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Impregnation of Thermally Sprayed Coatings for Microstructural Studies

J. Karthikeyan, A.K. Sinha, and A.R. Biswas

Sprayed coatings are normally impregnated with an epoxy prior to metallographic specimen preparation procedures. The effect of specimen impregnation technique on the structural and microhardness measurements of sprayed coatings has been evaluated. Independent, simultaneous, and controlled evacuation of both the specimen and the resin ensures complete impregnation of pores, yielding metallographic specimens with minimum abrasive loss and minimum scatter in the measured coating properties.

1. Introduction

IN GENERAL, thermal spray coatings have complex, composite structures consisting of a metal substrate, a metallic bond coat, and a ceramic overlay. Moreover, coating properties such as porosity, composition, and stress vary across the specimen cross section. Preparation of specimens for microstructural study is a difficult task, because the mechanical stresses that arise during the mounting, cutting, grinding, and polishing operations can result in material pullout, alter structural features, and damage the specimen (Ref 1-6).

Thermal barrier coatings (TBCs) represent a case study. Thermally sprayed TBCs are widely used in aircraft engines, turbines, and other advanced systems. Because a TBC has a porous and fragile ceramic coating and two or more interfaces with residual stresses, extreme care is required when preparing metallographic specimens in order to avoid altering the microstructure. Several studies (e.g., Ref 6) have concentrated exclusively on this aspect of TBC research.

It has been well established that the first step in metallographic specimen preparation of sprayed coatings is to impregnate the sample in vacuum with an epoxy resin (Ref 1-6). Vacuum impregnation results in filling of pores, voids, microcracks, and other surface defects in the specimen with resin. As a result of the additional strength contributed by the impregnated epoxy, the specimen can withstand much higher mechanical stresses and can be cut, ground, and polished without damage.

A few vacuum impregnation systems have been reported (Ref 1, 2). In these systems, the epoxy resin is kept at or close to ambient pressure and only the specimen is evacuated. After evacuation, the viscous epoxy is made to flow into the specimen chamber by tilting the entire vacuum assembly. Considerable degassing and frothing occur in the resin as soon as it flows into the vacuum chamber, and numerous bubbles form in the epoxy. The formation and collapse of bubbles on the specimen surface likely results in cavitation damage of the specimen.

Keywords hardness measurements, metallography, microstructural studies, yttria-stabilized zirconia

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Some researchers practice the "through epoxy evacuation" to avoid these difficulties. In this technique, the specimen is immersed in the epoxy at ambient pressure and is then evacuated through the resin. Since the evacuation of the pores in the specimen is carried out through the highly viscous resin, the evacuation (and hence the impregnation) will be largely ineffective. A simple vacuum impregnation system that has been built in the authors' laboratory allows independent, simultaneous, and controlled evacuation of the specimen, immersion of the specimen into evacuated resin without breaking the vacuum, and impregnation of the specimen with the epoxy by opening the air vent. The effect of specimen impregnation technique on the structural and microhardness measurements of sprayed coatings has been studied. This article presents the results.

2. Experimental Method

2.1 *Preparation of Test Specimens*

Nickel-base superalloy (Inconel 600) disks 20 mm in diameter and 3 mm thick were used as substrates. Feedstocks of 95Ni-5Al and 8 wt% yttria-stabilized zirconia powders were used to prepare the bond coat and the ceramic overlay, respectively. An atmospheric plasma spray system (Metco 7MB) was used for the spray operations. Duplex TBCs with a 0.13 mm thick bond coat and a 0.38 mm thick ceramic overlay were plasma sprayed onto grit-blasted substrates and used as test specimens.

2.2 *Vacuum Impregnation System*

Figure 1 is a schematic of the vacuum impregnation system. It consists of a vacuum chamber (150 mm diam and 80 mm high) with a transparent cover and a small rotary vacuum pump along with the associated pipelines, a thermocouple gage, a vacuum valve, and an air vent. For vacuum impregnation of the specimen, the impregnating epoxy (premixed Araldite resin CY 230 and hardener HY 951 in a 4-to-1 weight ratio) is kept in a beaker inside the vacuum chamber. The specimen is placed on a stand inside the chamber just above the beaker, as shown in Fig. 1, and the chamber is evacuated slowly.

