Combustion Chemical Vapor Deposited Coatings for Thermal Barrier Coating Systems

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Abstract

Thermal barrier coatings (TBC's) are presently used on hot section components of gas turbines. Their role is to provide thermal insulation for blades, vanes, etc., and thereby permit an increase in firing temperature which provides corresponding gains in thermodynamic efficiency and possibly extensions of component life. The objective of this research program is to produce TBC's that exhibit increased lifetimes.

This research addresses two general failure modes of air plasma sprayed (ATS) TBC's: spallation associated with a failure near the bond coat/YSZ interface and spallation associated with corrosion/erosion of the outer surface of the YSZ. The strategy for producing improved TBC's is to incorporate an additional coating layer into conventional TBC's. This layer is deposited either directly to the bond coat, and is termed an "interlayer" coating; or alternately, this additional layer is deposited over top of the YSZ at the outer surface; this layer is termed an "overlayer" coating. This technique being used to deposit the thin films is combustion chemical vapor deposition (CVD). Combustion CVD is a novel, flame-assisted deposition process that is performed in the open atmosphere, and therefore does not require the coating chambers and vacuum hardware associated with traditional thin film deposition processes.

This project is a joint effort between Georgia Tech (GT) and General Electric Power Generation (GEPG), which is subcontracted to GT to supply substrates, plasma sprayed bond coats and YSZ top coats, testing services, and technical expertise. GT is depositing the combustion CVD coatings, performing oxidation testing, and characterizing the tested specimens.

Combustion CVD deposition parameters have been developed for: alumina, nickel-spinel, magnesium-spinel, ceria, and YSZ, all under investigation for use in TBC's. The Ph.D. thesis research of G. W. Book quantified the relationship between reagent concentrations, aerosol droplet size distribution, and the resulting population of particle clusters in the coatings. Dr. Book determined that deposition rates can be increased without the formation of clusters by increasing the reagent concentration and simultaneously decreasing droplet size.

Michelle Hendrick, who completed her Master's thesis under this program, studied the oxidation protection provided by the thin (<1 µm thick) alumina coatings deposited using combustion CVD. She quantified the degree of oxidation protection provided by the alumina for a proto-typical bond coat material: Ni-20Cr, using isothermal thermogravimetric analysis (TGA) in air. Ms. Hendrick's results indicate that for deposition temperatures between 850 and 1,250°C alumina deposits primarily as nano-dimensional 0-Al_2O_3, which reduces the oxidation rate of the underlying substrate during isothermal oxidation for temperatures ranging between 800 and 1,100°C. Vacuum annealing the coated specimens prior to TGA testing further reduces the oxidation rate, and causes the formation of an approximately 1 µm thick layer of spinel.

David Stollberg, who is in the final year of his Ph.D. program, has developed the deposition parameters for depositing $NiAl_2O_4$ and $MgAl_2O_4$ using combustion CVD. These have been applied to GE-provided samples for use as interlayers and overlayers, respectively, within TBC systems. Tests for corrosion and erosion of the overlayer samples and furnace cycle tests for the $NiAl_2O_4$ interlayer will be begin in November, 1997.

Both alumina and ceria thin films have been incorporated into TBC's as interlayers between the bond coat and YSZ. N5 single crystal substrates were vacuum plasma bond coated, combustion CVD coated, and APS YSZ coated. One set of alumina coated specimens was vacuum annealed before being YSZ coated. Furnace cycle testing (FCT) was performed on these three sets of specimens (ceria coated, alumina coated, alumina coated + anneal) and on conventional TBC control specimens. The alumina coated/annealed specimens lasted 63% longer, on average, than the control specimens. Other combustion CVD coated specimens displayed no significant FCT lifetime increases. Perhaps more significantly, the alumina coated and annealed specimens displayed a different failure mode than the other specimens, which all failed within the YSZ. The alumina coated and annealed specimens consistently failed beneath the bond coat. Repeat experiments designed to differentiate between the effects associated with the anneal versus the effects associated with the coating in conjunction with the anneal, indicate that samples which have been annealed at the GT facility last approximately 25% longer than the control samples annealed by GEPG. Further, they consistently fail beneath the bond coat, thus indicating that the GT treatments improve the YSZ/bond coat interface.

