RESEARCH PAPER

Comparative analysis of CoCrAlY coatings at high‑temperature oxidation behavior using diferent reinforcement composition profles

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Abstract

The oxidation behaviour of plasma-sprayed carbides and oxides reinforced composite CoCrAlY coatings at 700 °C was examined in this paper. CoCrAlY was reinforced with Al_2O_3 + YSZ, CeO₂, WC–Co, Cr₃C₂-NiCr individually to produce four diferent coatings. SEM, EDS, and XRD are used to analyse the oxide scales formed during the oxidation process on the surface of the coating. From the results, it is found that all of these coatings had a parabolic weight gain, suggesting the existence of a shielding oxide scale on the surface, whereas changes in total weight gain are observed. The oxidation resistance of $CoCrAY + A1₂O₃ + YSZ$ coating is better and exhibits 28% less weight gain than other coatings. During oxidation, coating shows slow scale growth of kinetics due to thermodynamically stable A_1O_3 . The CoCrAlY +2% CeO₂ coating showed the lowest oxidation resistance of all the coatings.

Keywords Plasma spray · CoCrAlY · Carbides · Oxides · High-temperature oxidation

1 Introduction

High-temperature applications like boiler components are commonly used in power plant sectors, and these are subjected to extreme working conditions; it could be circulation of high pressure fuid/water or steam, reaction from sulphur element presence in coal $[1, 2]$ $[1, 2]$ $[1, 2]$. The component may undergo surface degradations in the form of erosion, oxidation, hot corrosion, and wear [[3,](#page-6-2) [4\]](#page-6-3). Boilers are recognised as one of the severe challenges in the course application, and development of the boiler using right combination of materials is in wide scope [\[5–](#page-6-4)[7\]](#page-7-0). Oxidation is a major material degradation mechanism that occurs mostly in high-temperature situations such as IC engines, jet engine components, gas turbines, coal

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pipelines, and offshore oil $[8-10]$ $[8-10]$. Oxidation of alloys reduces ductility, tensile strength, corrosion, and erosion properties. Currently, nickel-based super alloys having excellent hightemperature strength are being used to manufacture the above mentioned components. However, theses alloys exhibit poor surface properties at higher temperature [\[11–](#page-7-3)[13](#page-7-4)]. Hence, development of coatings which could able to combat against these degradations is of high interest. Several industrial components commonly have thick coatings of MCrAlY applied by plasma spray to increase service life in an oxidising environment $[1, 2]$ $[1, 2]$ $[1, 2]$ $[1, 2]$ $[1, 2]$. The efforts have been made by some researchers to advance the properties of MCrAlY coatings by the addition of active elements such as Ti, Zr, Hf, Si, Ta, Ce, Ru, Ir, and Mo [\[6](#page-6-5)]. However, these changes are not beneficial to work against critical environment like combined oxidation, corrosion wear, and erosion. Furthermore, the MCrAlY coating is reinforced with carbides and oxides to combat against both high-temperature oxidation and wear degradations [\[7](#page-7-0)].

WC–Co and Cr_3C_2 -NiCr are the carbide coatings added with small amount of metallic binder are used for hightemperature tribological applications. These coatings have exceptional wear resistant properties; they are not most preferable in high-temperature oxidation and corrosion environments [\[11\]](#page-7-3). The inclusion of metal oxide particles benefts to strengthen the high temperature coating materials, which also raises the coating's hardness and wear resistance.

The addition of oxide particles to high-temperature coating materials strengthens the coating and eventually increases the hardness and wear resistance [[9](#page-7-5)]. Most of the research articles published associated to hard phase reinforced composite coatings focussed on tribological behaviour and phase development [\[12–](#page-7-6)[14\]](#page-7-7). However, the studies on high-temperature oxidation behaviour of these composite coating is on mandate as these composite coatings exposed to various environment.

