixing ratio.

### 3.2. Effect of alumina mixing ratio on the coatings

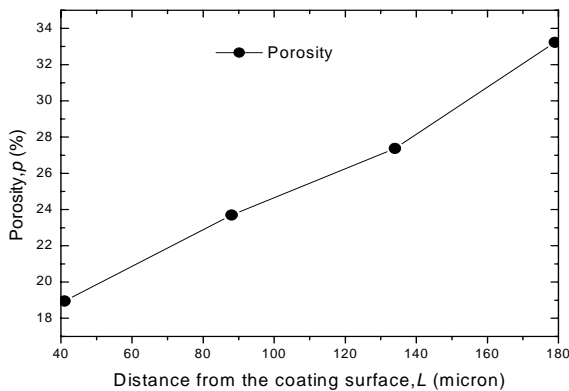
**Figure 4** shows the relationship between Vickers hardness and porosity of zirconia composite coatings formed by gas tunnel type plasma spraying with the alumina mixing ratio  $R$  (wt%). In this case, the power input was  $P=21\text{kW}$  and the spraying distance was  $L=40\text{mm}$ , with the traverse number of 4. The coating thickness approximately varied from 140 to 250 $\mu\text{m}$ .

The Vickers hardness on the cross section of zirconia composite coating increased with an increase in the alumina mixing ratio. The coating hardness corresponds to the high hardness of alumina particles. So that, the Vickers hardness of the alumina coating was  $Hv_{50}=1371$ . The hardness distribution of the composite coating has a remarkable graded functionality in the case of a large alumina mixing ratio. Because the part near the substrate did not change so much, the Vickers hardness near the coating surface became much higher due to the lower percentage of porosity. The effect of mixing gives higher thermal resistance in the transverse direction when the heat flux goes perpendicular to the coating surface.



**Fig.5** Optical micrographs of 80% ZrO<sub>2</sub>-20% Al<sub>2</sub>O<sub>3</sub> composite coatings produced by gas tunnel type plasma spraying.

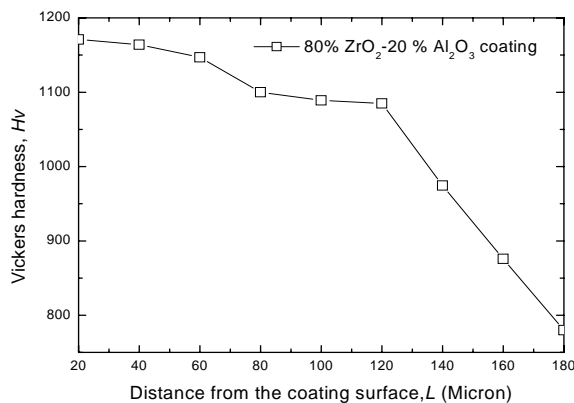
This leads to the development of thick and highly functional thermal barrier coatings. **Figure 5** shows the cross section micrograph of 80%ZrO<sub>2</sub>-20%Al<sub>2</sub>O<sub>3</sub> composite coatings sprayed at L=40mm. Porosity increase from the surface of the coatings towards the surface of the substrate is shown in **Fig. 6**. In the as-sprayed condition the absolute porosity variation ranges from 18.95% to 33.23%, from the surface of the coatings towards the surface of the substrate. This variation contributes to the low and high hardness of the coatings.



**Fig.6** Porosity distribution along the cross section at different distances from the coating surface of composite coatings (80% ZrO<sub>2</sub>-20% Al<sub>2</sub>O<sub>3</sub>).

### 3.3. Microhardness measurements

**Figure 7** shows the microhardness distribution of the ceramic top coatings for the as-sprayed condition. It can be observed that the microhardness increases towards the surface in composite coatings. Portinha et.al<sup>9)</sup> observed little decrease in the porosity values towards the surface which might be explained by the increase of the surface temperature during deposition, and justifies the small microhardness increase in the sample. Not only can the reduction in the porosity contribute to an increase of the hardness but also the thermal residual stresses within the coatings are important. The successive materials arriving at the substrate solidify on a surface at lower temperature that increases with coating growth and slightly decreases the cooling velocity of the splats.



