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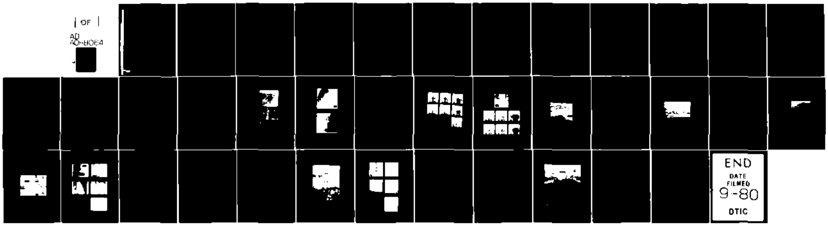

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DIP PROCESS THERMAL-BARRIER COATINGS FOR SUPERALLOYS

LEVEL II

Final Report

June 1980

By: I. M. Allam, R. W. Bartlett and G. N. Krishnan

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A new technique of developing thermal-barrier coatings for superalloys has been investigated. It involves hot dipping of superalloy samples into a molten bath of low melting Ce-Co or Ce-Ni alloy. By internally oxidizing these coatings, it is possible to obtain a duplex with a CeO_2-rich oxide scale as the outer layer (thermal barrier) and an inner layer composite of fine cerium oxide particles dispersed in a matrix of Ni(Co)CrAl. This inner layer, or subscale, grades into the superalloy. Appropriate heat treatment can produce recrystallized CeO_2 particles

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ABSTRACT

A new technique of developing thermal-barrier coatings for superalloys has been investigated. It involves hot dipping of superalloy samples into a molten bath of low melting Ce-Co or Ce-Ni alloy. By internally oxidizing these coatings, it is possible to obtain a duplex with a CeO_2-rich oxide scale as the outer layer (thermal barrier) and an inner layer composite of fine cerium oxide particles dispersed in a matrix of Ni(Co)CrAl. This inner layer, or subscale, grades into the superalloy. Appropriate heat treatment can produce recrystallized CeO_2 particles in the subscale that should improve adherence of the oxide scale to the subscale and substrate superalloy. The coating process does involve limited reaction between the coating alloy and superalloy. The thickness of this interaction zone can be controlled and compensated for in the design (cross section) of the superalloy component.

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INTRODUCTION

Coatings that offer improved resistance to oxidation and hot corrosion have been developed for superalloys used as hot gas turbine components. The coatings have featured CoCrAlY and NiCrAlY films deposited by electron beam evaporation or sputtering. More recently, these metallic materials have been used as a base layer for a duplex coating involving overlay of an oxide as a thermal barrier (insulation). Some problems have developed in adhesion of the thermal barrier to the metal base. In principle, plasma spray techniques involving two guns can be used to apply a graded MCrAlY to refractory oxide coating. Yttria-stabilized cubic zirconia has been used for the oxide because of its high thermal expansion coefficient, which corresponds more closely to the high thermal expansion coefficients of the alloy substrates.

Electron beam vaporization is expensive, limited to line of sight, and tends to lead to variations in coating compositions because of differences in the vapor pressures of the coating constituent elements. Sputtering is expensive and also limited to line of sight. Plasma spraying techniques can lead to variable coating thickness, bare spots, pinholes, and microcracks, all of which can contribute to catastrophic failure. Also, plasma spraying often cannot be used on reentrant surfaces such as those on jet engine combustor liners.

Objecitves

This project concerns an alternative process to develop a duplex oxide thermal-barrier coating of the CoCrAl type for superalloys that would ultimately be: (1) easier and less expensive to apply than the present processes, (2) applicable to reentrant surfaces and to both large and small components, and (3) equal or superior to present thermal-barrier coating systems in thermal oxidation protection, particularly under extended thermal cycling, where adhesion of the thermal-barrier oxide has been a problem.

Methodology

The SRI coating approach is radically different from contemporary processes for providing thermal-barrier CoCrAlY coatings. It was stimulated by earlier findings[1,2] of one of the authors of this report during a study of the rapid selective internal oxidation of rare earth cobalt alloys at relatively modest temperatures.

Our approach is based on low-melting alloys, rich in cerium (rare earth) and containing minor amounts, in descending order, of cobalt, chromium, and aluminum. These alloys can be fused onto superalloys either by dipping the superalloy into the coating melt or by using a coating alloy powder slurried with an organic vehicle, then fired in an inert atmosphere. Our original concept was to selectively oxidize cerium, using CO/CO_2 gas mixtures, to provide a CeO_2-rich outer scale, useful as the thermal-barrier oxide, and an inner subscale containing dispersed internal CeO_2 in an alloy matrix of the other elements, primarily Co, Cr, and Al. CeO_2 matches the underlying alloy thermal expansion coefficient better than does stabilized ZrO_2. If necessary, a graded alloy coating was to be obtained by multiple layers of alloy, each successively richer in cerium. Once the cerium is oxidized, the remaining alloy is both refractory (high melting point) and resistant to oxidation.

As described in the next section, the coating process has been developed during the past year generally along this plan but with some differences. Most importantly, the goals of the modest one-year effort have been met for a cerium-rich dip coating on superalloy IN738. Excellent results have been obtained by using a 90 wt% cerium alloy, which is richer than planned but has a very low melting temperature. A film of this alloy is formed on IN738 by briefly dipping it into the Ce alloy melt. By an inert atmosphere heat treatment the resulting film is allowed to react further with the superalloy substrate to produce a cerium-rich outer layer and an interaction zone containing at least two conjugate layers. Subsequent selective oxidation of cerium in CO/CO_2

mixtures has produced an oxide scale (thermal barrier) very rich in CeO_2 and an inner layer alloy (coating) rich in Ni, Cr, and Al, but also containing Co and dispersed CeO_2. Because the interface between this interaction zone and the oxide scale is highly irregular, it is expected to promote good scale adhesion by mechanically anchoring the oxide scale to the underlying substrate. Dispersed CeO_2 particles in the interaction zone also act as pegs to anchor the outer oxide scale.

EXPERIMENTAL PROCEDURES

Two superalloys were investigated: nickel-base IN738 and cobalt-base MAR-M509. Standard test wedge bars were coated with cerium-rich low melting alloys by two techniques: hot melt dipping and fused slurry brazing.

Table 1 shows the composition of the seven cerium-rich alloy coatings that were studied. The alloys were prepared by melting the constituent metals. Initially, cold hearth arc melting was used with remelt of the button. Later, induction melting under argon in a deep molybdenum crucible was used in conjunction with dip melting. After this high-temperature homogenization melt, the alloy was cooled to room temperature and then transferred to the cold furnace used for dip coating without removing the coating alloy from the molybdenum crucible.

Table 1

COMPOSITION OF CERIUM (MISCHMETAL)-RICH COATING ALLOYS
(Weight Percent)

Alloy Number	Ce	Co	Cr	Al	Ni
1	72.1	21.2	5.3	1.4	--
2	50.0	35.0	10.0	5.0	--
3	60.0	40.0	--	--	
4	50.0	--	--	--	50.0
5	90.0	10.0	--	--	--
6	90.0	--	--	--	10.0
7	100.0	--	--	--	--

Powders for slurry coating were prepared by crushing arc-melted alloy buttons, followed by ball milling.

Fused slurry coatings were prepared by painting (usually multiple coats) or dipping the substrate alloy at room temperature into a slurried mixture of the coating alloy. This slurry consisted of:

- Coating alloy
- Organic cement (Nicrobraz 500, Wall Colmonoy Corp.)
- Mineral spirit (MPA-60, Baker Coastor Oil Co.).

The mixture was ground in a ceramic ball mill with Al_2O_3 balls for 5 hours and decanted into a sealable container. After the coated samples were dried in air for $\simeq$ 2 hours, they were fused at 1250°C in an inert atmosphere. Slow-flowing argon was passed over hot calcium chips to getter oxygen.

Dip coating temperatures must exceed the liquidus temperature, which declines with increasing Ce content in Co and Co(Ni) alloys. During the hot-dipping step (process step 1), the liquid bath temperature was held about 50-150°C higher than the liquidus temperature of the coating alloy used. The dip temperatures generally used are shown in Table 2. The alloy film was formed on the superalloy substrate by slowly lowering the test wedge bar, using a vertical push rod passing through a Cajon ultra-torr fitting. The specimens were submerged in the melt, typically for 1 minute, and then withdrawn. The dip coating furnace apparatus is shown in Figure 1.