Hatami et al., [[15](#page-7-8)] studied the oxidation behavior of 0–15% YSZ mixed (5% intervals) with CoNiCrAlY powder by HVOF technique. The result reported that coating with 5%YSZ obtained the better oxidation resistance at 1000 °C. Similarly, Ghadami et al. [[16](#page-7-9)] investigated the oxidation behaviour of HVOF-sprayed CeO₂-gradient NiCrAlY coatings at 1000 °C. It is reported that functionally graded NiCrAlY coating reinforced with nano-CeO₂ had a greater oxidation resistance than the mono-layered coatings with lower oxide growth rate. The rest of due to controlling Al and O difusion due to the formation of relatively thin, dense, and sticky Al_2O_3 oxide scale.

Hang et al. [[17](#page-7-10)] investigated the elevated temperature oxidation behavior of WC-12Co and rare earth-modifed WC-12Co HVOF coating. The $CeO₂$ -modified coatings have good oxidation resistance at the temperature below 550 ℃. Whereas above 650 ℃ due to the occurrence of anomalous oxidation, the oxidation mechanism changes. Similarly, comparative oxidation studies of NiCoCrAlYSiHf and NiCoCrAlYSiHf+NiAl composite coatings were reported by Yao et al. $[18]$ $[18]$ at 1100 °C. NiCoCrAlYSiHf + NiAl composite coating exhibited good oxidation resistance with the Kp value of less than two-ffths of that for NiCoCrAlYSiHf coating during isothermal oxidation. Shi et al., [\[19](#page-7-12)] recently reported the elevated temperature oxidation behaviour of Ni and NiCrAlY-based composite coatings by reinforcing with $TiO₂-ZnO$ elements. The reinforcement in the coating difuses to the surface at 800 °C and 1000 °C.

In case of NiCrAlY composite coatings, the difusion behaviour was slower compared to Ni-based coatings at 800 °C. Whereas, difusion behaviour greatly increased as temperature closer to 1000 °C. Similarly, Reddy et al., [[20\]](#page-7-13) studied the cyclic oxidation behavior of plasma-sprayed NiCrAly coating by addition of $TiO₂$ and $Cr₂O₃-YSZ$ at 700 °C. Cr₂O₃ + YSZ-reinforced NiCrAlY coating was found to exhibit highly resistive as compared with $TiO₂$ -reinforced NiCrAlY coating in the oxidation environment.

The comparative studies with both oxides and carbides were reported in Ref [[16–](#page-7-9)[20\]](#page-7-13) related to the high temperature oxidation and erosion behaviour of HVOF-sprayed NiCrSiB/ Al_2O_3 , NiCrSiB/n-Al₂O₃ and NiCrSiB/WC–Co coatings on an SS304 steel substrate. NiCrSiB/ Al_2O_3 coatings exhibits superior oxidation resistance to that of NiCrSiB/WC–Co coatings due to the presence of protective oxide layers like Cr_2O_3 , Al_2O_3 , NiCr₂O₄, and NiO. The weight gain of Al_2O_3 -reinforced coating was less than that of n-Al₂O₃ and WC–Co-reinforced coatings by 43% and 51% respectively. The presence volatile oxide WO_3 in WC–Co-reinforced coatings had least oxidation resistance.

From the above discussion, it can be inferred that, the addition of oxide or carbide-reinforced coating will be benefcial for both oxidation and erosion/wear resistance. In the present investigation, attempt has been made to develop four diferent coatings by addition of diferent oxides and carbides reinforcement such as Al_2O_3 , CeO₂, WC and Cr₃C₂ with CoCrAlY coating. A comparative study on cyclic oxidation behavior of these plasma-sprayed composite coatings at 700 °C have been studied and reported.

