



Effects of deposition temperature on the kinetics growth and protective properties of aluminide coatings



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ARTICLE INFO

Article history:

Received 3 January 2015

Received in revised form 21 January 2015

Accepted 22 January 2015

Available online 30 January 2015

Keywords:

Aluminide coating

CVD

Deposition temperature

Microstructure

Kinetics behavior

ABSTRACT

Chemical vapor aluminizing is a major production technique used for producing aluminide coatings on gas turbine blades to form dense and adhesive alumina surface layers and further prolong their service lives at high temperature. A chemical vapor deposited aluminizing process had been employed to deposit nickel aluminide coatings under the different deposition temperature. The phase structure, morphology, chemical composition, kinetics growth mechanism and oxidation/corrosion behavior of the aluminide coatings were studied in detail. The results show that the phase structure, microstructure and composition of the aluminide coatings are strongly influenced by the deposition temperature during chemical vapor deposition (CVD). The higher is the deposition temperature, the easier is the formation of pure β -NiAl phase. Along with the increment of the deposition temperature, the coatings' surface will become smooth and dense, while the ridges along grain boundaries can be simultaneously approach to flat in a certain extend. Large numbers of elements of Cr, Mo and W within diffusion zone that could probably form a thin film layer, which is usually so-called as diffusion barrier. The key step of growth of coating grain is the formation of a volatile aluminium subhalide by controlling of the suitable deposition temperature during CVD. Based on the results of isothermal oxidation and hot corrosion kinetics curves, the best deposition parameter can be gained among the four coatings when the deposition temperature is adopted to be 1323 K.

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1. Introduction

Nickel-base superalloys have been widely used in established military and commercial gas turbine engines for combustion cans, blades and other high temperature structural components due to their good combination of high temperature strength and excellent oxidation resistance for long-term exposure. In gas turbine applications, the components are subject to repeated oxidation and hot corrosion cycles, which occur during cruise and marine environment. The property of superalloys under oxidation and corrosion environments is an important issue. In addition, the operation temperature of modern gas turbine engines is getting higher to improve engine efficiency and performance [1,2]. For superalloys used at high temperature, it is a common practice to coat a high aluminium-containing layer onto the surface by pack cementation aluminizing [3], out-of-pack [4], slurry [5] and chemical vapor deposition (CVD) [6] processes. The diffusion

aluminizing process is one of the basic methods of surface protection for gas turbine blades and vanes against oxidizing influence of exhaust gases.

Independently from the used aluminizing method, the coating formation mechanism consist in the reaction high activity process, the inward diffusion of aluminium is the dominant growth mechanism. It has been proven by using several powders of Ni_2Al_3 , NH_4Cl and Al_2O_3 with proper proportion of mixture that β -NiAl is the basic phase constituent [7]. During the process which is so-called as the low activity aluminizing, the outward diffusion of nickel is the dominant growth mechanism. In the case of the high activity process it is necessary to perform the additional heat treatment in order to obtain the transformation of Ni_2Al_3 phase into β -NiAl [8]. The out-of-pack technique and vapor phase aluminizing are the extension of the pack cementation method. The Al powder and the Cr–Al granules are in proper distant above from the surface of coated elements and are used as the source of aluminium in those methods. The process is accompanied with the argon or hydrogen atmosphere in the retort furnaces. Aluminium fluoride or ammonium fluoride is usually used as an activating agent

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[9,10]. It has been proven, that it is possible to perform aluminizing of internal cooling channels of blades and vanes using out-of-pack method [9].

It should be also considered that there is a limitation of parameters control during the out-of-pack or vapor phase aluminizing process. It is a result of using the solid-state activating agent introduced before the process. The largest possibilities of parameters control are to be using of the chemical vapor deposition process. Up to now, large of blades and vanes are coated with diffusion aluminide coatings by chemical vapor deposition [2,9]. Sun et al. [11] conducted the CVD aluminizing process with application AlCl_3 and H_2 flow, which were produced inside the external generator over the aluminium and aluminium oxide powder. It has been proven, that the temperature growth in the AlCl_3 generator results in the growth Al concentration in the aluminide coating to above 60 at.%. The conducted research on obtaining platinum-modified aluminide coating combined with the CVD aluminizing process by Warnes and Punola [12] showed that the coatings obtained during the low activity process were characterized by the best oxidation resistance. Furthermore, Warnes [13] also proved the possibility of modifying the aluminide layers containing silicon and hafnium while using three external generators. Several authors confirmed previously that the deposition temperature of kinetics growth of aluminide coatings from 1123 K to 1323 K had a significant influence on the coating thickness [9,13].

However, kinetics growth and microstructure evolution of aluminide coatings preparation under the different deposition temperature have not been investigated in detail up to now, and no isothermal oxidation and hot corrosion compared results are available. In this study, a chemical vapor deposited aluminizing process has been employed to deposit nickel aluminide coatings on the nickel-based single-crystal superalloys under the different deposition temperature. The phase structure, morphology, chemical composition and kinetics growth mechanism of the aluminide coatings have been investigated in detail. Simultaneously, Isothermal oxidation and hot corrosion tests at 1323 K and 1173 K have been conducted to evaluate the performances of the aluminide coatings with different processing parameters, respectively.

