THERMAL BARRIER COATINGS ISSUES
IN ADVANCED LAND-BASED GAS TURBINES

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ABSTRACT

The Department of Energy's Advanced Turbine Systems (ATS) program is aimed at fostering the development of a new generation of land-based gas turbine systems with overall efficiencies significantly beyond those of current state-of-the-art machines, as well as greatly increased times between inspection and refurbishment, improved environmental impact, and decreased cost. The proposed duty cycle of ATS machines will emphasize different criteria in the selection of materials for the critical components. In particular, thermal barrier coatings (TBCs) will be an essential feature of the hot gas path components in these machines. In fact, the goals of the ATS will require significant improvements in TBC technology, since these turbines will be totally reliant on TBCs, which will be required to function on critical components such as the first stage vanes and blades for times considerably in excess of those experienced in current applications. Issues that assume increased importance are the mechanical and chemical stability of the ceramic layer and of the metallic bond coat; the thermal expansion characteristics and compliance of the ceramic layer; and the thermal conductivity across the thickness of the ceramic layer. Obviously, the ATS program provides a very challenging opportunity for TBCs, and involves some significant opportunities to extend this technology. A significant TBC development effort is planned in the ATS program which will address these key issues.

INTRODUCTION

The predicted worldwide need for new electricity-generation capacity in the 1990's is 600 GW. In the U. S., energy consumption is expected to increase by 46 percent in the next 10 years or so[1] which suggests a market in the U. S. alone for new base-load electricity generation capacity of 20 GW per year. Approximately 44 percent of the current electricity generating capacity in the U. S. will be more than 40 years old by the year 2010[2] and its replacement will be required to meet stringent emissions standards. The technology required by conventional fossil fuel-fired steam boilers to meet these requirements is ready and available; however, substantial reductions in CO_2 emissions will probably require some fuel switching, since approximately 40% of all the carbon-

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emissions in the U. S. currently are produced by the electric utilities[2]. The projected efficiency of the best available coal-fired steam boiler technology, represented by the Electric Power Research Institute's State-of-the Art Power Plant (SOAPP)[3], is 42 percent [8,100 Btu/kWh net at full load, based on the higher heating value of the fuel (HHV)] when an advanced supercritical steam cycle is employed, which compares with approximately 38 percent for the best coal-fired plants currently in operation in the U. S.

Gas-fired gas turbine combined cycle systems are expected to account for a significant fraction of the projected new capacity. Such plants are available in a range of sizes, up to more than 200 MWe per turbine, which allows the concept of modular build-up of new capacity to meet growth needs. Also, gas turbine power generation equipment offers a low risk, low capital cost, quick-return option. Current combined-cycle plants have claimed cycle efficiencies of the order of 49 percent (7,000 Btu/kWh, based on the HHV), and can achieve environmental compliance for SO_2 and NO_x with minimal effluent or byproduct streams. Further, they can be installed in a very short time frame as a turnkey operation—less than 12 months is possible. Given that gas turbines will likely account for an increasing fraction of the base-load generating capacity, developments to improve their efficiency and improve their environmental compatibility would have a significant positive effect on the economics of power industry.

OVERVIEW OF THE ATS PROGRAM

The overall goals of the ATS program are to develop advanced gas turbine systems to serve both utility and industrial power generation markets. The systems intended for utility application will have an efficiency rating of at least 60 percent (based on the lower heating value; better than 54 percent, based on the HHV), whereas the industrial power generation systems will be at least 15 percent more efficient than current gas turbines. The baseline fuel is natural gas. The ATS machines will have superior environmental compliance, with NO_x levels less than 8 ppm, and CO and hydrocarbon emissions less than 2 ppm. The goal of these systems is to reduce the cost of electricity by at least 10 percent. There is also a requirement that the systems developed will be adaptable to coal and biomass firing; this assumes that gasification processes will be developed as planned[4] to allow a smooth transition from natural gas to coal when that is economical and strategically necessary.