2 Materials and methods

2.1 Feedstock and spray process

Cobalt-based composite powder of CoCrAlY (23Cr-13Al-0.65Y-balCo) was added with $Al_2O_3 + YSZ$ (28% +2%), CeO₂ (2%), WC–Co (30%), and Cr₃C₂-NiCr (30%) exclusively to prepare coating powders (feedstock). The coating powders were subjected to mechanical mixing to blend properly. The coating powders were developed by using mechanical mixing. $CoCrA_1Y + 28\%Al_2O_3 + 2\%YSZ$, $CoCrA1Y + 2\%CeO₂$, $CoCrA1Y + WC-Co$, and $CoCrA₁Y + Cr₃C₂-NiCr$ coating powders were plasma sprayed over the bond coat. The reinforcement was added in weight percentage. The spraying parameters specifed by the feedstock powder manufacturer were used during plasma spray deposition. METCO USA 3 MB equipment was used to conduct the coating by plasma spray technique. The detailed plasma spray parameters are listed in Table [1.](#page-1-0) Coating particle size was between−45+15 µm.

2.2 Oxidation test

Plasma-sprayed CoCrAlY + Al_2O_3 + YSZ, CoCrAlY + CeO₂, $CoCrA₁W_C$ – Co, and CoCrAlY + Cr₃C₂-NiCr coatings are subjected oxidation at 700 °C in static air for 50 cycles. At the end of each cycle, the specimens were visually examined. Thermogravimetry is a technique for studying the oxidation

Table 1 Plasma spray parameters

| Parameters | Ouantity |
|---------------------------------------|-----------------|
| Flow rate of primary gas (argon) | 40 L/min |
| Flow rate of secondary gas (hydrogen) | 7 L/min |
| Spray distance | $100 - 125$ mm |
| Powder feed rate | 60 g/min |
| Voltage | 60V |
| Current | 490A |

mechanism reaction rate and kinetics. XRD and SEM/EDAX methods were used to identify and investigate the structural properties of reaction products from oxidised specimens.

3 Result and discussion

The cumulative weight increase $(mg/cm²)$ plots of $CoCrA₁Y+A₁,O₃+YSZ$ coatings as a function of time indicated in cycles are shown in Fig. [1.](#page-2-0) Table [2](#page-2-1) shows the total weight increase of the CoCrAlY + Al_2O_3 + YSZ, CoCrAlY + CeO₂, $CoCrA₁Y+WC-C₀$, and $CoCrA₁Y+Cr₃C₂-NiCr$ coatings on MDN 321 and Superni 76 alloys after 50 cycles of oxidation. To analyse oxidation kinetics, Fig. [1](#page-2-0) presents weight growth square (mg^2/cm^4) data as a function of time. There was little diference in weight increase between the two coated alloys.

The rate of weight increase of the coated alloys was faster in the early oxidation cycles, which was due to the rapid formation of active element oxides. However, oxidation and oxidising species difusion at splat borders and open pores have caused the coating splat boundaries to oxidise. The difusion of the oxidising species along the splat boundaries and open pores is listed in Table [2](#page-2-1) as Kp in 10^{-10} g²cm⁻⁴ s⁻¹ for all coatings on both substrates. The parabolic rate constant value of all the coatings on both substrates is shown in Table [2](#page-2-1) in the form of Kp in 10^{-10} g²cm⁻⁴ s⁻¹.

3.1 Phase analysis

Figure [2](#page-3-0) shows the XRD patterns of oxidised $CoCrA1Y + Al₂O₃ + YSZ-$, $CoCrA1Y + CeO₂$ -, $CoCrA₁ + WC$ –Co-, and $CoCrA₁Y + Cr₃C₂$ -NiCr-coated

Fig. 1 (Weight gain/area)² vs. number of cycles plot of (a) $\text{CoCrAlY} + \text{Al}_2\text{O}_3 + \text{YSZ}$, $$ CoCrAlY+WC–Co, and (**d**) $CoCrAlY + Cr₃C₂-NiCr$ coatings after oxidation

Table 2 Weight gain and parabolic rate constant values of oxidised coatings

alloys at 700 °C. Cr_2O_3 , CoO, CoCr₂O₄, and CoAl₂O₄ are the primary peaks of the oxidised coated alloys. Minor peaks are indexed to Al_2O_3 and AlCo respectively. The spinel oxides found in $CoCrA1Y+WC-Co$ and $CoCrA1Y+Cr_3C_2-NiCr$ coatings are $CoWO₄$ and $NiCr₂O₄$, respectively. Because the base coating composition is the same, the primary phases observed in all coatings are comparable. Furthermore, there were no peaks of elements from substrate alloys in the XRD, indicating that no difusion from the substrate had occurred.