Table 2

EXPERIMENTAL DIP COATING TEMPERATURES

Alloy Number	Temperature
1, 2, 3, and 4	1150°C
5 and 6	600°C
7	850°C

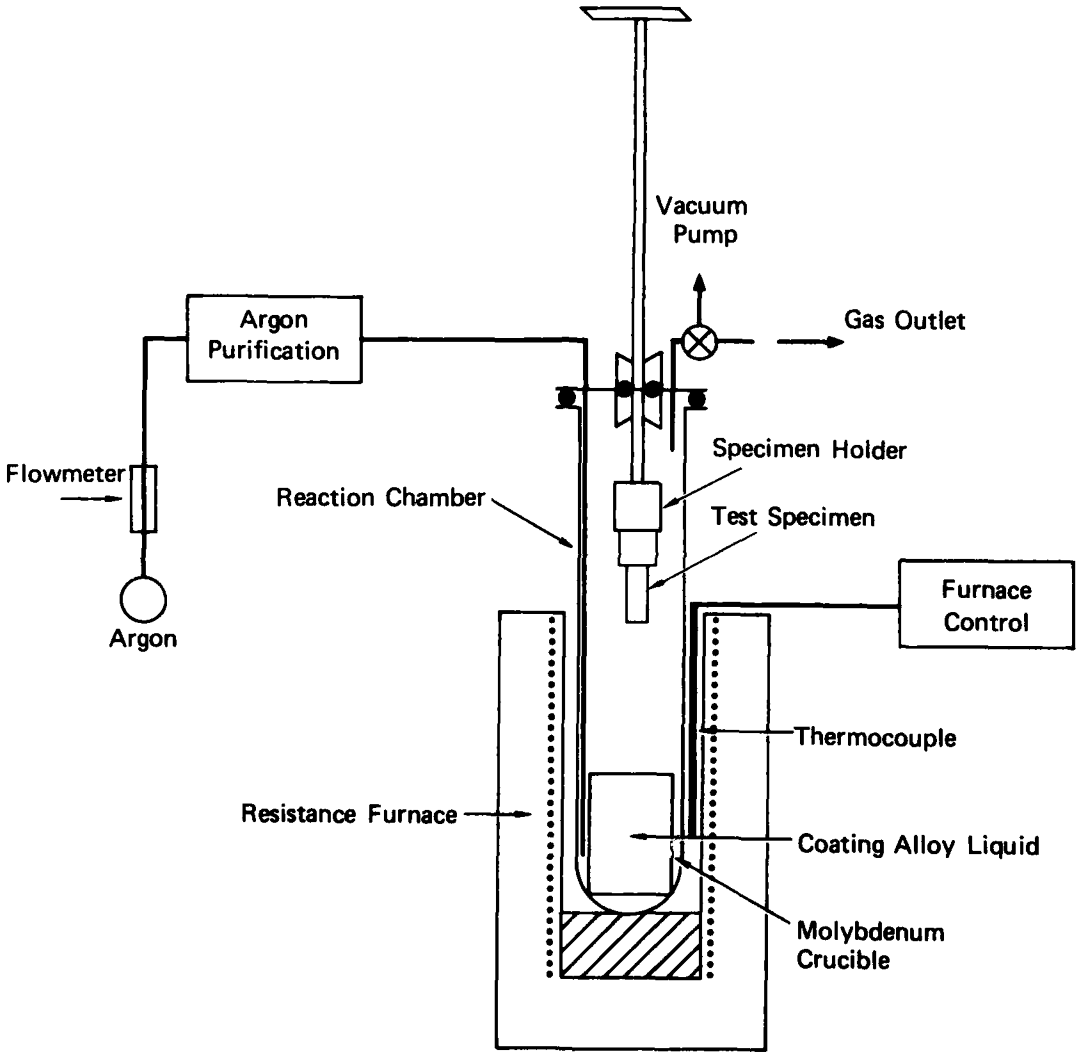

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FIGURE 1 SCHEMATIC DIAGRAM OF HOT DIPPING APPARATUS

Dip-coated specimens were usually held at furnace temperature for periods longer than 1 minute. This permitted formation of an extensive, conjugate interaction zone caused by a reaction between the coating and substrate alloys. This furnace annealing constituted the unplanned second step in what became a four-step process.

In the third step, coated specimens were selectively oxidized in 9.67% CO -90.33% CO_2 for periods ranging from 30 minutes to 2 hours at 800°C. The equilibrium oxygen partial pressure was 2.25×10^{-17} atmosphere. Several samples were internally oxidized at 900°C, which corresponds to $P_{O_2} = 7.19 \times 10^{-15}$ atm. The dissociation pressures of CoO calculated at 800° and 900°C were 2.75×10^{-16} atm and 3.59×10^{-14} atm, respectively. The dissociation pressures of NiO are higher than that of CoO at these temperatures; hence, cobalt and nickel were not oxidized. Both oxidation temperatures are below the melting temperatures of alloys 1, 2, 3, and 4, but above that of alloys 5, 6, and 7 (cerium). However, rapid reaction between the coating alloy film and the substrate during dipping or slurry brazing tended to severely diminish the actual cerium concentration so that incipient melting was not a problem.

The fourth step was an anneal in the absence of any oxygen, carried out in a horizontal tube furnace at either 900° or 1000°C for periods up to 2 hours. This was intended to recrystallize oxide grains inside the unoxidized alloy matrix.

EXPERIMENTAL RESULTS

The results are divided into three experimental categories: (1) dip coating with alloys containing less than 75% Ce and usually significant amounts of Cr and Al, (2) slurry fusion coatings of alloys containing less than 75% Ce, and (3) Ce-rich low-melting-point alloy dip coatings with 90 to 100% Ce. By far the best results were obtained with Ce-rich dip coatings, and the earlier two categories are no longer significant.

Dip Coatings ($<$ 75% Ce)

CoCrAl-type coatings require higher concentrations of chromium and aluminum than can be tolerated by superalloys used as blades or nozzle guide vanes. Therefore, we originally intended to maintain moderately high concentrations of both chromium and aluminum in the liquid dip alloy to provide a coating that, after complete oxidation of cerium, is within the composition range for typical CoCrAl protective coatings. The primary evaluation of each coating at this early stage was based on its ability to form an adherent, continuous, and pore-free layer.

Figure 2 shows the microstructure of the base alloy IN738 as coated by hot dipping into coating alloy 2 at 1150°C for 5 minutes. The coating layer produced by this process was nonuniform, and porous and contained a large amount of CeO_2-rich slag, which attached to the specimen during dipping. A thick interaction layer ($\simeq$ 600 μm) formed at the original interface between the coating and the unreacted substrace. This layer is attributed to the partial exchange of elements between the coating and substrate alloys--a process that radically alters the composition of both materials.

Figure 3 is an SEM micrograph of the same substrate IN738 after dipping into coating alloy 1 under the same conditions described above for coating alloy 2. The coating was porous and discontinuous and often

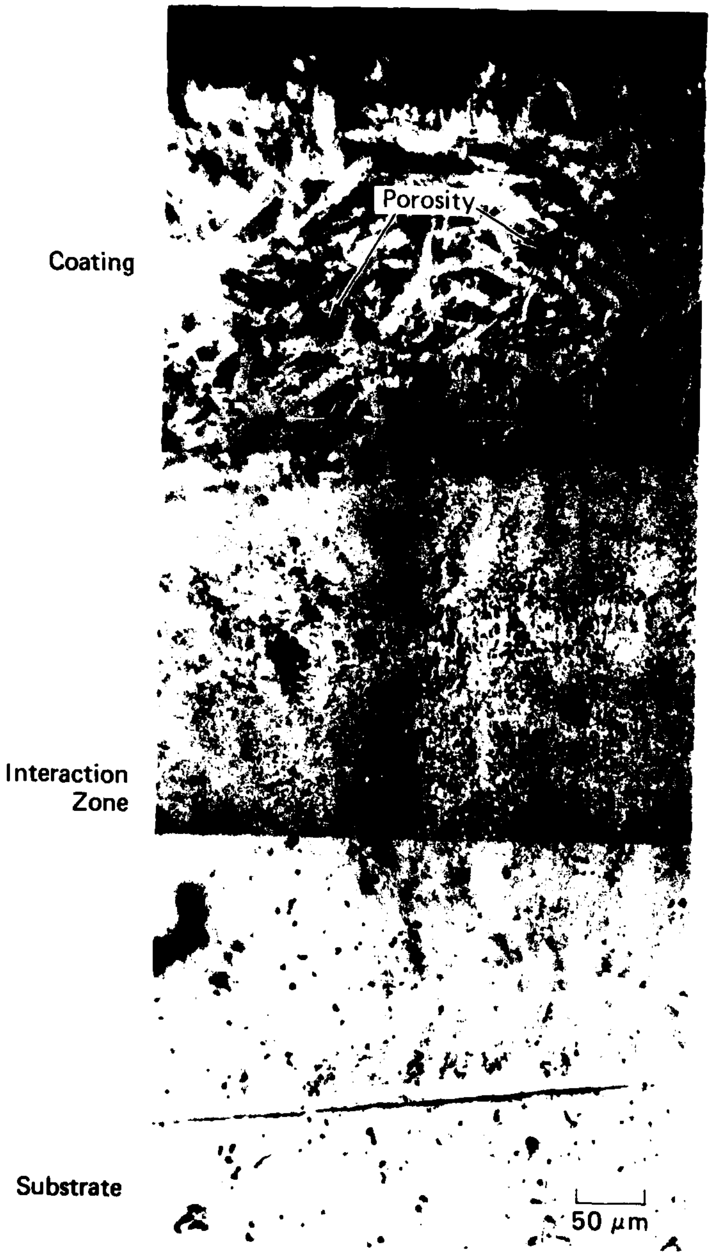

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FIGURE 2 CROSS SECTION OF IN738 COATED BY ALLOY No. 2 (50% Ce-35% Co-10% Cr-5% Al), HOT DIPPED AT 1150°C FOR 5 MINUTES