**Fig.7** Vickers hardness measurements in plasma sprayed coatings (80%ZrO<sub>2</sub>-20% Al<sub>2</sub>O<sub>3</sub>) along the cross section at different distances from the coating surface.

With this effect, the layers closer to the final coating top surface should present a more dense structure that has higher elastic properties, and is therefore favorable to the formation of higher residual stresses and also present more hardness.

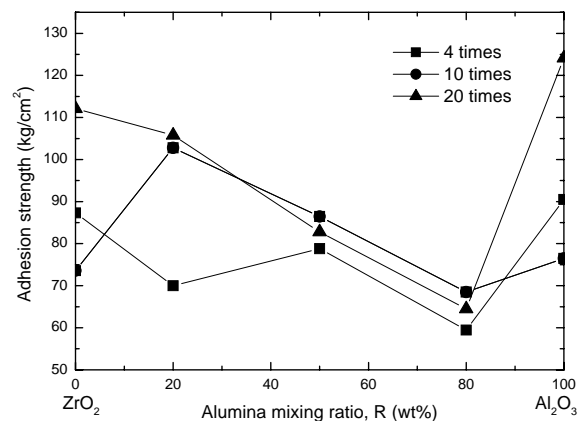
The decrease in microhardness for the graded samples is due to the increase in porosity along the cross section, this variation can be observed in Fig. 7 and it is clear as a reduction in the hardness values for the higher porosities. All the reported microhardness values are means of 10 indentations

### 3.4. Effect of adhesion strength on composite coating

Regarding the adhesive strength of zirconia composite coatings, which decreased with an increase in the alumina-mixing ratio, the adherence of zirconia composite coating was minimum when the ratio was R=50-80% and decreased from 105.78 to 64.5 kg/cm<sup>2</sup> which is shown in **Fig.8**. These composite coatings were formed by three different substrate traverse numbers i.e 4, 10 and 20 times. It is noted that the adherence improved when the traverse number was large. The alumina coating had a high bonding strength, and the fracture was at the interface between coating and substrate. The thicker topcoat is mechanically weakened with increasing pores and residual stresses. The higher hardness, higher adhesion strength and lower porosity can be obtained at a lower coating thickness of 140µm. The increase of the porosity amount will result in the decrease of the hardness of the coating<sup>10-15</sup>

### 3.5. Effect of wear resistance on composite coatings

**Figure 9** shows the relationship between adhesion strength and wear resistance of composite coatings with the alumina mixing ratio R (wt%). In this case, the power input was P=21kW and the spraying distance was L=40mm, with a traverse number of 20 times. The coating thickness approximately varied from 140 to 250µm. The adhesion strength of the zirconia composite coating decreased with an increase in the alumina mixing ratio and depends on the high hardness of alumina particles. Hence the higher strength and lower wear resistance of alumina coating is observed. At the same time the wear resistance also decreased from 8.38 to 1.49 mg/cm<sup>2</sup>.h. This behaviour indicates an improvement of high functionality thermal barrier coatings.



**Fig.8** Variation of adhesion strength for different alumina mixing ratios at three different substrate traverse times.

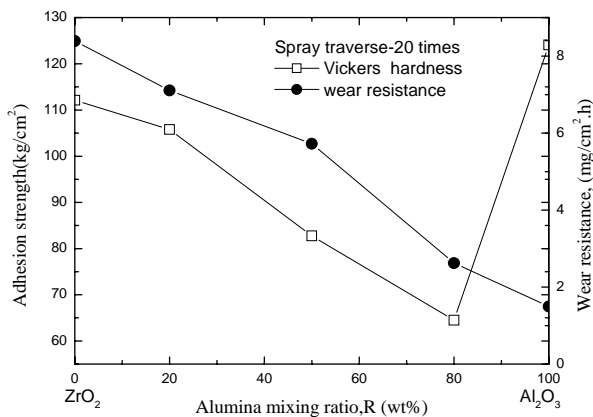


Fig.9 Variation of strength and wear resistance at different alumina mixing ratios.