During the initial stages of evacuation, considerable degassing and frothing occur in the epoxy. Hence, the epoxy is continuously monitored via the transparent cover, and the pumping speed is slowly increased in a controlled manner by using the vacuum valve until the chamber vacuum reaches about 10^{-3} torr. The chamber vacuum is maintained at 10^{-3} torr for another 30 min to ensure the complete evacuation of all the pores in the specimen. Then, the specimen is pushed into the resin using the handle (specimen feeder). When the specimen is completely submerged in the resin, the vacuum valve is closed and the air vent opened to allow air into the chamber. Due to atmospheric pressure, the resin penetrates into the pores and cracks in the specimen. After 5 min, the beaker containing the resin and specimen is removed and the specimen may be adjusted into a more appropriate orientation in the same resin.

2.3 *Specimen Impregnation Methods*

Sets of three TBC specimens were molded in the epoxy resin as follows. The first specimen was molded without vacuum impregnation; this specimen is designated as the as-received specimen. The second specimen was impregnated using the through epoxy evacuation technique, described earlier. The third specimen was vacuum impregnated by the procedure described in section 2.2, designated as the "independent evacuation" technique. Figure 2 shows the three different methods; the primary differences among them are summarized in Table 1.

2.4 *Specimen Testing*

All three sets of specimens were simultaneously prepared for microstructural investigations following the procedure outlined in Table 2. Microstructural investigations were carried out using a standard inverted-type optical microscope (Richert, Austria). The microhardness measurement and data analysis procedures are detailed in Ref 7. In brief, a standard microhardness microscope (Wilson Instruments, UK) was used with a load of 300 g over a loading cycle time of 17 s. Microhardness of the specimen was measured at precise locations in three different regions of the coating. At each location, a series of 30 hardness tests were carried out.

The procedure was as follows. The specimen was mounted on the microhardness table and oriented so that the x traverse was parallel to the substrate edge. The "average interface"—that is, the boundary between the bond coat and the substrate—was termed the I_{SB} (interface between substrate and bond coat), and a series of microhardness measurements were carried out at that location. Similarly, a series of measurements were performed at the average interface between the bond coat and the ceramic layer (designated as the I_{BC}), and a final series of measurements were carried out at the midplane of the ceramic coating (designated as C_{MP}).

Table 1 Evacuation-impregnation techniques

Method	Designation	Evacuation: Specimen	Evacuation: Resin
A	As-received	No	No
B	Through epoxy evacuation	Partial	Yes
C	Independent evacuation	Yes	Yes

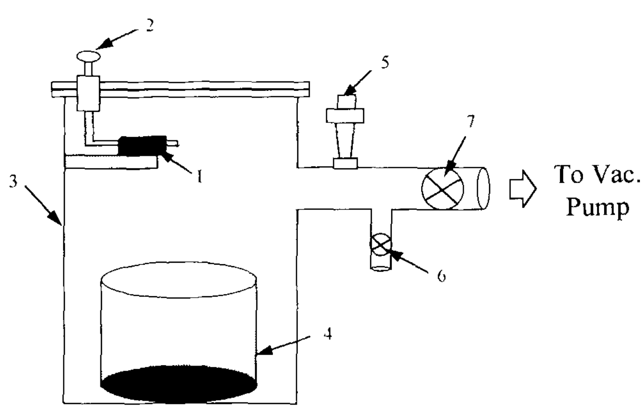


Fig. 1 Schematic of the vacuum impregnation system 1, sprayed coating; 2, handle; 3, vacuum chamber; 4, resin; 5, vacuum gage; 6, air vent

