Ablation behavior of monolayer and multilayer Ir coatings under carburizing and oxidizing oxyacetylene flames

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ABSTRACT

Iridium is one of the most promising candidates for protective barrier of refractory materials to endure high service temperature. The multilayer iridium coating was produced by a double glow plasma process on the polished tungsten carbide substrates, compared with monolayer. The ablation behaviors of the monolayer on the unpolished and polished substrates were investigated under carburizing and oxidizing oxyacetylene flames, respectively, at the same time the multilayer coating ablated under oxidizing flames. Multilayer coating was a polycrystalline phase with the preferential (220) orientation. Monolayer on the unpolished substrate had fine coarse grains and some small microcracks were present. Multilayer consisted of columnar grains with some voids between the grains boundaries. The formation of a WIr phase in the as-deposited multilayer was attributed to high deposition temperature. The monolayer could endure high temperature up to 1800 °C in carburizing flame. The substrates could be protected more effectively by multilayer than monolayer at 2000- 2200 °C in oxidizing flame.

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1. Introduction

Iridium (Ir) is one of the most corrosion-resistant metals. Due to a high melting point (T_m=2447 °C), low oxygen permeability, good chemical compatibility, superior oxidation resistance and the absence of carbide below the eutectic temperature of 2280 °C [1,2], the Ir acts as an effective diffusion barrier for inward oxygen and outward carbon [3]. The Ir is one of the most promising candidates for protective coating of either structural carbon material or rhenium (Re) components to endure high service temperature and ensure long service life [4,5]. The Ir coating can be prepared by chemical vapor deposition CVD [5–8], magnetron sputtering (MS) [1–4, 9,10], pulsed laser deposition (PLD) [11], laser-induced chemical decomposition (LICD) [12], electrodeposition (ED) [13–15], and double glow plasma (DGP) [16]. At present, the Ir/Re composite combustion chamber used as liquid rocket motor could operate at temperatures up to 2200 °C [17]. NASA has reported the development of Ir coated Re rocket chamber technology, allowing an increase in satellite life from 12 to 15 years, and gaining 30–60 M$ in the added revenue per satellite [18].

In previous publications, the monolayer Ir coating was produced by DGP onto the carbon/carbon composite [19,20], graphite [21,22], tungsten carbide (WC) [23], molybdenum [24–26], and niobium substrates [16,27]. Due to the large coefficient of thermal expansion (CTE) mismatch between the carbon material (carbon/carbon composite 1.1×10^{-6} °C^{-1} [28], or the graphite 3.8×10^{-6} °C^{-1} [22]) and the Ir coating (6.2×10^{-6} °C^{-1}), the Ir-carbon bond became weak after oxidizing ablation by an oxyacetylene torch at ~2000 °C [20,22]. Although the CTE of the molybdenum (5.0×10^{-6} °C^{-1}) is close to that of the Ir, the surface of the coating had pores, bulges and cracks after exposure to oxidizing flame [26]. The multilayer Ir coating could be more favorable as a protective layer against oxidation at ultra-high temperature [6]. The porosity on the surface of the multilayer Ir coating deposited on niobium substrate was less than monolayer after oxidizing ablation. However, the multilayer was debonded to the niobium substrate due to the effects of the underdense coating, volatile gas and interdiffusion between the coating and the substrate [27]. To date, the ablation behavior of Ir coating on WC substrate is not further studied. In the current work, the multilayer Ir coating was fabricated on surface of WC substrates (low CTE 5.2×10^{-6} °C^{-1} [29]) by a DGP process. We investigate the ablation behavior of monolayer and multilayer under carburizing and oxidizing oxyacetylene flames.

2. Experimental

A detailed description of the DGP process was reported elsewhere [30,31]. The Ir coating was performed in a double glow

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plasma surface-alloying device, in which a low temperature plasma was produced by a glow discharge process in a vacuum-sputtering chamber. Three electrodes were designed in the chamber: one anode and two negatively charged members, the cathode where the working piece was placed, and the source electrode where the desired element was located. An Ir plate (purity: 99.95%, ⌀ 50 × 3.5 mm) was used as the target material. The ceramic WC sheet with an irregular shape was the commercial product used as the substrate, which was obtained from cemented WC scrap by an electrolysis process. The substrates were polished by metallographic abrasive papers, then using the 1 μm diamond media, and cleaning with acetone followed the unpolished and polished substrates. The monolayer and multilayer were produced on the surface of the polished substrates. Argon was used as working gas. The deposition conditions were: base pressure 4×10^{-3} Pa, target bias voltage −900 V, substrate bias voltage −350 V, substrate temperature 850–900 °C, working pressure 35 Pa, target-substrate spacing 15 mm, deposition time 3 h for monolayer, total deposition time 3 h for multilayer repeating the same process three times. Meanwhile, the monolayer was also deposited on an unpolished WC substrate. The deposition parameters were: substrate voltage −400 V, target voltage −900 V, working pressure 35 Pa, target-substrate spacing 15 mm, and deposition time 1 h.