#### 4. Conclusions

The following results were investigated in this study:

- (1) With an increase in the alumina mixing ratio, the Vickers hardness of the high hardness layer became large, and changed from Hv=1083 to 1371, at the same time the porosity in the hardness layer decreased and changed from 33.7 to 7.65 %.
- (2) In the case of the same spraying distance, the value of the adhesive strength was highest at 112.2 kgf/cm<sup>2</sup> with a coating thickness of around 140μm. The adhesive strength became weaker as the coating thickness became greater. In this study, the thickness value reached approximately 250μm.
- (3) With an increase in the alumina mixing ratio, the adhesion strength of the zirconia composite coating decreased because of the high hardness of alumina particles. At the same time the wear resistance decreased from 8.38 to 1.49 mg/cm<sup>2</sup>.h.
- (4) When the traverse number increased, the adherence of the coating became higher, but the wear resistance of the sprayed coating became weaker. The properties of sprayed zirconia composite coatings would be better than at fewer traverse numbers when the other operating parameters were held the same.

#### Acknowledgement

This work was financially supported by a Grant-in-Aid for Scientific Research from the Japanese Ministry of Education, Science and Culture and JFE 21<sup>st</sup> Century Foundation. The authors are grateful to Mr. Yano for assistance in preparing the coating samples during experiment.

#### References

- 1) E. Celik, I.A. Sengil and E. Avci. *Surf. Coat. Technol.* **97** (1997) pp. 355–360.
- 2) F. Üstel, S. Soykan, E. Çelik and E. Avci. *J. Metall.* **97** (1995) pp. 31–37.
- 3) M. Friis, C. Persson and J. Wigren. *Surf. Coat. Technol.* **141** (2001) pp. 115–127.
- 4) J. Matejcek and S. Sampath. *Acta Mater.* **49** (2001), pp. 1993–1999.
- 5) Y. Arata, A. Kobayashi and Y. Habara, *J. Appl.Phys.* **62** (1987) pp. 4884–4889.
- 6) Y. Arata, A. Kobayashi, Y. Habara and S. Jing, *Trans.of JWRI*, 15-2 (1986) pp.227-231.
- 7) H.M. Choi, B.S. Kang, W.K. Choi and D.G. Choi. *J. Mater. Sci.* **33** (1998) pp.5895-99.
- 8) C.J. Li, Y. He, A. Ohmori, *Proceedings of the 15th International Thermal Spray Conference*, Nice, France (1998) pp.717.
- 9) A. Portinha, V. Teixeira, J. Carneiro, J. Martins, M.F. Costa, R. Vassen and D. Stoever, Characterization of thermal barrier coatings with a gradient in porosity: *Surf.Coat.Technol.*, 195 (2005) pp.245-251.
- 10) G. Shanmugavelayutham and A. Kobayashi, *Plasma Applications and Hybrid Functionally Materials*, 14 (2005) pp. 79-82.
- 11) A.Kobayashi, S. Kurihara, Y. Habara, and Y. Arata, *J. Weld. Soc. Jpn.*, 8 (1990) pp.457-462.
- 12) A. Kobayashi, Property of an Alumina Coating Sprayed with a Gas Tunnel Plasma Spraying, *Proc. of ITSC.*, (1992) pp.57-62.
- 13) A. Kobayashi, *Surface and Coating Technology*, 90 (1990) pp. 197-202.
- 14) Y. Li and K.A. Khor. *Surf. Coat. Technol.* **150** (2002) pp. 125–132.
- 15) S. Sampath, X. Jiang, A. Kulkarni, J. Matejcek, D.L. Gilmore and R.A. Neiser. *Mater.Sci. Eng. A* **A348** (2003) pp. 54–66.