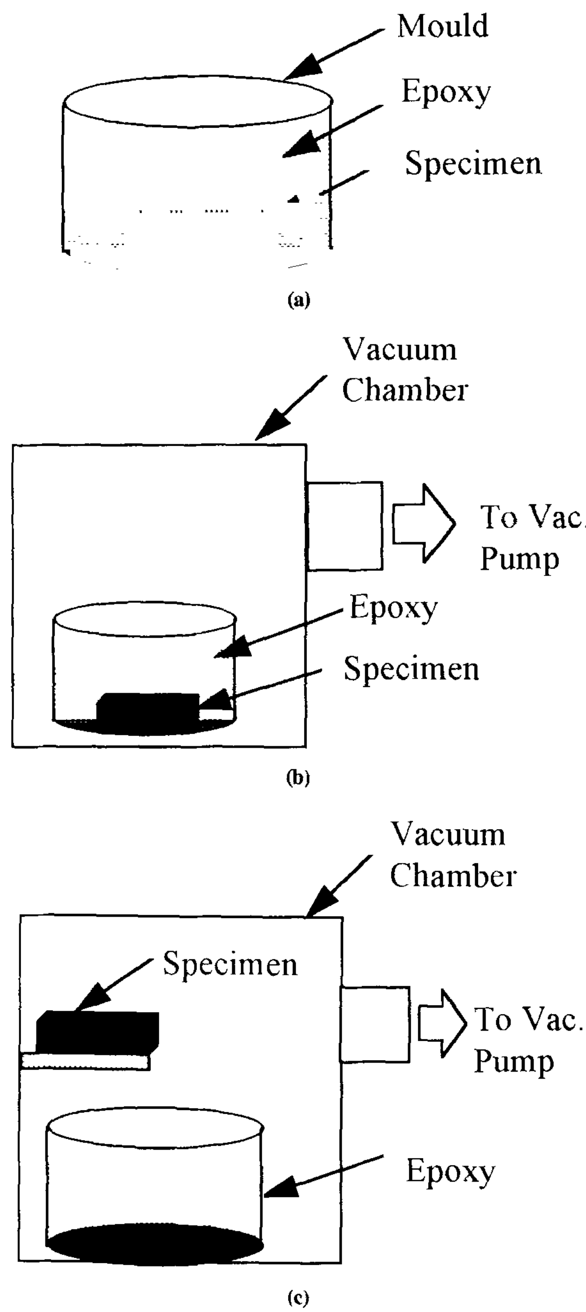


Fig. 2 Evacuation techniques (a) As received (b) Through epoxy evacuation (c) Independent evacuation

During microhardness measurement, each indent location was carefully selected. After indentation, each indent was thoroughly examined to ensure that it did not overlap voids or pores. The mean, standard deviation and the variance of each series were calculated by assuming that the data obeyed the normal (Gaussian) distribution. Weibull analysis of each series was performed to study the spread or scatter in the measured values (Ref 7-10).

3. Results and Discussion

3.1 *Microstructure*

Figure 3 shows the microstructures of typical specimens molded using the three procedures. Inherent porosity in all the specimens is clearly visible as regularly shaped (circular and lamella) black spots and lines. From the microstructures, it can be inferred that the specimens contain both closed pores (spherical shape, about 5.0 µm diam) and interconnected pores (lamella shape, about 5.0 µm diam and 3 to 20 µm long). Abrasive loss that occurs during specimen preparation manifests itself as large voids and is visible as large black spots, cavities, and canals of irregular shapes and sizes. As can be seen in Fig. 3(c), a small amount of abrasive loss occurred in the specimen that was impregnated after independent evacuation of specimen and epoxy. The material loss is particularly visible in the ceramic coating region of the specimen. No abrasive loss occurred at the two interfaces or at the bond coat region. By comparison, in the specimen molded by the through epoxy evacuation method (Fig. 3b), both the number and size of the voids in the ceramic region have increased. The bond coating also shows a small amount of material pullout, particularly at the interface between the substrate and the bond coat. Maximum abrasive loss has occurred in the as-received specimen (Fig. 3a), which has led to the formation of large canals and cavities in the ceramic coating. Moreover, a large number of small voids have formed at both interfaces and at the bond coat region.