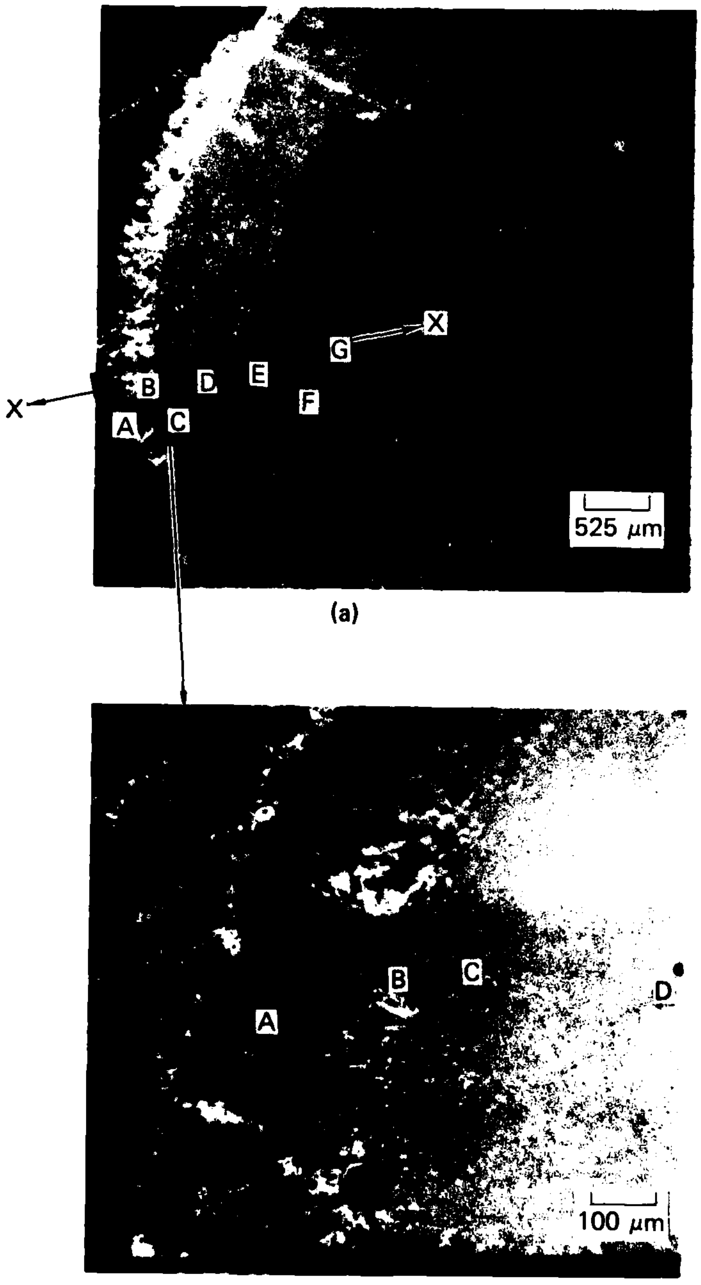

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FIGURE 3 SEM CROSS SECTION MICROGRAPH OF IN738 COATED BY ALLOY No. 1 (72.1% Ce-21.2% Co-5.3% Cr-1.4% Al), HOT DIPPED AT 1150°C FOR 5 MINUTES spalled along the coating/substrate interface during cooling. It appears that the thickness of the interaction zone (1000 μm) for alloy 1 increases as the cerium concentration in the dip alloy increases.

Figure 4 shows the concentration profiles of coating and substrate constituents across line X-X in Figure 3. Cerium partially replaced nickel across the interaction zone [point B to E]. The formation of the interaction zone did not cause changes in the concentrations of chromium or aluminum.

The performance of both coating alloys 1 and 2 was unsatisfactory because of the following factors:

(1) Incomplete coverage of the substrate alloy.

(2) Nonuniformity of the coating layer.

(3) High porosity and loss of adhesion.

(4) The formation of a thick interaction zone, which resulted in narrowing of the effective remaining cross section of the substrate.

(5) Partial oxidation of cerium to a CeO_2 slag on the melt surface; this slag adheres to the specimen. The slagging problem and the excessively thick interaction zone (item 4) were caused by high processing temperatures, which were required to exceed the liquidus temperature of the Ce alloy.

Figure 5 shows an EDAX analysis of IN738 specimen coated by alloy 2, then oxidized for 120 minutes in the CO/CO_2 mixture at 800°C.

The SEM photograph shows a crack that extends through the substrate parallel to its surface. The outer dense layer is mainly CeO_2 containing variable amounts of Al, Co, Ni, and Cr, probably in the form of mixed oxides. The concentration of cobalt drops gradually in the interaction zone with distance away from the gas/oxide interface. Ce appears in significant amounts within this zone. The nickel concentration remains about constant across the interaction zone, but it is below the original concentration in the substrate alloy. Figure 6 shows a cross section of the same sample shown in Figure 5. Extensive delamination and spalling