3.2 Coating morphologies and EDS analysis

Figure [3](#page-4-0) a depicts the surface morphology of corroded $CoCrA₁Y + Al₂O₃ + YSZ$ coatings. On the oxidised surface, bigger dark and light grey areas and spherical shapes have emerged. The existence of Co as the predominant element was discovered by EDS analysis of the dark grey area labelled region 'A' in Fig. $3a$, along with nominal O and Cr and traces of Al. The light grey layer labelled area 'B' suggests that O, Cr, and Co are the most abundant elements, with traces of Al. The oxide scale $CoCrA_1Y + CeO_2$ (Fig. [3b\)](#page-4-0) is made up of compact granules with peculiar dark grey patches over the oxidised coating surface. EDS examination of the dark grey patches labelled region 'A' reveals the presence of dominant Co and Cr elements with insignifcant levels of Al, Ce, and O. The existence of O and Co as primary constituents, with little Cr concentration, was discovered in the compact granular structure labelled area 'B'.

Similar to $CoCrA_1Y + Al_2O_3 + YSZ$ coating, the oxide scale on CoCrAlY+WC–Co indicates the presence of globular structure (Fig. [3c\)](#page-4-0). Figure [3](#page-4-0) c also shows microporous (sponge) structures with superfcial fssures. The porous structure recognised the presence of W-rich oxides, which might be tungsten-trioxide (WO_3) , and cracks were discovered in this defective oxide region. The surface oxide scale of the CoCrAlY + Cr_3C_2 -NiCr coating (Fig. [3d\)](#page-4-0) is characterised by a compact packed globular shape with patches of distributed peculiar dark grey areas over the oxidised coating surface. Globular structure is a key component of O, Cr, and Co, indicating the presence of Cr and Co oxides. Co and Ni with Al and O make up grey patches, which might suggest the existence of oxides of Co and Ni.

Figure [4](#page-4-1) shows a SEM picture of the cross section of oxidised coatings after 50 cycles. Figure [4](#page-4-1) b shows that all of the remaining coatings have compact, adherent oxide scale on the top surface; however, the $CoCrAlY + CeO₂$ coating has fssures and delamination of oxide. The EDS analysis at diferent places was indicated by the markings 1, 2, 3, and 4 on the cross section picture, and the corresponding elemental weight % is noted in Table [3](#page-5-0).

3.3 Comparative discussion

In Fig. [5,](#page-5-1) the cumulative weight gain $(mg/cm²)$ of all oxidised coatings is indicated. When compared to the other coatings, $CoCrA_1Y + Al_2O_3 + YSZ$ -coated alloys had signifcantly smaller weight gains. During the oxidation cycles,

Fig. 2 XRD patterns for (**a**) 1 CoO, 2 Cr₂O₂, 3 αAl₂O₃, 4 AlCo, 1 CoO, 2 Cr₂O₃, 3 α Al₂O₃, $CoCrAlY+Al₂O₃+YSZ-$ 5 $CoCr₂O₄$, 6 $CoAl₂O₄$ 4 CeO₂, 5 CoAl₂O₄, 6 CoCr₂O₄ **,** $**(c)**$ CoCrAlY+WC–Co-, and (**d**) $CoCrAlY + Cr₃C₂$ -NiCr-coated (a) (b) alloys subjected to oxidation ntensity ntensity Coated Superni 76 Coated Superni 76 Coated MDN321 Coated MDN321 $\dot{36}$ 40° 5^c 60 70 $\overline{80}$ $\dot{90}$ 30 40 50 60 70 $\dot{80}$ 90 1 CoO, 2 Cr₂O₃, 3 α Al₂O₃, 4 WO₃ 1 CoO, 2 Cr₂O₃, 3 αAl₂O₃, $5 \text{ CoCr}_2\text{O}_4$, 6 CoWO 4 Al_sCo₂, 5 NiCr₂O₄, 6 CoCr₂O₄ (d) (c) $\begin{array}{c} 6 \\ 5 \end{array}$ ntensity ntensity Coated Superni 76 Coating on Superni 76 Coated MDN321 Coating on MDN321 30 40 50 70 80 90 $\dot{30}$ 40 50 60 $\dot{70}$ 80 90 60 20 2θ