To summarize the microstructural observations, the microstructures of as-received and vacuum-impregnated specimens vary greatly. There are also perceptible differences between the microstructures of through epoxy and independent evacuated specimens. Material pullout and specimen damage are minimal in the case of the direct evacuated specimen.

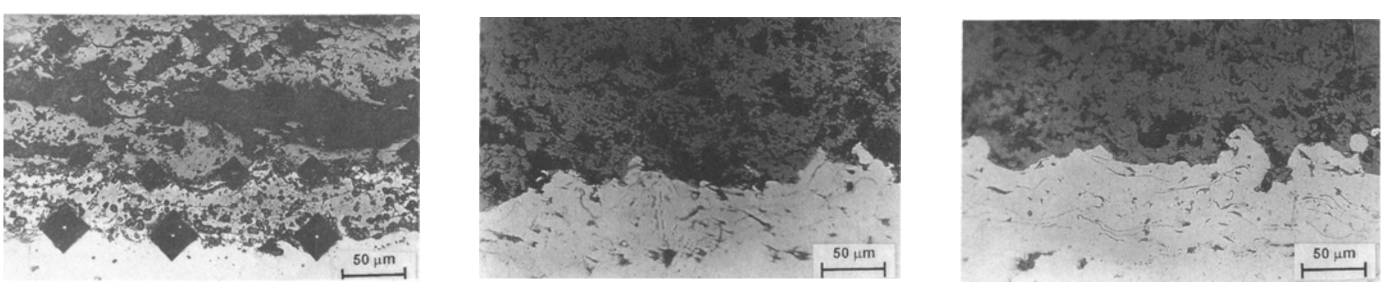
Table 2 Specimen preparation procedure

Operation	Surface	Abrasive size	Lubricant	Speed, rev/min	Time, min
Grinding	SiC paper	200 mesh	Water	120	5.0
	SiC paper	400 mesh	Water	120	2.0
	SiC paper	600 mesh	Water	120	2.0
Polishing	Napless cloth	6 µm diamond	Lapping oil	120	6.0
	Napless cloth	3 µm diamond	Lapping oil	120	2.0
	Napless cloth	1 µm diamond	Lapping oil	120	2.0

Table 3 Results of microhardness studies

Impregnation technique(a):	Substrate-bond coat interface			Bond-ceramic coat interface			Ceramic coating midplane		
	A	B	C	A	B	C	A	B	C
Mean, VHN	258	296	361	226	281	280	291	615	621
Std dev, VHN	39	38	38	41	41	36	73	102	86
Variance, %	15	13	10	18	14	13	25	17	14
Weibull modulus	6.8	8.4	10.3	5.7	7.4	8.4	4.5	6.5	7.6
R square	0.97	0.96	0.96	0.97	0.95	0.93	0.82	0.92	0.97

(a) See Table 2 for details of the impregnation techniques.



(a) (b) (c)

Fig. 3 Typical microstructures of test specimens (a) As received. (b) Vacuum impregnated after through epoxy evacuation. (c) Vacuum impregnated after independent evacuation

3.2 *Microhardness*

Weibull plots of specimens molded using different techniques are compared in Fig. 4. Results of the microhardness studies are summarized in Table 3. As shown, the Weibull moduli values are all very low, regardless of test location or specimen molding technique. Such low Weibull moduli values are not surprising, since all the thermally sprayed coatings show substantial variabilities in most of their measured properties, including strength (Ref 8), microhardness (Ref 7, 9, 10), and others (Ref 11, 12). Figure 5 compares the microhardnesses of specimens molded by different impregnation techniques. It shows that specimen molding technique affects both the absolute magnitude and the statistical scatter in the measured microhardness value and that the independent evacuation technique (method C) produces the minimum scatter in the measured values at all three test locations.