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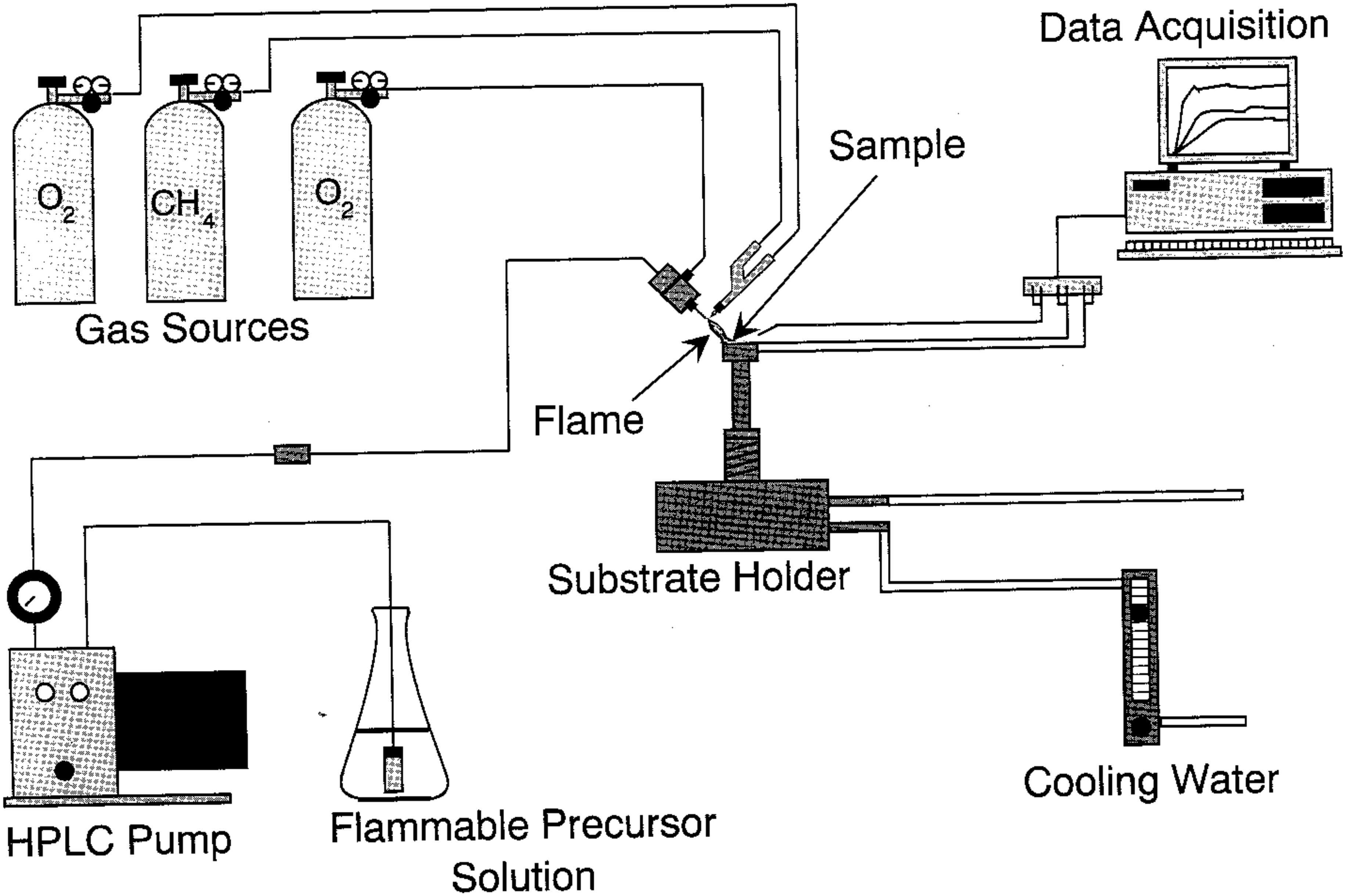
Introduction

