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PULSED LASER DEPOSITION OF SILICON CARBIDE ON HEAT RESISTANT MATERIALS

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ABSTRACT

Pulsed laser deposited coatings can be used to enhance the corrosion resistant of materials traditionally used for industrial applications. In this paper, we describe our initial results on coating HK40 (a material used for ethylene heater tubing) with silicon carbide (a carburization resistant coating) to increase tube life. A 1 um thick film of silicon carbide was successfully deposited onto a heated HK40 substrate. An array of characterization techniques (scanning electron microscopy, atomic force microscopy, and scratch tests) demonstrated that the processing conditions were suitable for good coverage and promising adhesion behavior.

Keywords: PLD, SiC, HK40, Adhesion, Sigma Phase

1. INTRODUCTION

Iron based alloys such as HK40 have been used for ethylene heater tubing with operating temperatures near 1000 C [1,2]. In service carburization has reduced tube life from 100,000 to ~ 50,000 hours [3]. Nickel based materials have been suggested for higher operating temperatures with only marginal improvement in carburization resistance [1,2]. In this paper, we report on our initial efforts to utilize pulsed laser deposited (PLD) coatings to reduce carburization and increase tube life. A thin (~ 1 um) silicon carbide (SiC) layer, a candidate material for this application, was deposited on as machined and shot peened HK40 substrates. Hou et al. [4] deposited SiC on Ni-Cr surfaces and found that adhesion improved when composition graded intermediate layers were employed. Surface preparation, which is an essential variable for good adhesion, was not reported in this work. Pulsed laser deposition has been previously used to deposit SiC on smooth semi-conductive surfaces [5-7]. To our knowledge, this paper represents the first study of the effect of surface preparation on the adhesion behavior of PLD films.

2. EXPERIMENTAL PROCEDURE

Centrifugally Cast A609 Grade HK40 [19.0-22.0% Ni, 23.0 –27.0%Cr, 0.35-0.45% C, 0.05 – 2.0% Si, 1.5% max. Mn] was obtained from Ultra-cast Inc. The target material, SiC [0.009% Al, 0.020% B, 0.031% Fe, 0.015% O and 0.017% Mn, traces of Ni and Ti], was supplied by Kurt J. Lesker Company. A KrF Excimer Laser (λ = 248 nm) was used to ablate and deposit SiC on as machined and shot peened (170/ 325 mesh glass beads) HK40 substrates. The films were deposited inside a stainless steel vacuum system pumped to a base pressure in the range of 10^{-4} to 10^{-5} torr. The KrF laser was focused using mirrors to a rectangular spot 0.125 cm x 0.078 cm onto the rotating (10 - 20 rpms) 2.54 cm in diameter SiC target. The pulse energy and frequency of the laser was ~500 mJ and 5-10 hertz respectively. The laser/ target angle of incidence was 45° and the target to substrate distance was 4 - 5 cm. Depositions were performed at room temperature and while resistively heating the substrate up to 500 C. These temperatures were selected to limit sigma phase formation [7] during the deposition process. Figure 1 shows a schematic of the experimental setup.

3. RESULTS AND DISCUSSION

3.1 Atomic Force Microscopy

Contact mode Atomic Force Microscopy (AFM) studies were performed on the target, substrate, and coating. All specimens were scanned at 50 um x 50 um x 2 um and no correction were made during analysis. The roughness of all surfaces were comparable and varied from 300 to 500 nm. The surface morphology of the coated and uncoated substrates did not change. Shot peening introduced compressive stress on the HK40 surface and produced a "dimple like" morphology. Machining produces a "scratch like" appearance and surface tensile stresses. The surface morphology of the machined and shot peened surfaces is shown in figure 2.

3.2 Scanning Electron Microscopy

Typical backscatter electron (BSE) microscopy images of as deposited SiC are shown in figure 3. During room temperature deposition on as machined "scratch like" surfaces, poor coverage and adhesion is apparent (figure 3a). Deposition at higher temperatures and on shot peened dimple like surfaces (figure 3b) showed significant coverage and improved adhesion.
Qualitative energy dispersive spectroscopy (EDS) studies of the target and coating shown in figure 3b showed similarities in the target and coating chemistry (see figure 4). A strong Aluminum (residual element from target) peak existed in the spectra for both the target and coated material.

3.3 Adhesion Test

The scratch-test method consists of generating scratches with a Rockwell C type diamond indenter (tip radius 50μm) which was drawn at a constant speed across the coating-substrate system [8]. Other test conditions are shown in table 1.