The hardness values of air-impregnated specimens are very low at all test locations, establishing that vacuum impregnation is highly recommended for any meaningful study of sprayed coatings. The microhardnesses of air-impregnated specimens at I_{SB}, I_{BC}, and C_{MP} are 72, 80, and 47%, respectively, of the specimen molded using method C, showing that maximum material pullout has occurred at the ceramic coating, followed by the two interfaces, I_{SB} and I_{BC}. Since ceramic coatings have large residual stresses and are fragile and porous, maximum material pullout would be expected in that region. Grit blasting of the substrate surface, followed by quenching of the molten metallic droplets, results in stress concentration at the I_{SB} and leads to more material pullout at this location. At the interface between the bond coat and the ceramic coating, the porous bond coating serves as a shock absorber; hence, the stress is less at this location as compared to the interface between the substrate and the bond coat. This explains the lower material pullout at this location.

Comparison of the microhardnesses of the two vacuum-impregnated specimens leads to two observations. First, scatter in the measured data points, given by the variance in Gaussian analysis and the Weibull moduli, varies depending on the impregnation technique and the test location. The independent evacuation technique produces the minimum scatter at all test locations. Second, the evacuation technique has no effect on the mean value of the microhardness measured at the midplane of the ceramic coating and at the bond coat/ceramic coating interface. However, an approximate 20% increase in microhardness value is observed at the substrate/bond coat interface when the evacuation procedure is changed from through epoxy (method B) to direct evacuation (method C).

It is believed that the epoxy can penetrate and fill the bulk of the ceramic coating during the vacuum impregnation procedure, since the ceramic coating contains a large number of interconnected pores through which the epoxy can penetrate. However, the metallic bond coat generally has a higher density and fewer interconnected pores. The evacuation of these pores thus requires much higher pumping speeds. In the through epoxy

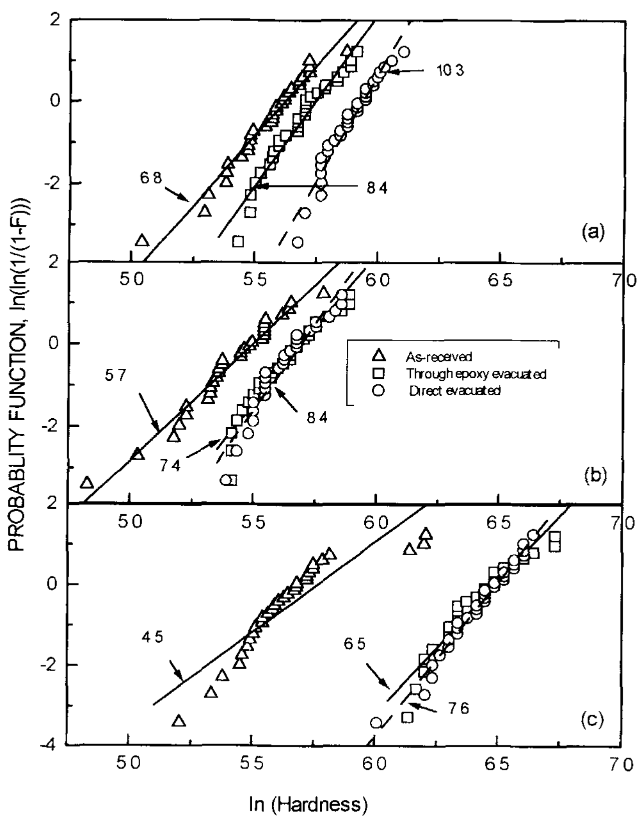


Fig. 4 Weibull plots of the microhardness data measured at interface between the substrate and bond coat (a), interface between the bond coat and ceramic coating (b), and midplane of the ceramic coating (c)