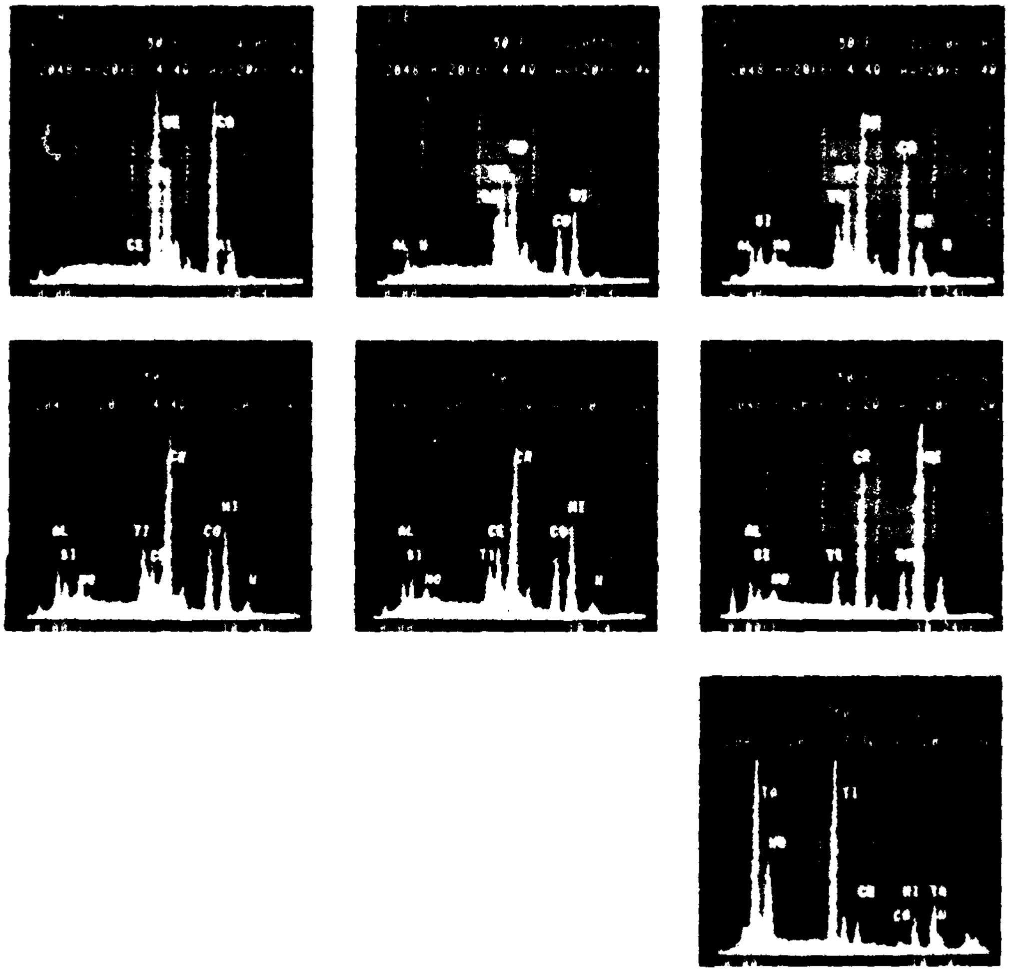

FIGURE 4 EDAX ANALYSIS OF THE COATING AND SUBSTRATE ACROSS LINE X-X SHOWN IN FIGURE 3

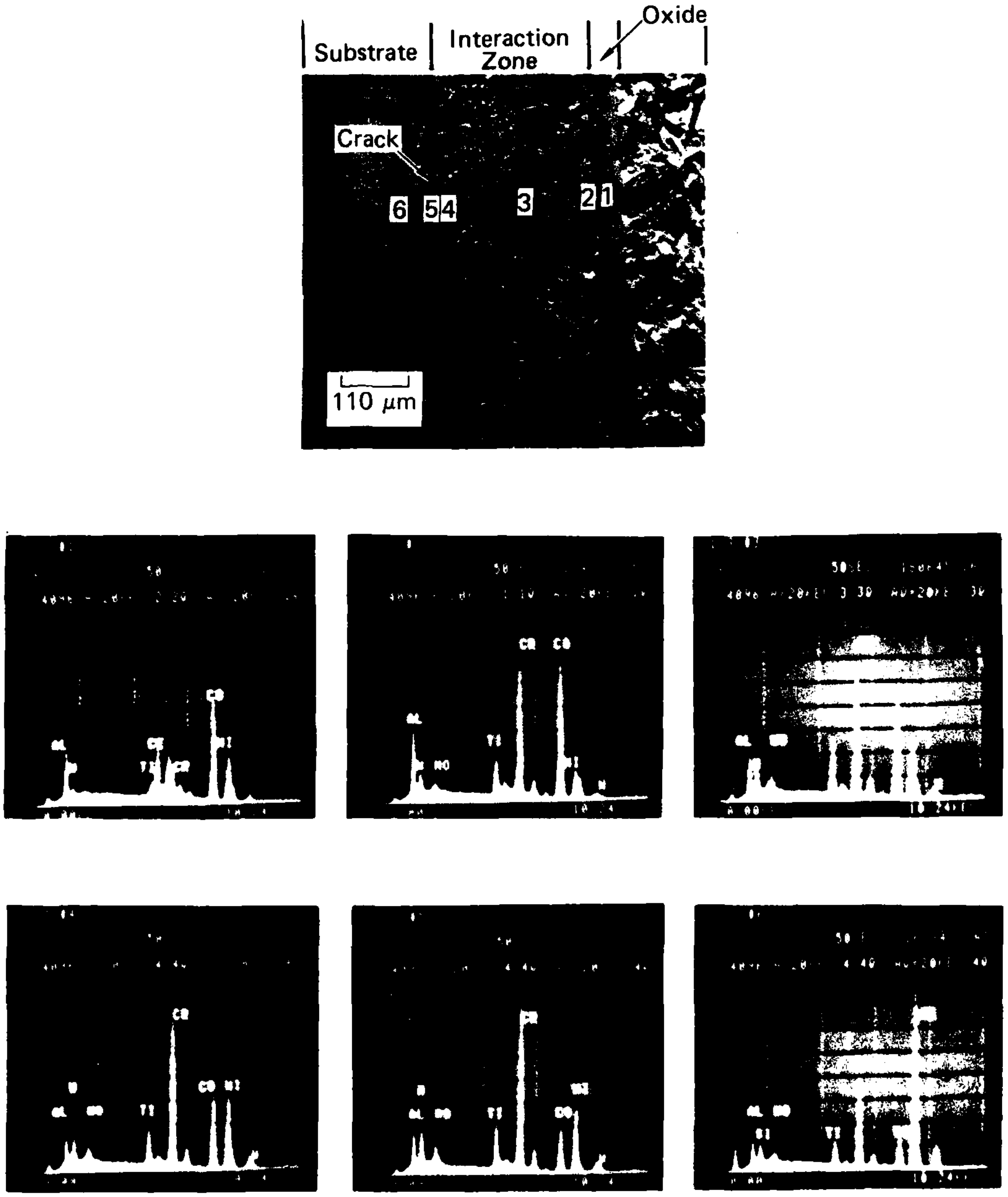

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FIGURE 5 EDAX ANALYSIS OF IN738 COATED BY ALLOY No. 2 (50% Ce-35% Co-10% Cr-5% Al), THEN OXIDIZED IN CO/CO_2 MIXTURE AT 800°C FOR 2 HOURS

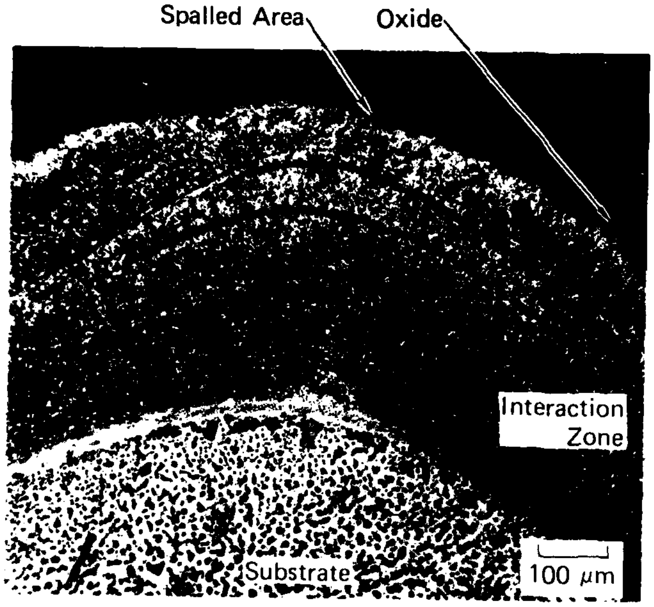

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FIGURE 6 CORNER DELAMINATION OF COATING-INTERACTION ZONE IN SPECIMEN SHOWN IN FIGURE 5 within the interaction zone commonly occur at specimen corners where thermally induced stresses are perpendicular to the relatively weak interfaces.

Hot dipping into coating alloys 3 and 4 did not show any significant improvement over coating alloys 1 and 2. Figure 7 shows the structure of an IN738 specimen coated by alloy 4, then oxidized for 2 hours at 800°C, and finally recrystallized at 1000°C for 2 hours. As in Figure 5, cracks develop parallel to the interaction zone or substrate interface as well as vertical to it, during cooling from recrystallization temperature.

The above results indicate that hot dipping at relatively high temperatures (1150°C) for alloys 2 and 3 is the major cause of coating degradation. This is attributed to:

(1) The excessive formation of CeO_2 slag, resulting in incomplete coverage of substrate by the liquid alloy and the nonuniformity as well as high porosity of the coating layer.

(2) The development of an excessively thick interaction zone and the consequent reduction in the substrate effective cross section.

Slurry Fusion Coatings

In dip coating, an almost infinite source of molten alloy is available to react with the substrate (superalloy). Slurry fusion coatings were developed to limit the amount of coating alloy that could react with the substrate and, thereby, limit the excessive thickness of the interaction zone. However, other problems were encountered and the slurry fusion coating process was eventually discontinued. Extensive oxidation of Ce results from ball milling, causing poor sintering of the coating alloy and a loose, friable coating with poor adhesion to the superalloy substrate.

