Journal of Alloys and Compounds 745 (2018) 271–281

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

An Nb_2O_5-SiO_2-Al_2O_3/$NbSi_2$/Nb_5Si_3 multilayer coating on Nb-Hf alloy to improve oxidation resistance

Yulin Ge [a, b], Yaming Wang [a, b, *], Junchen Chen [a, b], Yongchun Zou [a, b], Lixin Guo [a, b], Jiahu Ouyang [a, b], Dechang Jia [a, b], Yu Zhou [a, b]

[a] Institute for Advanced Ceramics, Harbin Institute of Technology, Harbin 150001, China
[b] Key Laboratory of Advanced Structural-Functional Integration Materials & Green Manufacturing Technology, Harbin Institute of Technology, Harbin, 150001, China

ARTICLE INFO

Article history:
Received 15 October 2017
Received in revised form
7 February 2018
Accepted 9 February 2018
Available online 10 February 2018

Keywords:
Nb alloy
Multilayer coating
Pack cementation
Microarc oxidation
High temperature oxidation resistance

ABSTRACT

An Nb_2O_5-SiO_2-Al_2O_3/$NbSi_2$/Nb_5Si_3 multilayer coating is fabricated by a two-step method of pack cementation and microarc oxidation. The isothermal oxidation resistances of Nb_2O_5-SiO_2-Al_2O_3/$NbSi_2$/Nb_5Si_3 multilayer coated and $NbSi_2$/Nb_5Si_3 coated Nb alloy are investigated at 1250 °C. The results show that the oxidation kinetics of $NbSi_2$/Nb_5Si_3 coated Nb alloy follows a parabolic-like law with the parabolic rate constant k_p of 0.79 $mg^2\,cm^{-4}\,h^{-1}$. Whereas the multilayer coated Nb alloy exhibits better oxidation resistance with a lower k_p of 0.06 $mg^2\,cm^{-4}\,h^{-1}$ for the first 15 h oxidation period, and a comparable oxidation resistance to that of $NbSi_2$/Nb_5Si_3 coated Nb alloy in the following oxidation period. The multilayer coating has successfully retarded the oxidation process, which is attributed to the formation of Al_2O_3 and glassy SiO_2 within the Nb_2O_5-SiO_2-Al_2O_3 outer layer.

© 2018 Elsevier B.V. All rights reserved.

1. Introduction

Nb alloys have great potential for metal thermal protective system (MTPS) due to the high melting point and the excellent mechanical properties at high temperature. However, the "pest oxidation" limits its extensive applications because of the non-protective oxidation products (Nb_2O_5 or NbO_2 phases) above 600 °C [1–3]. Thus, it still remains a great challenge for researchers to improve the oxidation resistance property of Nb alloys [4]. In addition, in ultrahigh temperature environment, the thermal radiation dominates heat transfer, which raises the demand of high emissivity coatings to intercept the heat flux to the metallic substrate by radiation. Therefore, the combined properties of high temperature oxidation resistance and high emissivity are highly required for Nb alloys as MTPS.

Currently, silicide coatings have been developed by methods of plasma spraying [4–6], molten salt [7], magnetron sputtering [8] or pack cementation (PC) [9,10], which were proved to be helpful to enhance the oxidation resistance for Nb alloys [11]. Compared with other coating systems, a silicide coating fabricated by PC has a series of advantages such as good compactness, metallurgical adhesion between coating and substrate and outstanding oxidation resistance performance. Furthermore, the silicide coatings modified with elements such as Al, Y, B, Ge, Cr [12,13], Re [14,15], Ce [16] or Zr [17] have been considered to be effective on elevating the oxidation resistance. The co-deposition of Al and Y inhibits oxygen diffusion by by-product of Al_2O_3 [18–20]. The co-deposition of B and Ge increases the fluidity and decreases the thermal expansion of the oxide scales [21–24]. However, the limited oxidation resistance of single silicide coatings drives researchers to develop multilayer coatings by method-cooperation [25]. Su [26] deposited Mo on Nb/Nb_5Si_3 substrate followed by co-deposition of Si and B. At the same period, Bacos [27] deposited Cr instead of Mo on Nb/Nb_5Si_3 substrate followed by co-deposition of Si and B. The two-step deposited coatings retard the diffusion of oxygen due to the formation of borosiliconized healing glass layers. Sun et al. [28] proposed a $MoSi_2$-$NbSi_2$ multilayer coating through PC followed by plasma spray technology, which possesses better oxidation resistance compared with the single $NbSi_2$ coating or sprayed $MoSi_2$ coating. It has been confirmed that the silicide coatings are effective to improve the oxidation resistance properties, which are attributed to the formation of protective oxide scales. Unfortunately the