APPROACHES TO ACHIEVING THE ATS PROGRAM GOALS

The approaches available for achieving the program goals were analyzed in an initial phase, in which systems studies were conducted by Allison Engines; ABB Power Generation, Inc., the General Electric Co.; Solar Turbines, Inc.; United Technologies; and Westinghouse Electric Corp. The major areas identified where improvements could be made, and where significant investment would be required were:

- Recuperation to preheat the air entering the combustor.
- Advanced combustors/catalytic combustion to achieve significant reduction in NO_x levels.
- Methods for increasing the TRIT: An increase of 55°C (100°F) in turbine inlet temperature (TRIT) can provide an 8 to 13 percent increase in power output, and a 1 to 4 percent increase in simple cycle efficiency. Suggested approaches include items such as:
  —introduction of single crystal alloys,
  —introduction of low-sulfur alloys,
  —adaption of advanced cooling designs,
  —development of closed-circuit cooling with steam,
  —development of new alloys and advanced cooling designs, which could require ceramic core and mold development,
  —use of thermal barrier coatings (TBCs; a TBC is essentially a materials system which consists of a thermally-insulating ceramic bonded to an oxidation-resistant metal coating which is integral with the substrate), and
  —development of ceramic vanes and blades.
- Increased aerodynamic efficiency from the use of improved sealing and advanced airfoil designs (also applies to steam turbine).
- Improved steam cycles: generation of higher-temperature steam (from 10 MPa/538°C/538°C (1450 psi/1000°F /1000°F) to 12.4 MPa/593°C/593°C (1,800 psi/1100°F /1100°F), or 10 MPa/816°C (1500 psi/1500°F) once-through).
- Thermodynamic recuperation: use of exhaust heat in a methane reforming process to produce fuel gas which would augment the normal fuel.

Some indication of the potential gains associated with some of these approaches can be seen from the following:

Approach	Potential Efficiency Gain
Advanced materials, TBCs	3-4%[5]
Thermodynamic recuperation	2%[6]
Advanced air cooling	1-2%[7]
Reduced leakage (clearance)	0.5-0.6%[5]
Improved aerodynamics (gas turbines & steam turbines)	0.2-0.4%[5]

MATERIALS DEVELOPMENT NEEDS

Some of the materials developments required to realize the goals of the ATS program will be based in part on technology already in use in modern aircraft engines. These engines emphasize maximization of specific thrust, and especially peak thrust in the case of military applications, which is achieved largely through increased turbine inlet temperatures and increased pressure ratios. The pressure ratios of military engines may be as high as 40:1, with the gas temperature at the first stage blade (TRIT) upwards of 1500°C (2732°F). Compact engine dimensions are obtained through the use of multi-

spool designs to achieve high pressure ratios, and high individual high-pressure turbine stage loadings. A premium has, therefore, been placed on the development of alloys with appropriate strength at extreme temperatures, and the improvements in aircraft engine performance with time can be accurately tracked by developments in alloy design and processing. Current single crystal alloys experience bulk metal temperatures up to 1000°C (1832°F) in aircraft engine service, reaching 1075 to 1100°C (1967 to 2012°F) at hot spots, but the useful lifetime under these conditions (in the absence of a TBC) is usually less than 5,000 hours. It is considered that alloying technology will allow one more generation of single crystal alloys (which are now in the 3rd generation), which may give a further 28°C (50°F) increase in temperature capability. Further increases in the temperature capability of metallic blade and vanes will probably have to rely on advances in cooling techniques and TBCs, in the absence of alloy technology breakthroughs[8].

Since superalloys begin to melt at 1260-1290°C (2300-2350°F) or so, it has been necessary to use advanced cooling of the critical components, such as combustors, transition ducts, and the first stage vanes and blades. This is currently accomplished by a combination of air cooling, and the use of thermal barrier coatings. The air for turbine cooling is bled from the compressor, and can be as much as 20 percent of the compressor capacity. Since this air bypasses the combustion process, a significant efficiency penalty results. The need for more efficient use of this cooling air has led to progressively more complicated cooling passage designs, hence more complicated blade and vane castings. TBCs were introduced in combustors of advanced engines approximately ten years ago, and now are used routinely on transition ducts as well as combustor liners. Trials are in progress using TBCs on the platforms and, in some cases, as patches on the airfoils of first stage vanes and blades; in this application, the TBC is an added benefit, and no credit is taken for it by reducing the amount of cooling air.