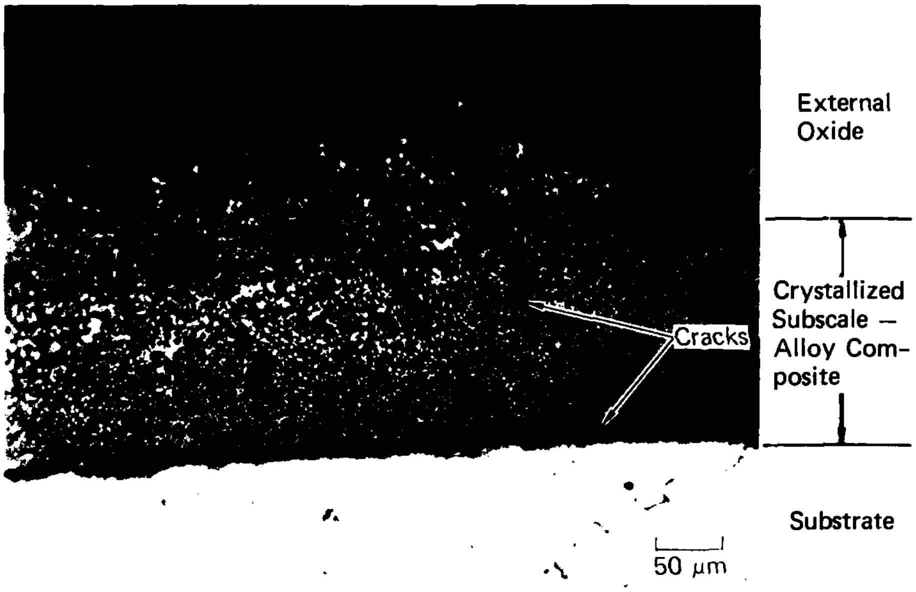

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FIGURE 7 MICROSTRUCTURE OF IN738 COATED BY ALLOY No. 4 (50% Ce-50% Ni), THEN OXIDIZED FOR 2 HOURS AT 800°C, AND RECRYSTALLIZED AT 1000°C FOR 2 HOURS

Ultra-Rich Cerium (≥ 90%) on IN738

The best results were obtained when using low-melting-point coating alloys rich in cerium, that is, with alloy 5 (90% Ce-10% Co, melting temperature ≃ 450°C) and alloy 6 (90% Ce-10% Ni, melting temperature ≃ 480°C).

For all coating alloy compositions, a fairly thick interaction layer forms between the coating alloy and unreacted substrate; as shown in Figure 8. This layer is essentially a partial conversion or substitution of elements in the superalloy substrate in a well-defined band that shows a drastic change in composition from the original superalloy substrate. The thickness of the interaction layer is dependent on the dipping temperature and dipping time, and also on the composition of the coating alloy and substrate alloy. Figure 9 shows SEM micrographs of IN738, coated by a uniform layer of 90% Ce-10% Co alloy. The Ce concentration profile measured at the middle section of the micrograph shows abundant Ce across the interaction zone. Although there is a Ce concentration gradient across the interaction zone, the Ce concentration is nearly uniform. Cerium content drops discontinuously at the unaffected substrate, but increases sharply in the primary coating (right). Unexpectedly, the concentration of Ni in the interaction zone is lower than that in the bulk substrate as shown in Figure 10. Also, there is a high Ni concentration in the primary coating alloy layer, which originally, before dipping, contained no Ni. It appears that a coating alloy film freezes onto the substrate after extracting a considerable amount of Ni from the substrate near its surface.

Figures 11(a) and (b) are the phase diagrams of Ce-Co and Ce-Ni.[3] The compositions of both coating alloys at room temperature are: cerium + Ce_3Co intermetallic for the first system and Ce + Ce_3Ni for the second system. A possible driving force for this reaction is the formation of the intermetallic compound, Ce_3Ni, which exists in both the primary coating layer and the interaction zone. This hypothesis is supported by the formation of a much thicker interaction zone when the same substrate alloy was dipped in 100% Ce, alloy 7, under the same

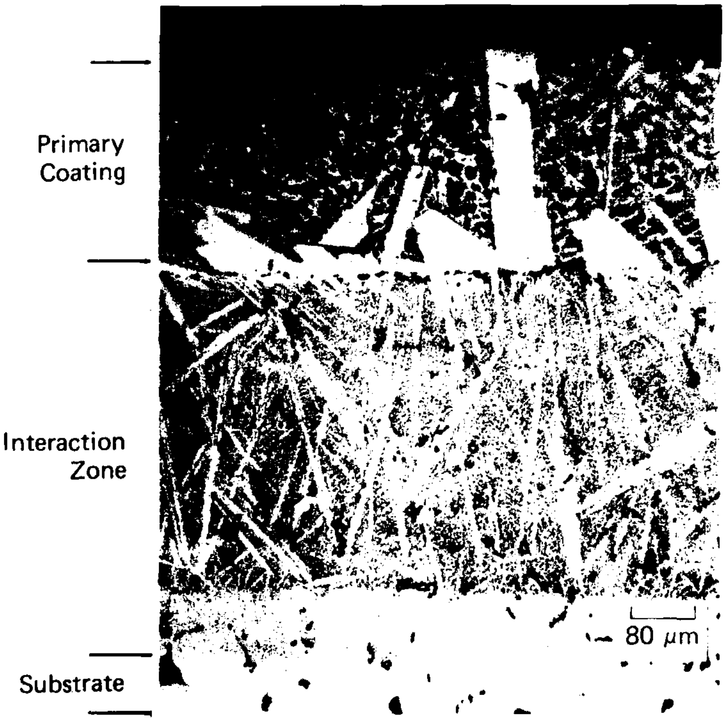

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FIGURE 8 MICROSTRUCTURE OF IN738 COATED BY ALLOY No. 5 (90% Ce-10% Co)

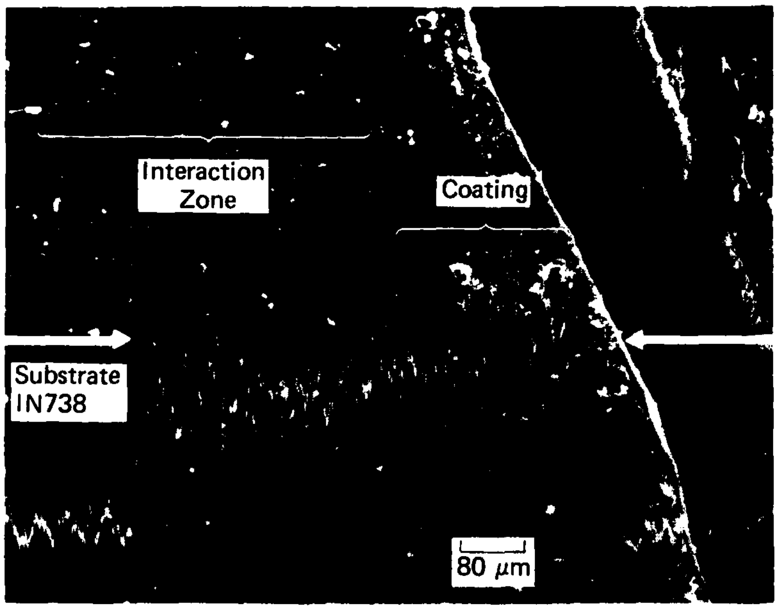

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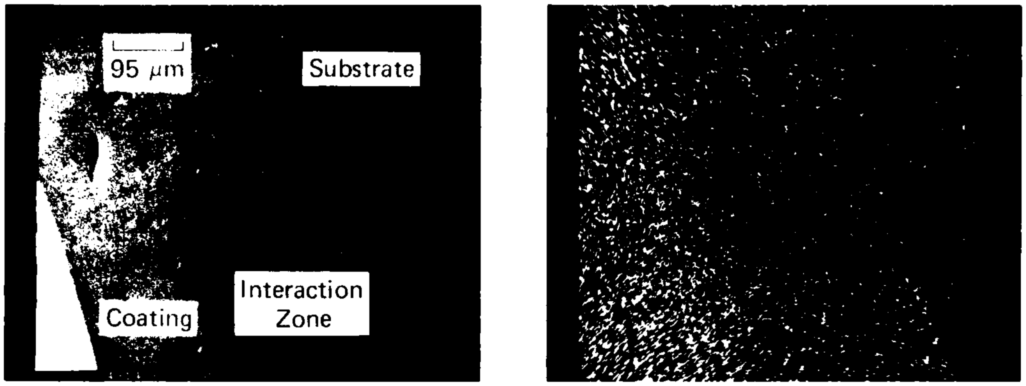
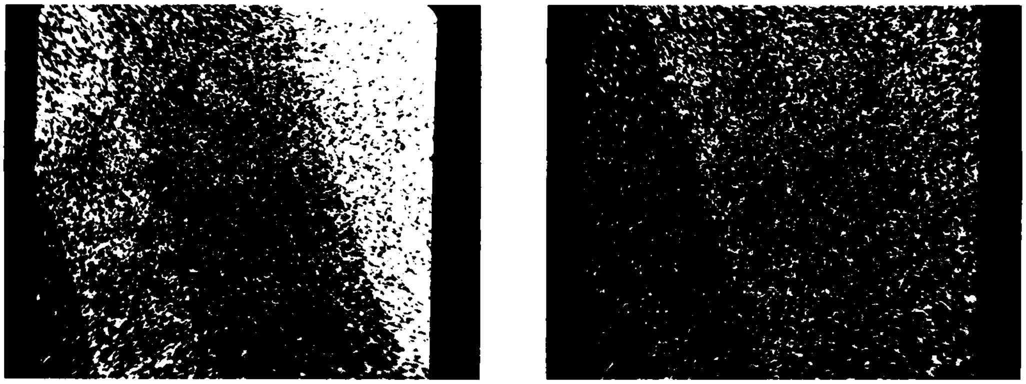
FIGURE 9 CERIUM CONCENTRATION PROFILE ACROSS IN738 SUBSTRATE COATED BY UNIFORM LAYER OF 90% Ce-10% Co