* Corresponding author. Institute for Advanced Ceramics, Harbin Institute of Technology, Harbin 150001, China.
E-mail address: wangyaming@hit.edu.cn (Y. Wang).

https://doi.org/10.1016/j.jallcom.2018.02.107
0925-8388/© 2018 Elsevier B.V. All rights reserved.

thermal emissivity of the silicide coatings mentioned above is as low as 0.1–0.3 [29,30]. The low thermal emissivity of silicide coating restrains the heat radiation, thus increasing the heat flux to the Nb alloy substrate.

Therefore, we propose an alternative strategy to elevate the surface emissivity by introducing a high emissivity layer on the silicide coatings surface. It has been confirmed in our previous research that fabricating high emissivity coatings using microarc oxidation (MAO) is feasible [29]. In this study, we emphasize on the fabrication of multilayer coatings of PC/MAO and exploring the effects of MAO outer layer on high temperature oxidation resistance properties of PC coated Nb alloy. Firstly, an $NbSi_2/Nb_5Si_3$ layer is formed on the bare Nb by depositing Si by PC process. Then, MAO is applied to grow an Nb_2O_5-SiO_2-Al_2O_3 outer layer on the $NbSi_2/Nb_5Si_3$ layer, aiming to enhance the oxidation resistance and surface emissivity. The present study focuses on analyzing the isothermal oxidation behaviors of the single PC ($NbSi_2/Nb_5Si_3$) coated samples and the multilayer (Nb_2O_5-SiO_2-$Al_2O_3/NbSi_2/Nb_5Si_3$) coated counterparts by comparing the oxidation kinetics.

2. Experimental procedures

2.1. Coating fabrication

The Nb-Hf alloy, with a chemical composition of Nb-8.9Hf-1.2Zr (at.%), was prepared by vacuum arc-melting. The samples were machined to the dimensions of 10 mm × 10 mm × 0.5 mm. All the sharp corners and edges were chamfered. All samples were polished with 600, 800 and 1000 grit SiC papers successively, and then cleaned in ethanol and dried by a hot air stream.

In the first step, the samples were coated by the PC method. The diagrammatic sketch of PC process is shown in Fig. 1 (a). The pack powders were mixed uniformly at the ratio of 16Si:79Al_2O_3:5NaF (wt.%) before the Si-deposition process. The samples were fully embedded in a cylindrical alumina crucible with a distance more than 8 mm from one to another as well as the inner wall of the crucible. The remaining spaces were filled with the mixed pack powders. Then the crucible was sealed with aluminum phosphate binder, and put into a tube furnace. The furnace was heated under argon atmosphere with a constant heating rate of 8 °C min^{-1} up to 1300 °C, held for 8 h at 1300 °C, and then cooled down to room temperature with a constant cooling rate of 5 °C min^{-1}. The as-achieved Si-deposited coatings with a thickness of 100 ± 3 μm are named as $NbSi_2/Nb_5Si_3$ coatings.

In the second step, the MAO process was performed in 15 g L^{-1} $NaAlO_2$ aqueous electrolyte, and the diagrammatic sketch of MAO process is shown in Fig. 1 (b). The PC coated samples were placed in the electrolyte bath working as anode and a steel plate was used as cathode. A 500 V square wave voltage was applied on the two electrodes for 5 min, and the frequency and duty cycle were 600 Hz and 8% respectively. Thus a MAO layer of 4–8 μm in thickness was formed on the $NbSi_2/Nb_5Si_3$ coating surface. Then, the as-achieved multilayer coatings are named as Nb_2O_5-SiO_2-$Al_2O_3/NbSi_2/Nb_5Si_3$ multilayer coating.