The design of heavy frame turbines has been typically conservative, with greater emphasis on long service life and minimized maintenance requirements than on higher performance and efficiency. However, the performance of these turbines has been gradually improved without much sacrifice of service life and reliability as new materials and designs incorporating aircraft engine experience have been evolved. As a result, the TRIT of a state-of-the-art heavy frame engine is of the order of 1300°C (2370°F), and pressure ratios range up to 16:1. It is expected that the application of advanced air-cooling techniques as well as TBCs will be required to allow an increase in the TRIT to 1427°C (2600°F), together with a lifetime in excess of 25,000 hours. Further increases in the TRIT probably would require the use of newer cooling technologies such as closed-loop cooling with air or steam.

The transfer of aircraft engine technology for the ATS application must be made with the recognition that the different duty cycle envisioned for these engines will impose different requirements on the critical components and associated materials. In particular, ATS engines will run for a large fraction of their operating lives at close to full power, in contrast to the use of close to full power by aircraft engines only during

take off and landing (and combat). In addition, the required time between overhauls and refurbishment is 25,000 hours, which is some five times that required for aircraft engines. The inherently longer time at temperature suggests that time-dependent processes such as creep, corrosion, and sintering (of ceramic thermal barrier coatings) will play an increased role in the degradation processes experienced by ATS engine components. Since the bulk metal temperature must be commensurate with the creep allowances, blade temperatures may have to be somewhat lower than those tolerated in aircraft engines, or more creep-resistant alloys must be introduced.

The short-term and long-term materials needs for developing advanced land-based turbines were recently identified using input from gas turbine manufacturers, materials suppliers, universities, and government laboratories[9]. The key technical areas were considered to be:

- coatings and process development,
- directional solidification and single crystal airfoils manufacturing technology,
- turbine airfoil development; ceramics development,
- materials characterization,
- catalytic combustor materials, and
- efficient technology information exchange to fully use parallel efforts.

Materials issues inherent in coatings and airfoil development are: corrosion, fouling, uniformity of cooling, the impact of steam cooling, leakage, and operation on coal or coal-derived fuels. The development of TBCs with improved bond coats was rated by many respondents as the most important element for advanced land-based turbine applications.

TBC DEVELOPMENT NEEDS

TBCs are currently used on combustors, vane and blade platforms (and airfoil surfaces, but usually as patches) of aircraft engines as a means of increasing the component life, rather than as life-dependent coatings. TBC thicknesses of 500 to 1,000 μm (20 to 40 mil) are used routinely on combustors, and 250 to 500 μm (10 to 20 mil) on first stage vanes whereas, on rotating parts, the thickness is limited by weight and aerodynamic considerations to approximately 125 to 250 μm (or possibly 380 μm) [5-10 mil (or possibly 15 mil)]. Given that ATS machines will operate for significantly longer times at close to full power than do aircraft engines, it may be necessary to reduce the average bulk metal temperature of the first-stage blades below that permitted for aircraft engines to minimize creep. Even without such a requirement, the relatively thin TBC layer currently applicable to the blade surfaces suggests that substantial cooling of the blade still will be necessary. As a result of these considerations, it appears that developments are needed to increase the effectiveness of the TBC that can be applied to blade surfaces. The necessary improvements may involve developements to allow the use of increased coating thickness, coatings with decreased density, or decreased thermal conductivity. A further approach to minimize the efficiency penalty associated

with cooling requirements may involve cooling the air before it enters the blades, or the introduction of steam-cooling for the first-stage vanes to provide more air for the blades.