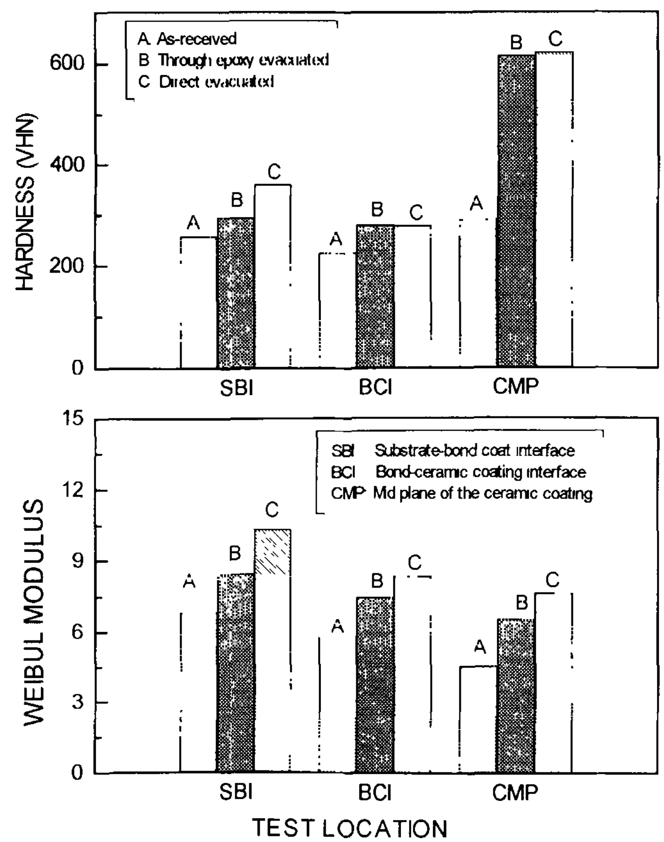


Fig. 5 Variation of microhardness with impregnation techniques

evacuation procedure, the epoxy may penetrate into the ceramic coating but reach only up to the interface between the ceramic coating and the bond coat, resulting in impregnation of the ceramic coating but not of the bond coat. In the independent evacuation technique, however, because of the direct evacuation of the specimen, all the pores in the bond coat will also be evacuated, allowing epoxy to impregnate both the ceramic and the bond coatings, leading to a higher microhardness value at the substrate/bond coat interface.

As noted earlier, during microhardness measurement each indent location was carefully selected and each indent thoroughly examined to ensure that it did not overlap any voids or pores (see Fig. 3). This procedure resulted in similar microhardness values at I_{BC} and C_{MP} for both impregnated specimens regardless of evacuation procedure. However, the presence of larger pore and void fractions in the through epoxy evacuated specimen reduces the number of such indent locations and thus increases data scatter.

3.3 *Advanced Thermal Spray Coatings*

This study has shown that a higher density bond coat can be effectively evacuated and impregnated only by the direct evacuation technique. Advanced thermal spray processes such as high-velocity oxyfuel, D-gun, low-pressure plasma spraying (LPPS), and high-energy plasma produce coatings that generally exhibit little porosity. Hence, the through epoxy evacuation technique will not be very effective with these types of coatings. Similarly, engineered coatings such as graded coatings have porosity, hardness, composition, and other properties that vary across the specimen cross section. Hence, if these specimens are not properly evacuated, then, depending on pumping speed, time, and epoxy properties, they will be impregnated only up to a certain depth, resulting in misleading microstructures. Variation in the amount of abrasive losses across the specimen thickness will also result in false variations in microhardness and other property measurements. Therefore, it is recommended that high-density coatings and other engineered coatings be vacuum impregnated in a system similar to the one reported here.

4. Concluding Remarks

The first step in metallographic specimen preparation of sprayed coatings is to impregnate them in vacuum with an epoxy resin. A simple system that allows direct, simultaneous, and controlled evacuation of both the resin and the specimen has been devised. Structural and microhardness studies carried out on plasma-sprayed TBCs show that the properties of specimens prepared using this system are superior to those of specimens prepared by conventional metallographic techniques.

Acknowledgment

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