Fig. 3 Surface morphology of oxidised (a) $CoCrAlY + Al₂O₃+YSZ-$, (b) $CoCrAlY + CeO₂$, (c) $CoCrAlY + WC-Co$ -, and (d) $CoCrA1Y + Cr_3C_2-NiCr-coated$ alloys subjected to oxidation

Fig. 4 Oxidised coating cross section of (**a**) $CoCrAlY+Al₂O₃+YSZ-$ (**b**) CoCrAlY + CeO₂-, (c) CoCrAlY+WC–Co-, and (**d**) $CoCrAlY + Cr₃C₂ - NiCr-coated$ alloys subjected to oxidation

Table 3 EDS analysis of oxidised coatings cross section

the parabolic weight of all of the coatings increased. Based on the data available from these cyclic oxidation trials, the sequential relative-oxidation resistance of all coatings is given in the order below:

 $CoCrAlY + Al₂O₃ + YSZ > CoCrAlY + Cr₃C₂-NiCr > Co$ $CrA₁W_C - Co > CoCrA₁Y + CeO₂.$

On the surface morphology of oxidised CoCrAlY + Al₂O₃ + YSZ coating, a continuous globular with dense in structure can be seen (Fig. [4](#page-4-1)). Figure [4](#page-4-1) a shows a thin, thick, non-porous, and well adhering oxide scale on the coating surface in a SEM image of a cross section of oxidised coating. Furthermore, the oxide scale is free of cracks. Crack-free, thin, dense, non-porous, and well adherent oxide scale are the most common topographies of protective oxides on the coated surface [\[21](#page-7-14)[–23\]](#page-7-15).

The oxygen enters minute gaps in coatings such as pores, splat boundaries, and rare voids, during the frst phases of oxidation until all the accessible interior surfaces are completely oxidised. Later, the active components of the coatings Al and Cr are partly oxidised, reducing the coating's availability to supplemental oxygen. Throughout the oxidation, the distribution of thermodynamically stable AI_2O_3 in the $CoCrA₁Y+A₁,₀Y+A₂YA₃$ roading underwent delayed oxide scale development kinetics $[16–19]$ $[16–19]$ $[16–19]$. One of the explanations for the lesser weight growth might be because the reinforcements Al_2O_3 YSZ are in a pre-oxidised phase and will not oxidise further. As a result, oxidation occurs solely on the outer surface of the coating.

The coating's basic element, Co, is oxidised to CoO, followed by Cr, which is oxidised to Cr_2O_3 . With respect to $CoCrA₁ + A₁, O₃ + YSZ$ coating, the oxidation of two distinct oxides leads in the creation of $CoAl₂O₄$ and $CoCr₂O₄$ spinels, which inhibits oxide development and results in a smaller

Fig. 5 Total weight gain (mg/ cm^2) of uncoated and plasmacoated alloys subjected to oxidationWhen comparing the weight gain of the same coating on two distinct alloys, the coatings on two substrates with the larger total weight gain are evaluated. $CoCrAlY + Al₂O₃ + YSZ$ coatings gained roughly 28% less weight than $CoCrAlY + Cr₃C₂-NiCr$ coatings, 34% less weight than CoCrAlY+WC–Co coatings, and 44% less weight than $CoCrAlY + CeO₂ coatings.$ After oxidation, the main phases visible at the surface of coatings on two diferent alloys are identical

oxide scale thickness. Internal oxidation occurs in open pores during the earliest phases of oxidation for thermal sprayed metallic coatings, according to some research publications [\[20](#page-7-13)[–22](#page-7-16)], corroborating the fndings of our study. The major phases identifed by XRD examination of an oxidised coated surface are Cr_2O_3 , $CoCr_2O_4$, and $CoAl_2O_4$, which are thermodynamically stable and have a dense-packed globular shape.