The key technical issues to be addressed in the development of current TBCs for use on the hot section components of ATS machines are, therefore, the mechanical and chemical stability of the ceramic layer (YSZ) and of the bond coat; the thermal expansion characteristics and compliance of the YSZ layer; and the thermal conductivity across the thickness of the ceramic layer.

PROPOSED EFFORT ON TBCs IN THE ATS PROGRAM

The effort proposed on the ATS program in support of TBCs will address the following issues:

(1) TBC Materials and Process Development

In order to hold the turbine blade temperatures to the limits required for $\geq 25,000$ hours, there are several materials issues to be considered for designing and manufacturing TBCs. These issues include sintering, phase changes, damage due to high angle impact of ingested particles, corrosion and destabilization of the YSZ layer, and long-term thermal and oxidation stability at the bond coat interface.

The mechanical integrity and compatibility with the substrate depend on careful control of the TBC microstructure. For rotating components, a columnar grain microstructure (grain boundaries oriented perpendicular to the surface) as produced by electron beam-assisted physical vapor deposition (EB-PVD) is preferred, since it produces a lower elastic modulus parallel to the metal substrate, and a higher modulus across the thickness. The increased coating compliance parallel to the substrate results in significantly improved spallation resistance. The ability to control this columnar orientation along with coating uniformity, while accommodating cooling holes and maintaining the desired aerodynamic airfoil surfaces, is a major consideration in the development of processes for coating large airfoils. TBCs are also deposited by plasma spraying, which often results in a more porous structure consisting of an agglomeration of solidified splats oriented parallel to the substrate surface. This process is less expensive than EB-PVD, and better-suited to coating large components.

Sintering and phase changes of the TBCs have been shown to occur at the turbine operating temperatures. Sintering increases the tendency for spallation, as well as thermal conductivity, both of which result in increases in the temperature of the metal substrate. The longer-term exposure to high-temperatures projected for ATS machines will accentuate this problem. Characterization of the thermal and physical properties of the TBC as a function of exposure conditions and of its mechanical performance at rated heat fluxes is, therefore, an essential part of any process development.

The deposition of corrodents and their adverse influence on the chemical and mechanical integrity of the TBC layer is a potentially serious threat for coal- and biomass-based fuels. For example, the Y_2O_3 phase in the YSZ system is known to be susceptible to fluxing in the presence of molten salts containing sodium and vanadium[10]. Also, infiltration of the surface of the TBC by deposited salts that are molten at operating temperature could promote spalling of the YSZ when they solidify on cool-down.

Current coating processes, especially plasma spraying, can result in plugging of cooling holes in turbine airfoils, and so lead to the need for secondary processing for removal of the coating in the holes. In advanced turbine airfoils, shaped holes are used to enhance film cooling; a coating that distorts the hole shape, hence the cooling air flow, could negate the benefits of the TBC. The surface finish of the coating is also important to maintain aerodynamic performance. Secondary processing that uses coolants that could contaminate the relatively porous TBC would be highly undesirable.

Manufacturing process developments will be required that improve the reliability and reproducibility of TBCs. Intelligent processing of materials is being emphasized in programs sponsored by the National Institute of Standards and Technology and the Advanced Research Development Agency.

(2) Bond Coat and Metallic Coating Development

Two general types of bond coat have been used: overlaid MCrAlY, where M is typically Ni or NiCo, and diffusion aluminide (or Pt-aluminide). These are shown schematically in Figure 1. The first coating is formed by plasma-spraying directly on to the component, whereas the second is formed by the diffusion of aluminum into the alloy substrate.