Thermal Barrier coatings (TBC's) are of importance to gas turbine manufacturers due to their ability to allow increased firing temperatures, thereby improving engine efficiency and potentially also providing concomitant gains in component life. This research focuses on materials selected for examination as either overlay coatings on top of TBC systems, or as interlay coatings between industrially applied layers of bond coat and yttria stabilized zirconia. The method utilized for depositing these materials is a new technology, termed combustion chemical vapor deposition, or combustion CVD.

Combustion Chemical Vapor Deposition

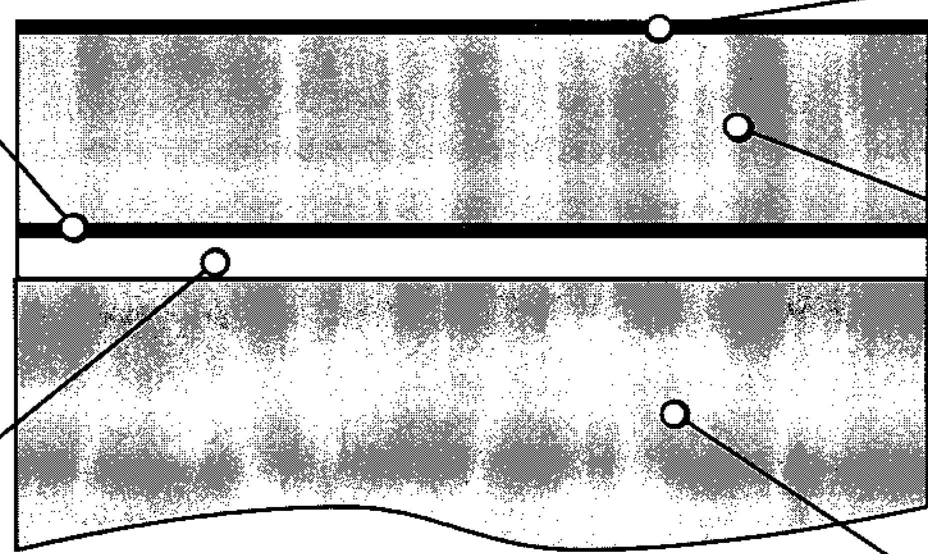
- Inexpensive -- little capital investment, precursors readily available
- Performed in open atmosphere; potential for assembly-line production of thin films
- Advantages of CVD without associated cost
- Heat for CVD provided by combustion process, no need for external heating of substrates
- Versatile, potentially portable