2.2. Oxidation tests

The isothermal oxidation test of the bare alloy, $NbSi_2/Nb_5Si_3$ and Nb_2O_5-SiO_2-$Al_2O_3/NbSi_2/Nb_5Si_3$ coated samples were conducted in static air at 1250 °C for 50 h. All of the samples were heated to 1000 °C with a constant heating rate of 4 °C min^{-1} and then to 1250 °C with a heating rate of 2 °C min^{-1}. After that, the samples were held at 1250 °C for 0, 2, 5, 10, 15, 20, 30, 40 and 50 h, respectively, and cooled down to room temperature out of the furnace naturally. Each sample was weighted by a precision analytical balance (CPA225D, Sartorius, Germany) with an accuracy of 10^{-5} g before and after the isothermal oxidation test.

2.3. Microstructure characterization

The surface and cross sectional morphologies of the coatings were investigated by using a scanning electron microscope (SEM; Helios Nanolab600i, FEI, U.S.A.) with an operating voltage of 20 kV. The phase compositions were characterized by an X-ray diffraction (XRD; Empyrean, PANalytical, Netherlands) with a Cu K_α radiation working at 30 mA and 40 kV. The phase composition of the Nb_2O_5-SiO_2-$Al_2O_3/NbSi_2/Nb_5Si_3$ multilayer coating was further analyzed by a transmission electron microscope (TEM; Tecnai F30, FEI, U.S.A.)

3. Results and discussion

3.1. Microstructure of coatings

The silicide coating was firstly deposited by PC method that works as an oxidation resistance coating. The microstructure and composition of the $NbSi_2/Nb_5Si_3$ coating are shown in Fig. 2. The coating surface is smooth with an average grain size of 6.5 μm (Fig. 2 a). $NbSi_2$ (JCPDF No. 00-008-0450) is the main phase of the dark grey bulk layer according to the XRD pattern (Fig. 2 b). The white transitional layer (3 μm in thickness) beneath the $NbSi_2$ layer is identified as Nb_5Si_3 according to the EDS result (in position A) and the Nb-Si binary phase diagram. As seen in Fig. 2 (c) and (d), the stripe inclusions within $NbSi_2$ layer are speculated to be columnar crystal grains growing perpendicular to the surface [31]. The light

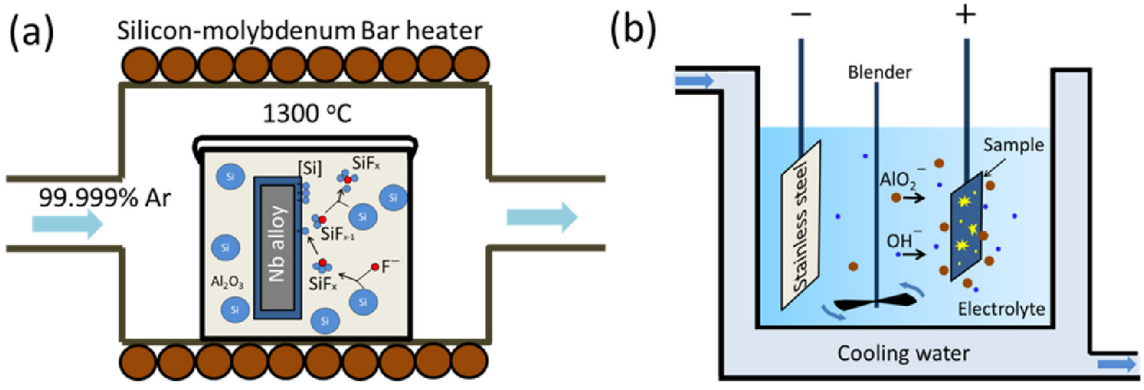

Fig. 1. Diagrammatic sketch of (a) PC and (b) MAO process in this work.

Download English Version:

https://daneshyari.com/en/article/7992728

Download Persian Version:

https://daneshyari.com/article/7992728

Daneshyari.com