A major concern associated with the longer design life for advanced land-based gas turbines is the stability of the bond coat. The ease of transport of oxygen through YSZ ensures that the bond coat will be subject to continued oxidation, as well as hot corrosion if deposited salts can penetrate to the bond-coat region. The ability of the bond coat to continue to support the growth of an alumina scale depends on the total amount of aluminum available; the effective reservoir of aluminum in the bond coat is depleted not only by consumption by the growth of the alumina scale, but also by diffusion into the alloy substrate. If a point is reached where the aluminum level of the bond coat falls below that at which alumina can be formed preferentially, faster-growing oxides of the other constituents of the bond coat will form, and the adherence of the TBC could be significantly degraded. Another factor is that the continuing growth of the otherwise protective Al_2O_3 at the bond coat-YSZ interface is a source of increasing strain, which may affect the adherence of the external ceramic layer.

Spallation of the YSZ layer induced by the oxidation of the bond coat is considered to be the ultimate failure mode for EB-PVD TBCs. Failure usually occurs at the interface between the bond coat and its oxide scale, whether the bond coat is diffusion aluminide

or overlaid MCrAlY. This observation suggests that the life of these TBCs is largely controlled by the factors that govern the bonding at the scale-bond coat interface. For air plasma-sprayed TBCs, spallation usually occurs within the ceramic layer, but very near to the bond coat-YSZ interface. The major sources of stress leading to degradation of TBCs appear to be[11]: thermal mismatch with the bond coat; temperature gradient through the TBC; stress at the YSZ-bond coat interface from oxide growth; and transformations in the YSZ, especially tetragonal to monoclinic which results in a 3 to 5 percent volume change.

A recent discovery is that even trace amounts (≈1 ppm) of sulfur impurity in airfoil alloys can degrade the integrity of the bonding at the scale-alloy interface. This sulfur effect can be minimized by reducing the inherent sulfur impurity level in the alloy through in-melt or post-solidification processing, or through the incorporation of reactive elements additions (such as Y). The use of Y-containing alloys requires the use of alumina for cores and molds, with inherent difficulties of handling and core extraction. The alternative is modification of current melt practices, as well as bond coating processes. There is, therefore, a need for definitive microstructural studies of the effectiveness of approaches to minimize the sulfur effect on the rate of scale growth on bond coat alloys, and on the adhesion of TBCs to such alloys, as a function of exposure time and temperature. These issues are shown schematically in Figure 2.

(3) TBC Analytical Modeling, Life Prediction, and Maintenance and Repair

The development of an analytical model that could predict TBC performance and life based on key TBC parameters and operating conditions could be a major step toward enhanced TBC reliability in an application which is totally dependent on the TBC to reduce airfoil temperature as well as increase airfoil life. The model should address interrelationships among geometric factors (thickness and surface curvature); thermomechanical considerations (gas temperature, cooling characteristics, mechanical stresses, and mechanisms for damage accumulation); material parameters (conductivity and coefficient of expansion; coating strength; bond strength; long-term creep properties; oxidation resistance, corrosion resistance, and microstructural stability). These issues are summarized in Figure 3.

It may be necessary to incorporate several different but related models into the overall model, such as: heat transfer model (prediction of temperature and gradients); materials models (prediction of thermal conductivity and stress-strain response); and mechanistic model (prediction of thermal and mechanical stress fields in the coating, substrate, and interface as a function of configuration, temperature, environment, and time). Experimental validation of the interrelationships in these models is key to the implementation and use of the overall model in the TBC manufacturing process. In this manner, such a model and its experimental verification could aid in establishing system tradeoffs and synergistic improvements.

The time-temperature exposures and projected thermal gradients in TBCs to meet ATS needs are beyond the current TBC experience base. Laboratory test facilities will be

required that can measure properties needed for modeling as described above. In addition, rig test facilities that can simulate the temperatures and high thermal gradients, gas velocities, and pressure on the TBCs are required. Built-in sensors capable of predicting imminent TBC failure in the field would reduce the risk associated with applications where the higher inlet temperature and reduced cooling air are dependent on the TBCs.

Given the critical role of TBCs, reliable inspection procedures will be needed to determine coating section thicknesses and detect defects. A methodology must also be established for the repair of components, either on-site or at a designated depot. Techniques that can establish non-destructively the remaining life of turbine airfoils are actively being sought for aircraft turbines, and such techniques will become even more critical for land-based gas turbines, where time-dependent properties such as creep assume greater importance.