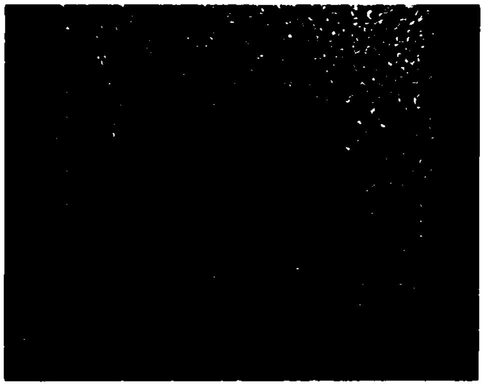
(a) ELECTRON IMAGE

(b) Ce X-RAY



(c) Ni X-RAY

(d) Cr X-RAY



(e) Co X-RAY

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FIGURE 10 X-RAY MAP OF IN738 COATED BY ALLOY No. 5 (90% Ce-10% Co)

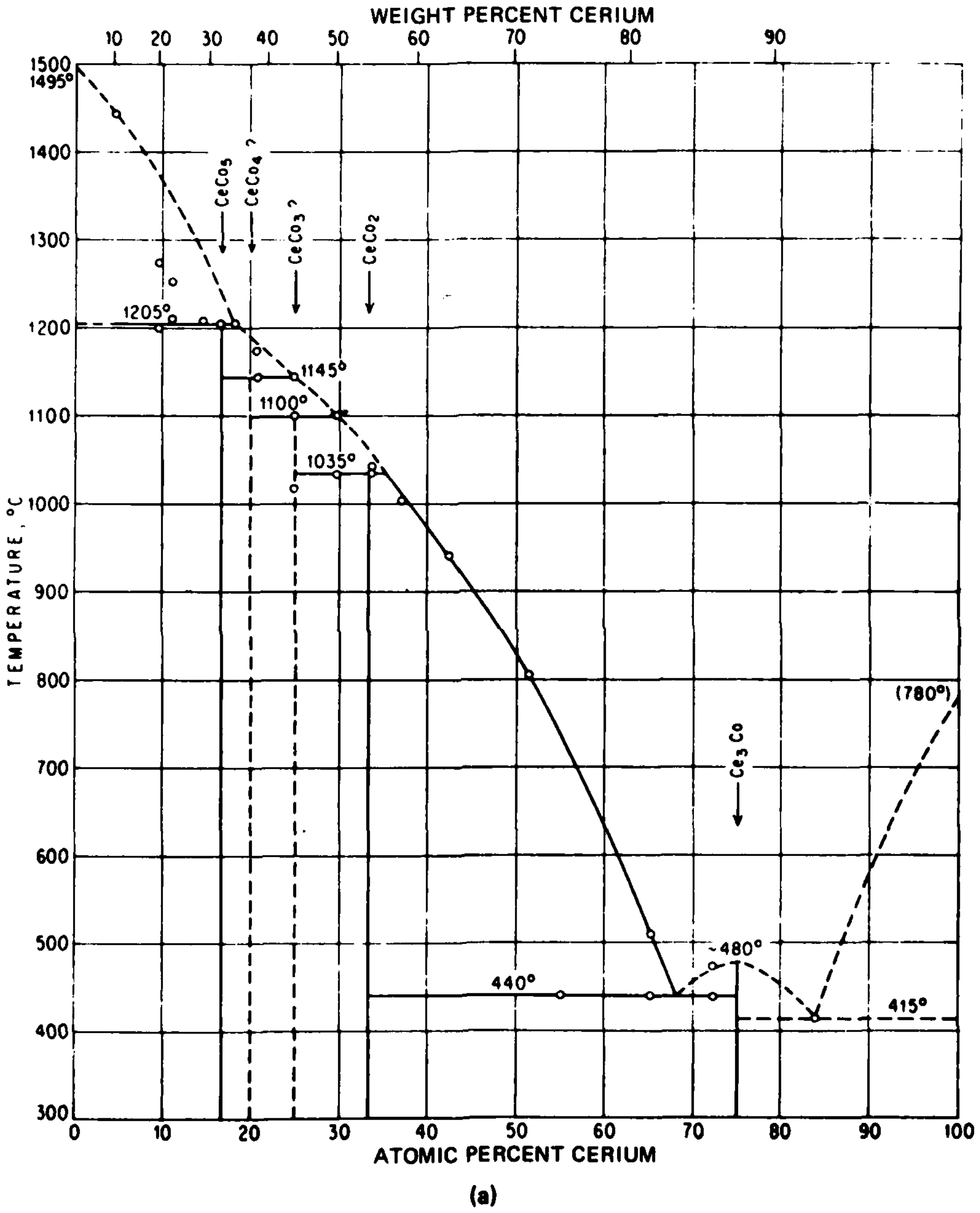

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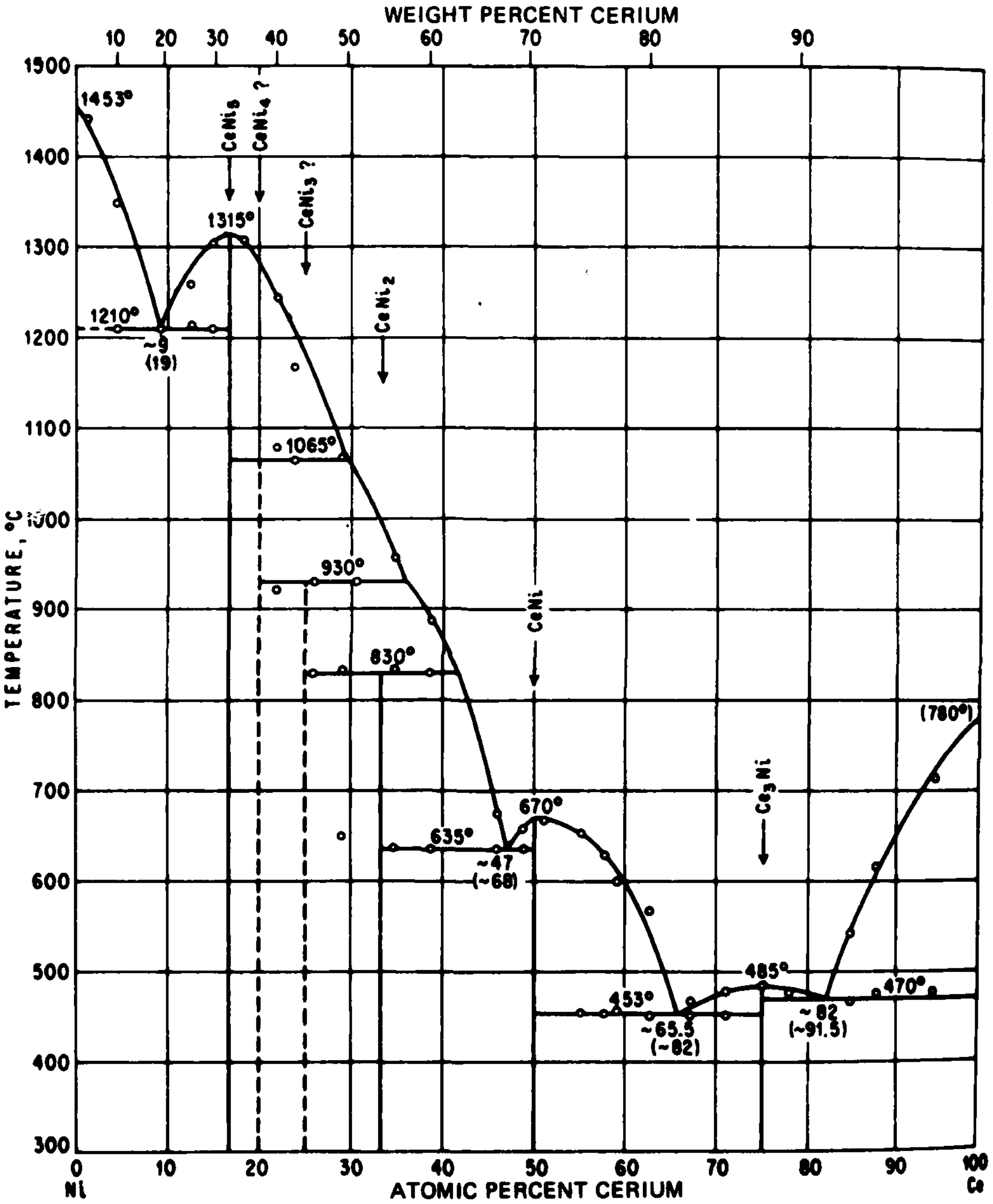
FIGURE 11 PHASE DIAGRAMS OF Ce-Co (a) AND Ce-Ni (b)