2. Experimental

2.1. Preparation of aluminide coatings

The substrates (15 mm × 10 mm × 1.5 mm) were ground before the diffusion aluminide coatings of NiAl were deposited by CVD reactor. In the CVD aluminization method, AlCl_x ($x < 3$) precursor vapors reacted with Ni from the Ni-based superalloy to form β -NiAl in the presence of H_2 and Ar, at temperatures of about 1233–1323 K. The vertical hot-wall reactor was used in this study. The gas pipe lines were wrapped with heating tapes from the vaporizer to the reactor, and then the temperature was controlled to ~673 K, which was slightly above the sublimation temperature of AlCl_x and further prevented its condensation in the gas lines. Since an excess of AlCl_x was used to ensure the uniformity of the NiAl coating, a water-cooled chamber was simultaneously used to collect unreacted AlCl_x waste before it entered into a liquid-ring vacuum pump. The parameters concerning particular processes of the four aluminide coatings are presented in Table 1.

2.2. Isothermal oxidation and hot corrosion tests

The isothermal oxidation of the four aluminide coatings was carried out at 1323 K in a furnace with airflow to evaluate of the coating samples' performance of oxidation resistance influenced by the deposition temperature. Before the oxida-

tion experiment, the specimen crucibles were heated at 1373 K until a constant weight was obtained. After the oxidation for a certain time, the oxidized specimens were removed from the furnace and air-cooled to room temperature. The specimens were then balanced with an automatic electronic balance (BS110S, 0.1 mg), and the weight change as a function of time was recorded.

The solution including of mixed of aviation fuels and artificial seawater was used as corrosive medium during hot corrosion at 1173 K in a gas-fired furnace. The composition of artificial seawater was including of NaCl, MgCl_2 , KCl and CaCl_2 with concentration (g/L) ratio of 27:12.8:1:1, respectively. After the hot corrosion for 25 h, 50 h, 75 h and 100 h at a fixed time, the four coating samples were then removed from the furnace and air-cooled to room temperature. Similarly, the four samples were also balanced with an automatic electronic balance, and the weight change as a function of time was similarly recorded.

2.3. Characterizations

After preparing of the coating samples, the surface of the aluminide coatings was characterized using the scanning electron microscope (SEM, FEI-Quanta 600). The coating samples were embedded in a transparent cold-setting epoxy and then sectioned, ground and polished with diamond pastes down to 1 μm . SEM equipped with EDS (Oxford INCAx-sight 6427) was applied for the microstructure and composition evaluation. X-ray diffraction (XRD, Bruker D8 Advance) with Cu K_α radiation at a scan rate of 4°min^{-1} was employed to analyze the phase structural evolution of the four coatings with the difference of deposition temperature. Coatings without polishing were used directly for XRD measurement.

3. Results and discussion

3.1. Phase structure of aluminide coatings

The XRD patterns of the four aluminide coatings fabricated under the different deposition temperature are compiled in Fig. 1. Contents of the four aluminide coatings are mainly including of Ni and Al elements. Correspondingly, the dominant phases of the four coatings are detected to be β - $\text{Ni}_{0.9}\text{Al}_{1.1}$ for samples A and B, β - $\text{Ni}_{1.1}\text{Al}_{0.9}$ for sample C and β - $\text{Ni}_{0.58}\text{Al}_{0.42}$ for sample D, respectively. Several mixed phases of the aluminide coatings for samples A and B are still coexistence, which are including of phases of α -Cr, α -Co, Al_9Co_2 and Al_2Cr_3 . The inward diffusion of Al element and outward diffusion of Ni element have simultaneously in progress due to the different compositions of Al/Ni elements within the coating and substrate [14]. It can be calculated from the results of the XRD patterns shown in Fig. 1 that content of Al element in the four coatings has a value change from 55 at.% down to 42 at.% accompanying with the increment of the deposition temperature. This phenomenon is primarily related to the interdiffusion behavior between Ni and Al elements. Meanwhile, it is also interesting to see that all the peaks (Fig. 1b–d) slightly shift to the larger 2θ -value (the smaller d -values) compared with the XRD pattern shown in Fig. 1a and the crystal lattices become gradually lower, the main reason is probably attributed to the outward diffusion of Cr and Co elements from the substrate to the coating. The higher is the deposition temperature, the more is the outward diffusion of refractory metal elements within substrate [15].

The activity of Al element is lower than that of Ni element when the deposition temperature is adopted to be above 1223 K. The diffusion velocity of Ni element from the substrate to the coating is faster than that of Al element occurred in opposite direction. At that time, AlCl_x ($x < 3$) precursor vapors can easily react with Ni from the Ni-based substrate to form β -NiAl in the presence of carrier gases, at temperature of above 1233 K. Hence, the growth pattern of coating grains is defined to be outward growing on top of

Table 1

The list of parameters used for deposition of the four aluminide coatings.

Sample	Temperature (K)	Pressure (mbar)	H_2 flow (L/min)	Ar flow (L/min)	HCl flow (L/min)
A	1233	150	10	5	1.5
B	1263	150	10	5	1.5
C	1293	150	10	5	1.5
D	1323	150	10	5	1.5

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