(4) Revolutionary TBC Concepts

TBCs with better thermal properties than achievable by current concepts (YSZ) may be needed to reach the target operating temperature of the ATS hot gas path components. Simply increasing the thickness of a given monolithic coating does not necessarily produce a corresponding increase in thermal resistance, since radiation contributes significantly to the overall heat transfer process at the ATS design temperature. The goal of radically new concepts for TBCs would be to maximize thermal resistance while minimizing coating thickness; thinner coatings might be expected to exhibit improved mechanical properties. There are several approaches for increasing the thermal resistance of a TBC; examples include:

(1) controlled increase in the population of microstructural discontinuities (grain boundaries, voids) in currently-used materials, to increase the thermal resistance
(2) multiple layers of precisely-controlled thickness to provide light-refracting interfaces for selected wavelengths, or
(3) compositionally-graded structures to provide a transition in mechanical properties (such as coefficient of expansion) across the bond coat-TBC interface; and, less revolutionary,
(4) diffusion barriers to reduce the permeability of the TBC to oxygen (and to alkali metal salts), and slow the growth of the oxide on the bond coat.

SUMMARY

The ATS program has the goal of developing gas turbine systems for both utility and industrial power generation applications that will have efficiency ratings significantly higher than current gas turbine systems, that will operate for significantly longer periods between scheduled maintenance outages and, in addition, will have superior emissions performance. Realization of these goals will make use of hot gas path component technologies developed for aircraft engines, but adapted to the quite

different duty cycle of ATS machines, which will operate mostly at close to full power rating and exhibit significantly longer lifetimes. A major difference will be that the ATS machines will <u>depend</u> on TBCs to allow a reduction in the cooling air requirements and to realize extended component lifetimes whereas, in aircraft engines, TBCs are used on blades and vanes only to gain added service life benefits, and no reliance is placed on them so that cooling air can be reduced. A significant TBC development effort is, therefore, planned in the ATS program which will address the following issues: improvement of the performance of TBC materials, including the ceramic insulator and the metallic bond coating; development of processes for applying TBCs to large, complex structures with good control of the coating thickness and structure; analytical modeling of the coating degradation processes to allow life prediction, maintenance and repair, and the exploration of revolutionary TBC concepts that have potential for increased thermal resistance and thinner coatings. The Department of Energy is currently pursuing solicitations for major research programs in these areas.
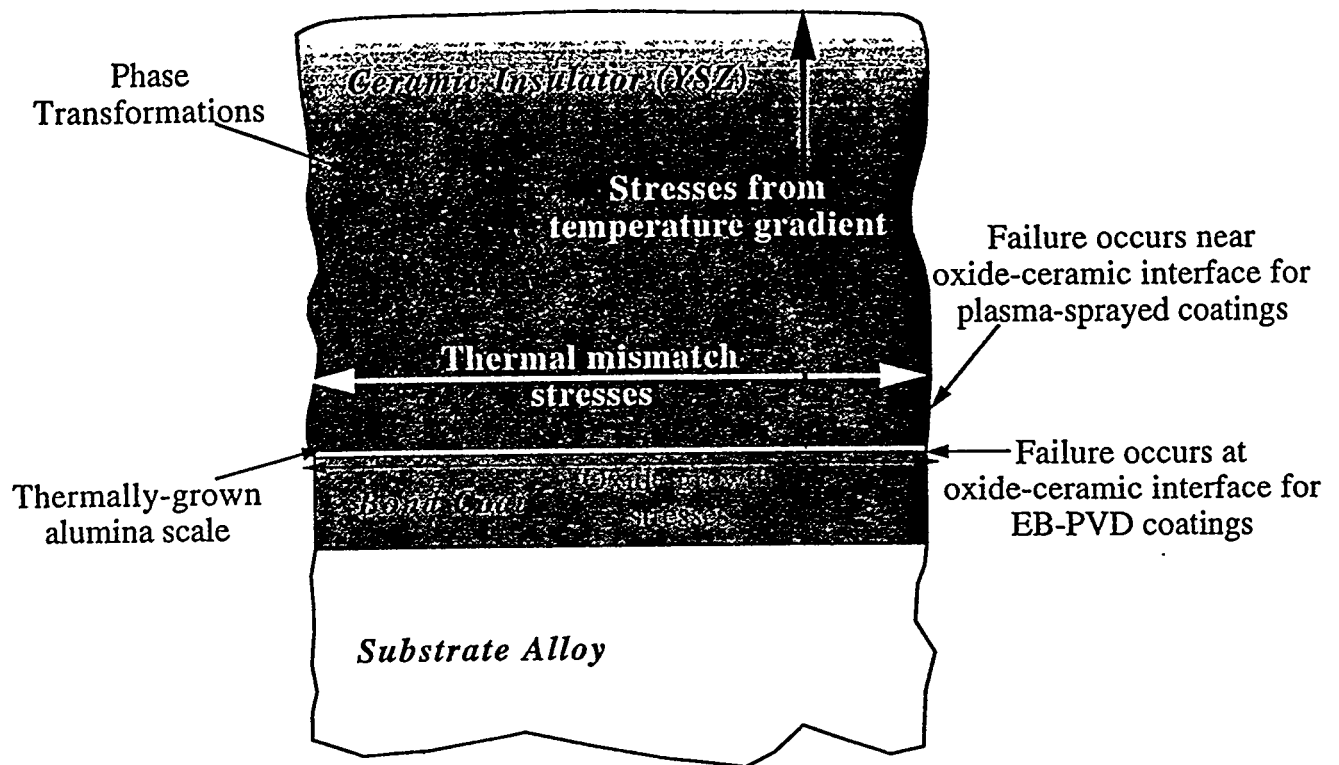
ACKNOWLEDGMENTS