WEIGHT PERCENT CERIUM
10
20
30
40
50
60
70
80
90
1500
1400
1300
1200
1100
1000
900
800
700
600
500
400
300
TEMPERATURE, °C
1453°
1315°
1210°
~9
(19)
1065°
930°
830°
635°
~47
(~68)
670°
453°
~65.5
(~82)
485°
~82
(~91.5)
470°
(780°)
CeNi5
CeNi4 ?
CeNi3 ?
CeNi2
CeNi
Ce3Ni
0
Ni
10
20
30
40
50
60
70
80
90
100
Ce
ATOMIC PERCENT CERIUM

(b)

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processing conditions. In this case, during coating, large amounts of cerium react with Ni to form Ce_3Ni, whereas with 90% Ce-10% Co, part of the cerium is tied to Co, to form the intermetallic compound Ce_3Co. Exact compositional in situ analysis of both the coating and the interaction zone remains to be carried out.

The behavior of Co-base (MAR-M509) alloy was fairly similar to that of the Ni-base IN738 explained above. However, some minor differences in the microstructure and composition of the coating and the underlying interaction zone were observed. Further investigation is required to study their compositions and the influence on the subsequent internal oxidation process.

As mentioned earlier, the thickness of the interaction zone increases with the time and temperature of the dipping process. Hence, it was desirable to lower both these parameters to reduce the thickness of the interaction zone and consequently increase the effective cross section of the unaffected superalloy substrate. This was attempted by dipping the substrate into the lowest m.p. alloys (alloys 5 and 6) for different periods. The shortest dipping time, $\simeq$ 5 seconds, resulted in incomplete wetting and coverage of the substrate by the coating alloy. A period of 1 minute was found to be optimum and the thickness of the interaction zone for this period was 300-350 μm, as shown in Figure 8.

Coating alloys 5 and 6 have been exposed to the internal oxidation and crystallization procedures (process steps 3 and 4). The other alloys were not investigated extensively because of the unsatisfactory results described earlier during the hot dipping step.

Figure 12 is a typical section of an IN738 specimen coated by alloy 5, then oxidized in CO/CO_2 at 800°C for 90 minutes. The oxidation temperature was much higher than the m.p. of the initial coating alloy (450°C), but because of prior interaction no processing difficulties were encountered. The SEM-EDAX analysis in Figure 13 shows high Ce concentration in both the outer oxide scale and the underlying internal oxide (subscale) network. The outer layer consists mainly of CeO_2,

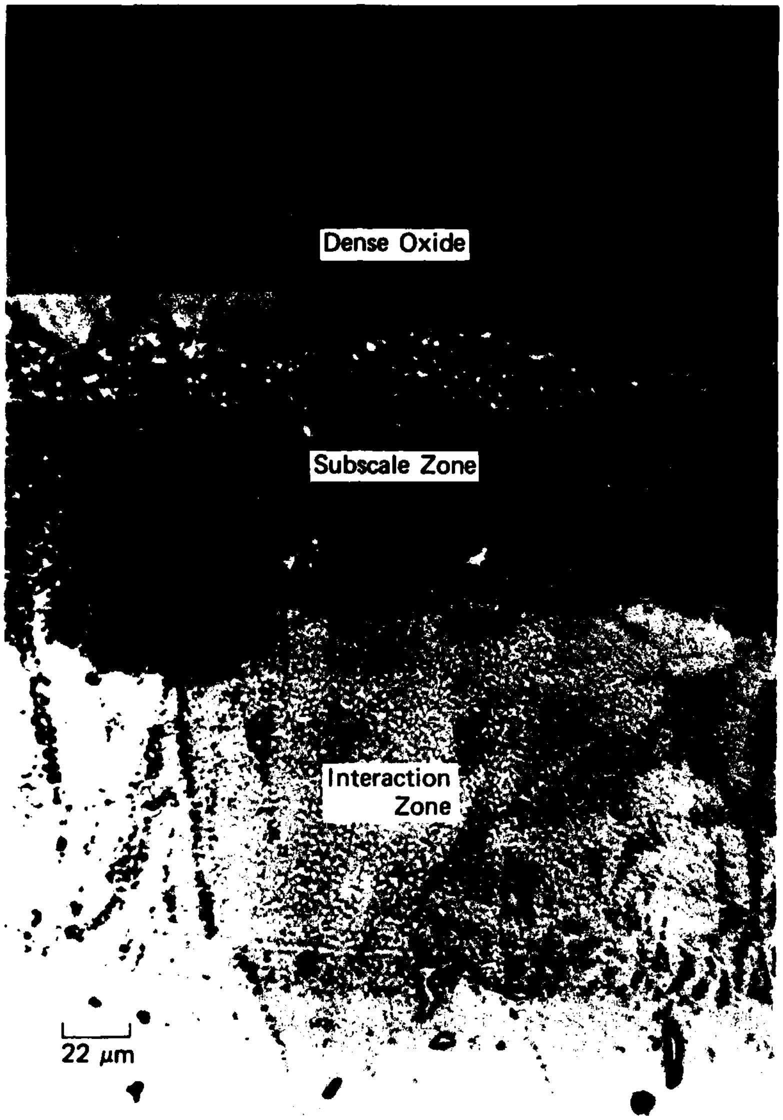

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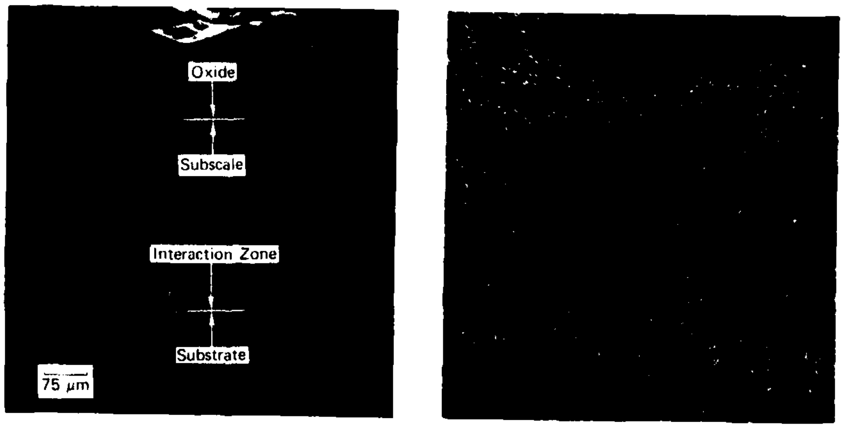
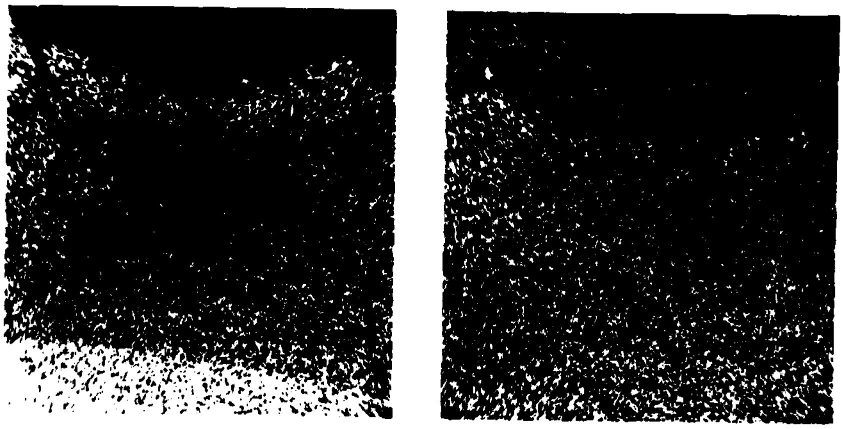
FIGURE 12 TYPICAL CROSS SECTION OF AN IN738 SPECIMEN COATED BY ALLOY (5) (90% Ce-10% Co), THEN OXIDIZED IN CO/CO_2 AT 800°C FOR 90 MINUTES