A critical load (i.e. the smallest load at which a recognizable failure occurs, see figure 5a) was attained at 0.48 ± 0.06 N. Complete delamination (see figure 5b) occurred at higher loading 4.37 $\pm$ 0.4 N. The coating thickness was determined by SEM of the scratch tested specimen. Figure 6 shows that the coating thickness is ~ 1 um.

4. CONCLUSIONS

In this paper, we presented our initial results on ablation and deposition of SiC onto HK40 substrates. It was determined that the surface condition and processing temperature play a role in the adhesion properties of the resulting films. Shot peening tended to improve coverage and adhesion properties. Substrate temperature during processing also improved coverage and adhesion, however, limitation must be imposed to avoid compromising the mechanical properties of the substrate. These efforts will be extended to determine the suitability of the coating/ substrate system for in-service testing conditions.

5. ACKNOWLEDGEMENTS

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6. REFERENCES

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TABLES

Table 1. Scratch Test Conditions

Loading range	0 to 6 N
Loading rate, dL/dt	6 N/min
Scratch Length	4 mm
Scratching speed, dx/dt	4 mm/min

FIGURES

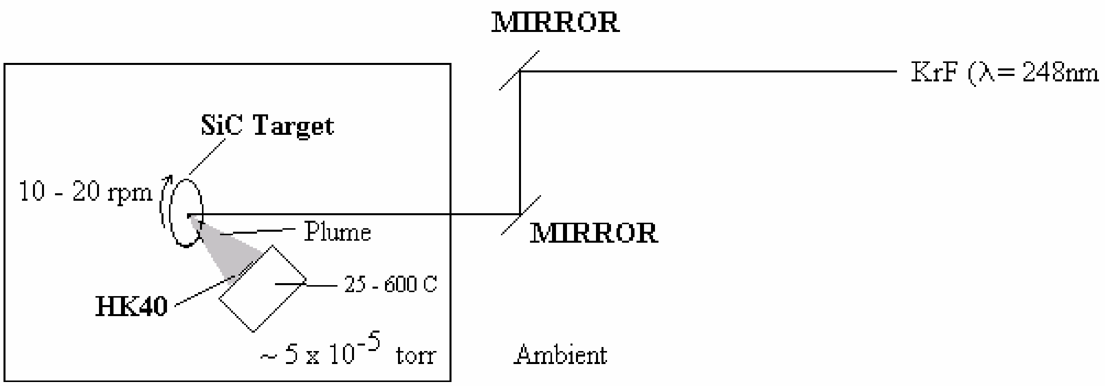


Figure 1. PLD System equipped with rotating target and substrate heating stage.

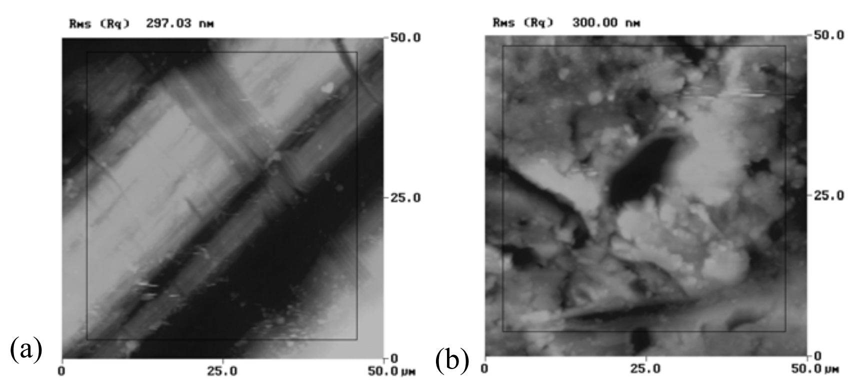
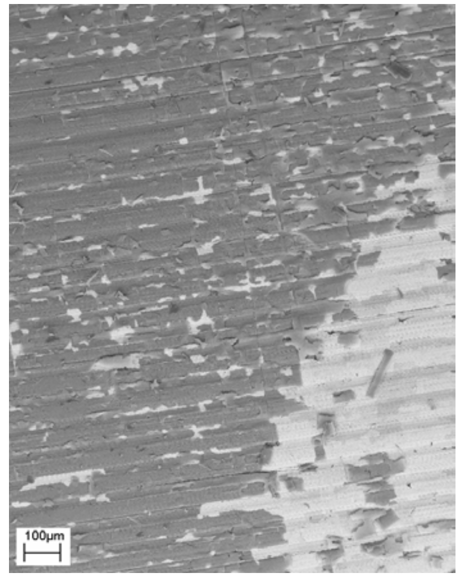


Figure 2. a) Scratch like AFM image of machined HK40 substrate. b) Dimple like image of shot peened HK40 substrate.