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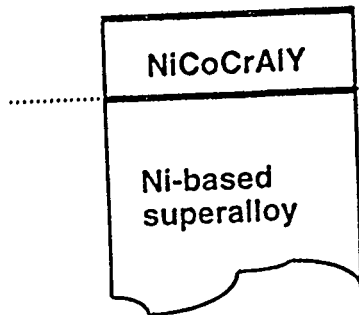
Phase Transformations

Stresses from temperature gradient

Failure occurs near oxide-ceramic interface for plasma-sprayed coatings

Thermal mismatch stresses

Failure occurs at oxide-ceramic interface for EB-PVD coatings

Thermally-grown alumina scale

Ceramic Insulator (YSZ)

Bond Coat

Oxide growth stresses

Substrate Alloy

Overlay NiCoCrAlY

NiCoCrAlY
Ni-based superalloy

VPS
APS
EBPVD

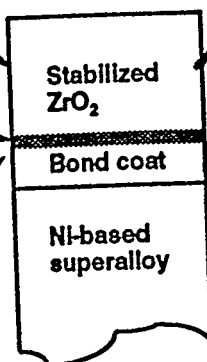## Diffusion Aluminide (NiAl)

NiAl
Ni-based superalloy
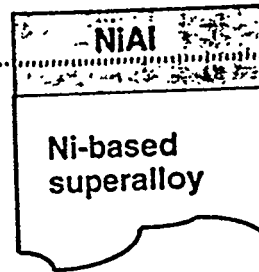
CVD
Gas phase pack
Pack Cementation

Goals

Higher thermal stability

Reduced spallation at the interface

Improved oxidation/corrosion Protection

Stabilized ZrO₂

Bond coat

Ni-based superalloy

Issues & Challenges

- Microstructure, Stabilizer
- APS, EBPVD, etc.

Understanding damage mechanisms
    Oxidation, Fatigue, Creep, Phase & Property changes, Hot corrosion, Erosion, Diffusion, etc.
Reliable life prediction

- NiAl, MCrAlY, Desulfurization
- VPS, HVOF, CVD etc.