The as coated specimen was exposed in the oxyacetylene flame. The specimen was laid on an aluminium oxide (Al₂O₃) firebrick. The surface temperature of the specimen was monitored with an optical pyrometer. The ablation temperature was fixed at 1800–2200 °C by adjusting the acetylene/oxygen ratio. The specimen was placed vertically to the oxyacetylene torch having a nozzle tip with 2 mm diameter. The distance between the oxyacetylene torch and the specimen was ~25 mm. The monolayer on the unpolished substrate was exposed to air for 90 s under carburizing flame. The monolayer and multilayer on the polished substrates were ablated under oxidizing flames for 60 s and 90 s, respectively. The carburizing and oxidizing flames were used according to the changes of the acetylene/oxygen ratio.

The top surface and fracture surface morphologies of the coating were imaged using scanning electron microscopy (SEM, Quanta 200, FEI Company). The chemical composition was examined by energy dispersive X-ray spectroscopy (EDS, LINK-860) attached to the SEM. The phase and texture were determined by X-ray diffraction (XRD, D8Advance, Bruker Company) using Ni-filtered Cu Kα radiation at a scanning rate of 0.5 s^{-1} scanning from 10° to 90° of 2θ.

3. Results and discussion

3.1. Characterization

In previous publication [23], the monolayer was a polycrystalline phase with a preferred (220) orientation. A new WIr phase was examined by XRD, which was attributed to the high deposition temperature. The interfacial reaction between the coating and the substrate led to WIr formation, which was related to the high-temperature substrate if the compound formation was thermally activated. The surface was the homogenously fine stripy-grained aggregates. Fig. 1 shows the macroscopically photograph and SEM image of the monolayer on the unpolished substrate. The coating was completely deposited on the surface of the substrate (Fig. 1 (a)). Some large particles were apparent due to the rough surface of the unpolished substrate. It was found that the thin Ir coating was deposited at the margin of the specimen (dashed line) due to the effect of substrate edge [32]. Here, the thin coating would be easily oxidized. The surface was nonhomogeneous, as observed in Fig. 1(b). Some small microcracks were found on the surface due to the effect of the substrate surface. In previous publication [23], the coating was smooth and no cracking formed on the surface of the polished WC substrate.

Fig. 2 shows the XRD pattern and SEM micrographs of the multilayer. Fig. 2(a) displays the XRD pattern of the coating. A WIr phase was also found on the XRD pattern. The scan data exhibited 2θ peaks at 40.65°, 47.40°, 69.18° and 83.46°, respectively, corresponding to the (111), (200), (220) and (311) peaks as compared with the standard d-values taken from JCPDS (46-1044). The multilayer was a polycrystalline structure. The preferential orientation can be obtained by a texture coefficient ($TC_{(hkl)}$) that can indicate the strongest reflection along the (hkl) plane at different conditions. This factor can be calculated by the following relation [33]:

$$TC_{(hkl)} = \frac{I_{(hkl)}/I_{0(hkl)}}{1/N\left[\sum I_{(hkl)}/I_{0(hkl)}\right]} \tag{1}$$

where TC is the texture coefficient of the (hkl) plane, $I_{(hkl)}$ is the measured intensity (hkl) plane, $I_{0(hkl)}$ is the corresponding standard recorded intensity in JCPDF card file, and N is the number of preferred growth directions. From the calculation, $TC_{(111)}$, $TC_{(200)}$, $TC_{(220)}$ and $TC_{(311)}$ are 0.13, 0.31, 3.35 and 0.22, respectively. It indicates that the multilayer exhibited a preferred (220) orientation. The preferential orientation of the grain was due to the initial nuclei preferred growth on the surface of the substrates. The (111) orientation is favored because the (111) planes are closest-packed
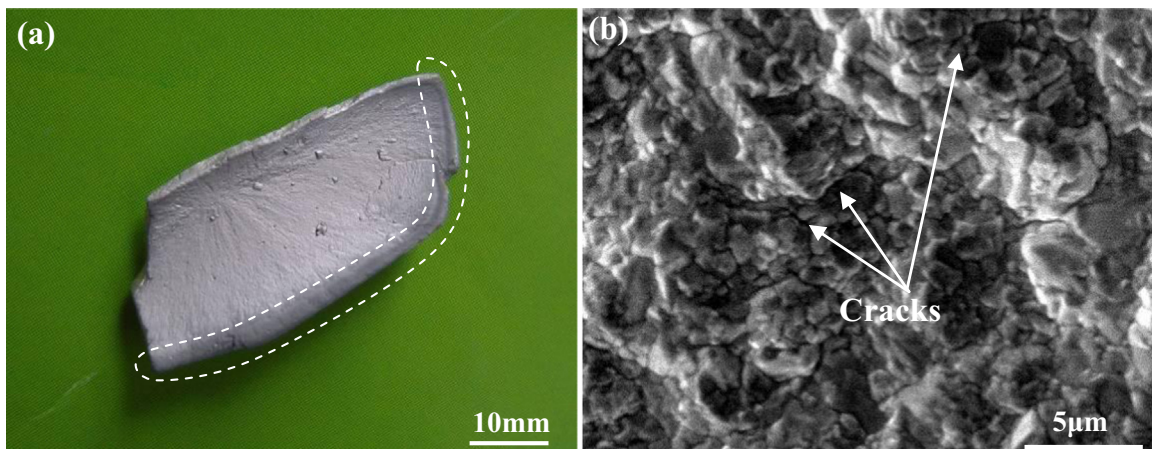

Fig. 1. Macroscopically photograph (a) and SEM image (b) of the monolayer on the unpolished substrate.