(a)



(b)

Figure 3. a) BSE micrograph of PLD of SiC on HK40 at room temperature. High z-contrast areas (white) represent HK-40 (Fe-Ni-Cr) and low z-contrast areas (dark) represent SiC coating. Poor adhesion and coverage are apparent. b) BSE micrograph of PLD of SiC on HK40 at 500 C. Complete coverage of SiC on HK40 is shown.

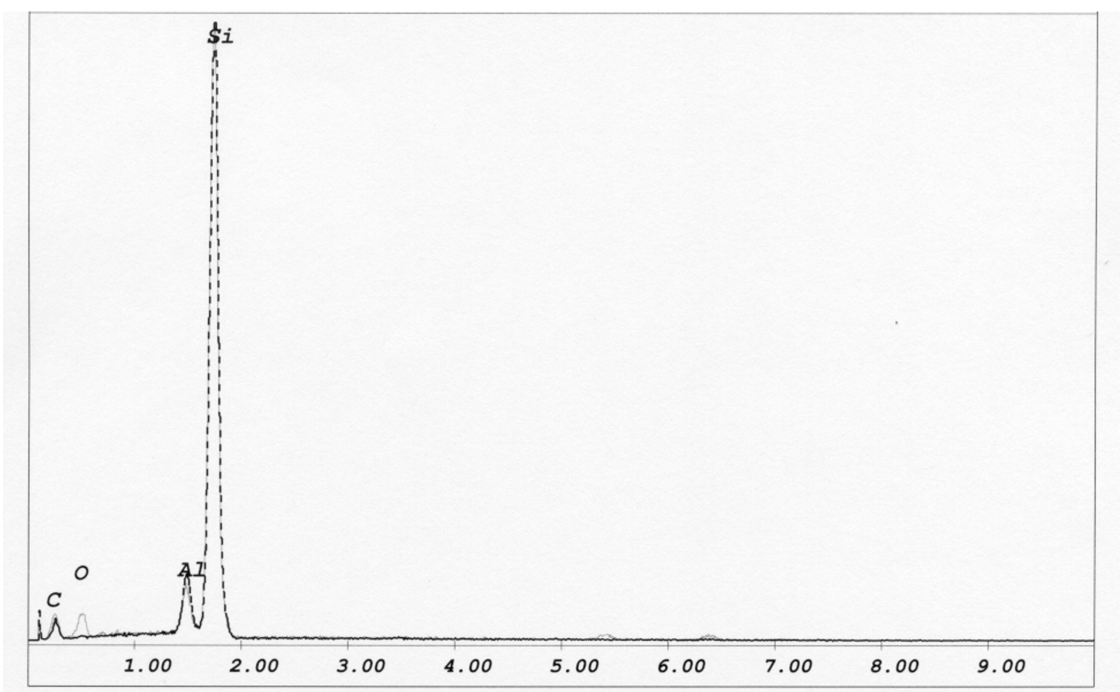
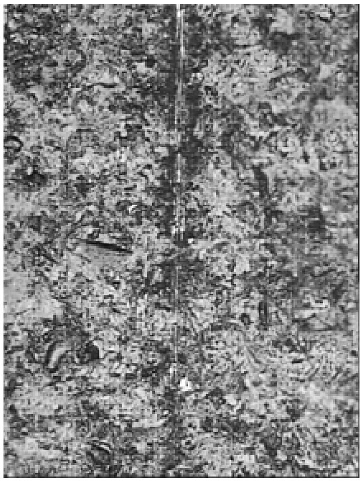


Figure 4. Adhesion test performed on specimen shown in figure 3b. a) Region of initial failure. b) Complete delamination.

a 

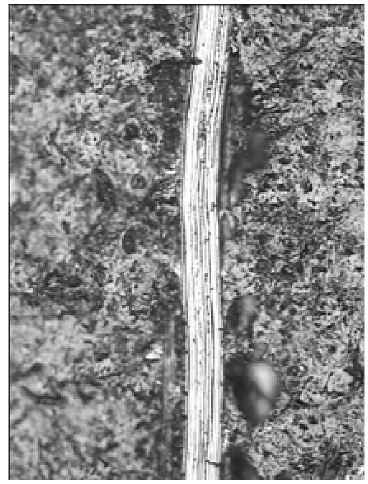
b 

Figure 5. Adhesion test performed on specimen shown in figure 3b. a) Region of initial failure. b) Complete delamination.

Figure 6. BSE of scratch tested specimen. The specimen was tilted 45 degree. The dark region (low z- contrast) is the SiC film, the bright region (high z- contrast) is the HK40 substrate.