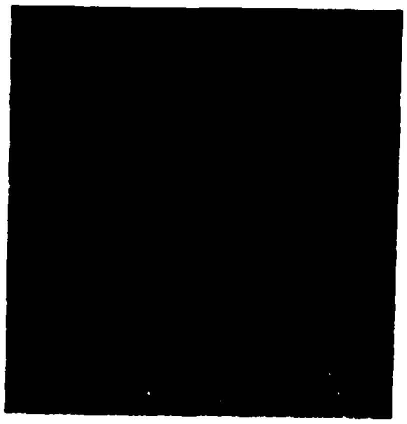
(a) SECONDARY ELECTRON IMAGE

(b) Ce X-RAY



(c) Ni X-RAY

(d) Cr X-RAY



(e) Co X-RAY

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FIGURE 13 X-RAY ANALYSIS OF DENSE SPECIMEN IN FIGURE 12 which contains small concentrations of Ni, Cr, and Al, probably diffused from the substrate alloy. Although the outer layer is fairly incoherent and contains numerous cracks, the cracks would not necessarily impede the performance of the oxide subscale as a thermal barrier. The internal oxide subscale, which extends into the interaction zone, consists of a network of oxide in a metallic matrix. After the internal oxidation treatment, it was difficult to identify the original interface between the primary coating and the interaction zone. The high Ni concentration in the interaction zone ahead of the ingressing external oxide/subscale interface is probably due to Ni accumulation as the P_{O_2} was too low for NiO formation. The subscale layer adherence to the substrate is very good because of the intermixing and uniform blending of the internal oxide particles and the substrate matrix. The oxide scale seems to be very adherent to the subscale layer, despite the cracks.

It was desired originally to convert all Ce existing within the interaction zone to CeO_2, but complete internal oxidation of Ce has not yet been achieved. This may be attributed to either the relatively low temperature internal oxidation treatment (although it is higher than the m.p. of the coating alloy), or to an insufficient time of internal oxidation.

Further oxidation of the remaining cerium in the interaction zone would increase the thickness of subscale layer and would enrich Cr and Al in the interaction zone to depletion of Ce. This Cr and Al enrichment in the interaction zone should provide better service oxidation resistance. The cerium oxide scale and subscale layers would act as thermal barriers.

The oxide recrystallization thermal treatment, step 4, was intended to transform the submicrometer internal oxide fibers into larger, discontinuous oxide particles of 1 to 10 μm diameter. This effect was partly achieved by heat-treating the internally oxidized specimens at 900°-1000°C for 2 hours. However, dense agglomeration of internal oxide particles in numerous areas, particulatly near the interface between the internal oxidation zone and the interaction zone (see arrow in Figure 14), leads to the formation of semicontinuous layer, which may affect the adhesion to the interaction zone adversely. This has not been investigated. Nevertheless, by careful control of the oxide recrystallization time and temperature, it is possible to establish an optimum size and distribution of these particles. Clearly, the amount of oxide depends on the concentration of cerium in the interaction zone found after the step 2 anneal. This may be managed to some extent by adjusting the coating composition and the controlling parameters in process steps 1 and 2.

In summary, a very promising thermal-barrier MCrAl coating system for IN738 has been initiated and we have achieved our goal of obtaining both conjugate layers and interspersed phases (oxide/alloy). However, we recommend that more research be performed to optimize this coating system and evaluate its performance characteristics.

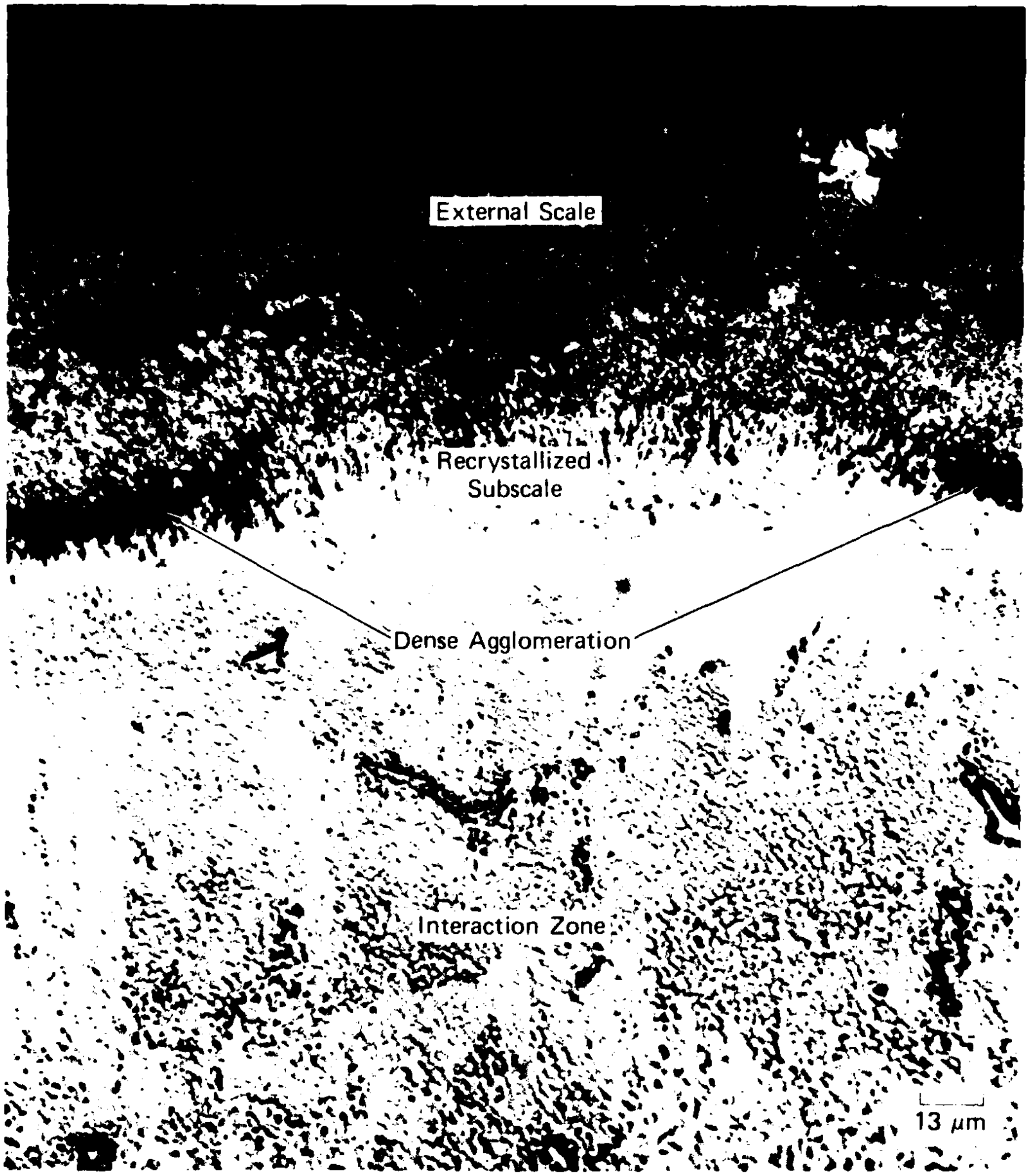

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FIGURE 14 SCALE MICROSTRUCTURE OF IN738 COATED BY ALLOY No. 5 (90% Ce-10% Co), THEN OXIDIZED IN CO/CO$_2$ MIXTURE AT 800°C FOR 45 MINUTES, AND RECRYSTALLIZED AT 900°C FOR 2 HOURS

CONCLUSIONS

(1) Low-melting-temperature rare earth coating alloys ($\simeq$ 90% Ce) provide the best wetting and adhesion to the base superalloys during the hot dipping coating process and also provide a uniform conjugate coating system of moderate thickness with a CeO_2-rich thermal barrier (oxide scale).

(2) An interaction zone rich in cerium develops in the substrate at its original surface as a result of the hot dipping process. The nickel content in this zone is less than originally contained in the superalloy. The formation of this layer does not alter the original chromium and aluminum concentrations in the superalloy; both are critical in resisting high temperature corrosion. However, following internal oxidation of cerium that has substituted for Ni(Co) in the interaction zone, the matrix alloy is effectively enriched in Cr and Al.

(3) Internal oxidation results in the development of a cerium-rich outer scale and a subscale or inner layer that is a mixture of CeO_2 dispersed in the residual Ni(Co)CrAl alloy. These latter layers are highly adherent to the substrate alloy.

(4) Control over the CeO_2 ripening conditions may be required to prevent overly dense agglomeration of the internal oxide particles, since such agglomeration could lead to scale spallation during service thermal cycling.

(5) A continued research program is needed to optimize process conditions developed during this investigation and to evaluate the oxidation and hot corrosion resistance of the new coating systems.

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