 $CoCrAlY + Cr₃C₂$ -NiCr coating, on the other hand, comprises 36 (wt%) Cr, including Cr contained in Cr_3C_2-NiCr reinforcement. Because of the sophisticated Cr element in the coating, Cr is quickly transformed into Cr_2O_3 at the start. The presence of Cr_2O_3 as a dominant phase may be seen in the XRD results. Cr_2O_3 is a stable oxide that prevents oxidation and corrosion by limiting oxygen transport inward below 800 °C. Cr₂O₃ will change to CrO₃ at this temperature, which is a volatile oxide that is not suited for oxidation protection at higher temperatures [[19\]](#page-7-12). The spinel oxides $CoCr₂O₄$ and $NiCr₂O₄$ are the predominant and minor phases in XRD analysis, respectively. $CoCr_2O_4$ and $NiCr_2O_4$ spinel oxides are formed via the nucleation of CoO-Cr₂O₃ and NiO-Cr₂O₃.

The active coating components namely Co, Cr, Al, and W have been oxidised to form CoO, Cr_2O_3 , Al_2O_3 , and WO₃ with respect to $CoCrA₁Y + WC-Co$ coating. $CoWO₄$ as a strong phase with Cr_2O_3 and $CoCr_2O_4$ creates a thick, nonporous oxide scale with future oxidation cycles, providing resistance to oxidation. Similarly, during oxidation tests of W-based coatings, several researchers [[23](#page-7-15)[–25\]](#page-7-17) found the occurrence of Cr_2O_3 , CoWO₄, and CoCr₂O₄ as a strong phase. WO_3 is viewed as a porous structure that allows oxygen to diffuse inward until it converts into $CoWO_4$.

The oxidation of active components Al, Co, and Cr results in the production of oxide scale on the coated surface in the case of $CoCrA₁Y + CeO₂$ coating. Ce ions restrict outward diffusion of Co due to the segregation of $CeO₂$ at the grain boundaries of the oxide scale, and oxide scale growth is mainly controlled by inward diffusion of oxygen. $CeO₂$ segregation, on the other hand, occurs slowly, resulting in greater weight gain and cracks.

4 Conclusion

• CoCrAlY + Al_2O_3 + YSZ coatings have higher oxidation resistance than other coatings. Based on thermogravimetric data, the relative oxidation resistance of the various coatings is arranged in the following order.

 $CoCrAlY + Al_2O_3 + YSZ > CoCrAlY + Cr_3C_2 - NiCr >$ $CoCrAlY + WC - Co > CoCrAlY + CeO₂$

• Slow-scale growth kinetics during oxidation was caused by an oxide layer of Al_2O_3 , Cr_2O_3 , and $CoCr_2O_4$ covering the outermost surface of $CoCrAlY + Al₂O₃ + YSZ$ coatings.

By selectively oxidising Al and Cr at the cobalt-rich splat boundary, air cannot penetrate the coating through pores or voids, keeping the oxidation rate constant.

- Because the CoCrAlY + Cr₃C₂-NiCr coating has a greater Cr concentration than the other coatings, Cr_2O_3 was formed quickly during the initial phases of oxidation. Cr_2O_3 with spinal oxides of $CoCr_2O_4$ and NiCr₂O₄ may hinder the inward passage of oxygen, lowering the rate of oxidation.
- The increased weight gain of CoCrAlY + WC–Co coating was due to the quicker oxidation of W into WO_3 , which appears as a porous structure that enables oxygen to permeate until it transforms into CoWO4. As a result of several oxidation cycles, thick, non-porous oxide scale made of Cr_2O_3 , CoO, and Al_2O_3 was produced, which ofered oxidation resistance.

Declarations

Conflict of interest The authors declare no competing interests.

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