Combustion CVD System



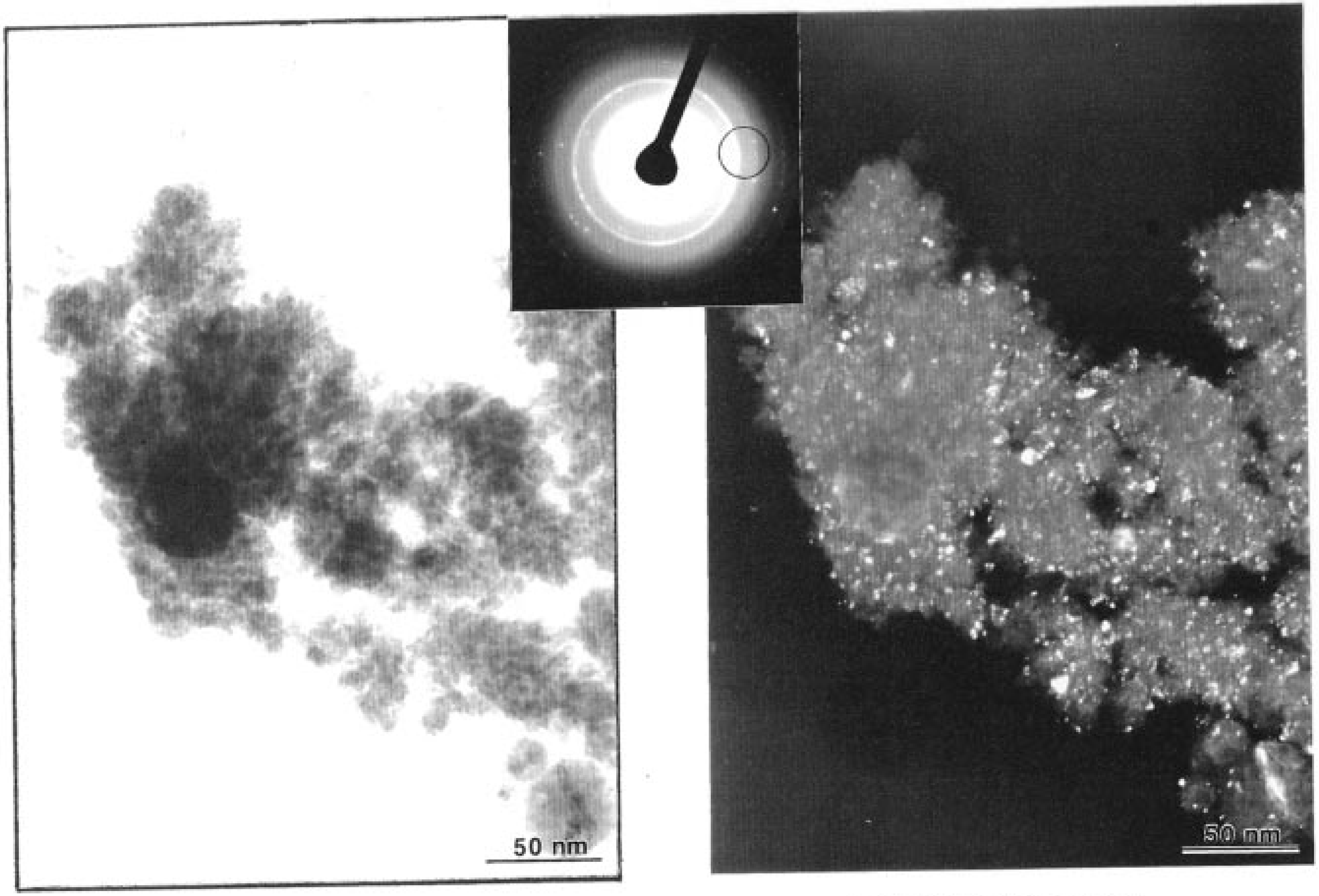
OBJECTIVE

Produce and Evaluate TBC's Modified with Combustion CVD Layers



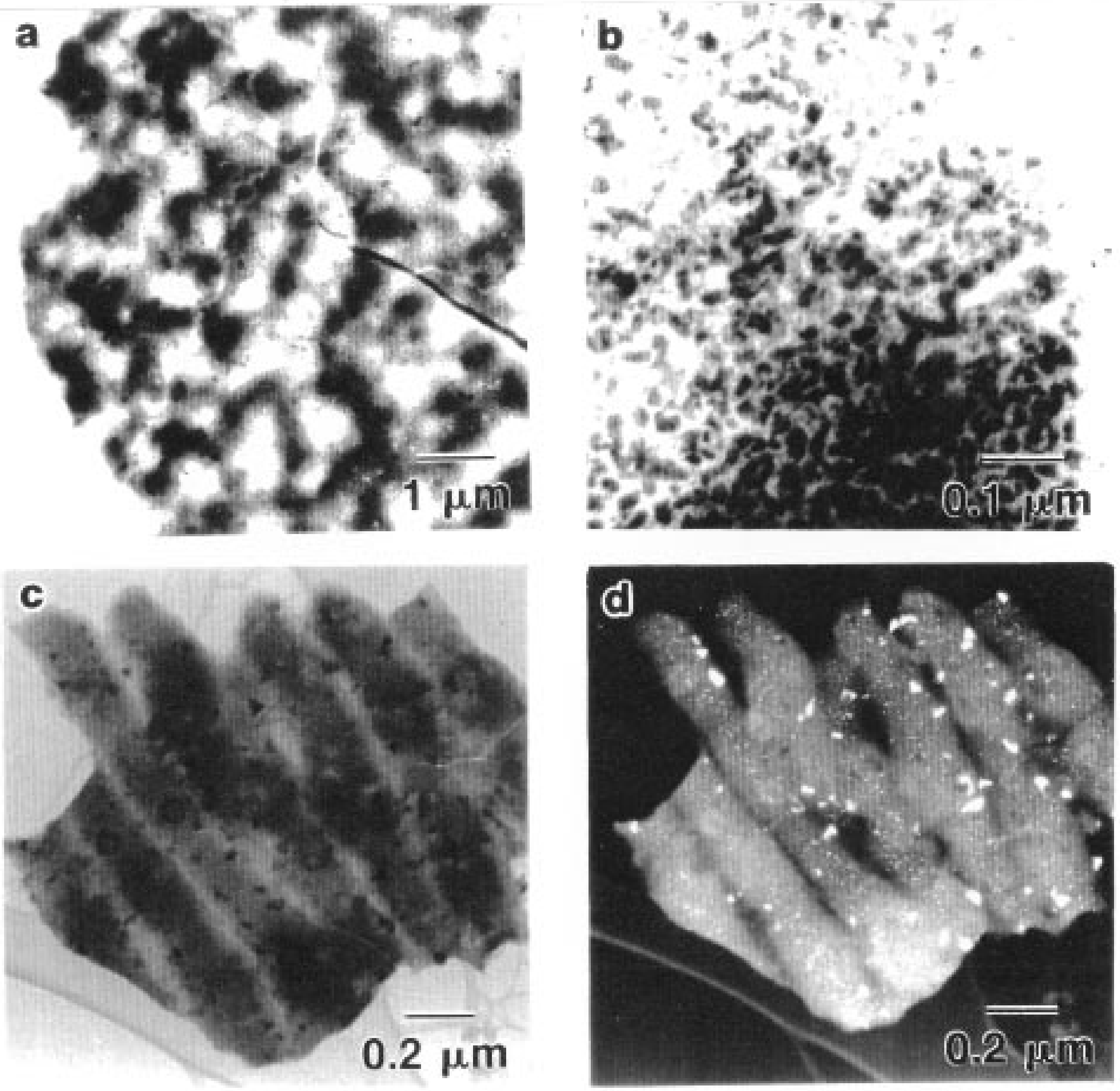
Material	Precursor	Conc.	Microstructure
alumina	Al acetylacetonate	0.00135 M	nano-dimensional (<20 nm grain size) theta-phase
silica	tetraethyloxysilicate	0.0025 M	amorphous
YSZ (7%)	Y and Zr ethylhexanoate	0.001M-0.005 M	equiaxed grains 0.1-1.0 μm
ceria	Ce ethylhexanoate	0.001 M	grain size a function of primary aerosol diameter
$NiAl_2O_4$	Ni acetylacetonate, Al acetylacetonate	0.004 M	spinel, nano-dimensional (< 100 nm in grain size)
$MgAl_2O_4$	Mg naphthenate, Al acetylacetonate	0.002 M	spinel, nano-dimensional

Alumina and Silica



BRIGHT FIELD DARK FIELD

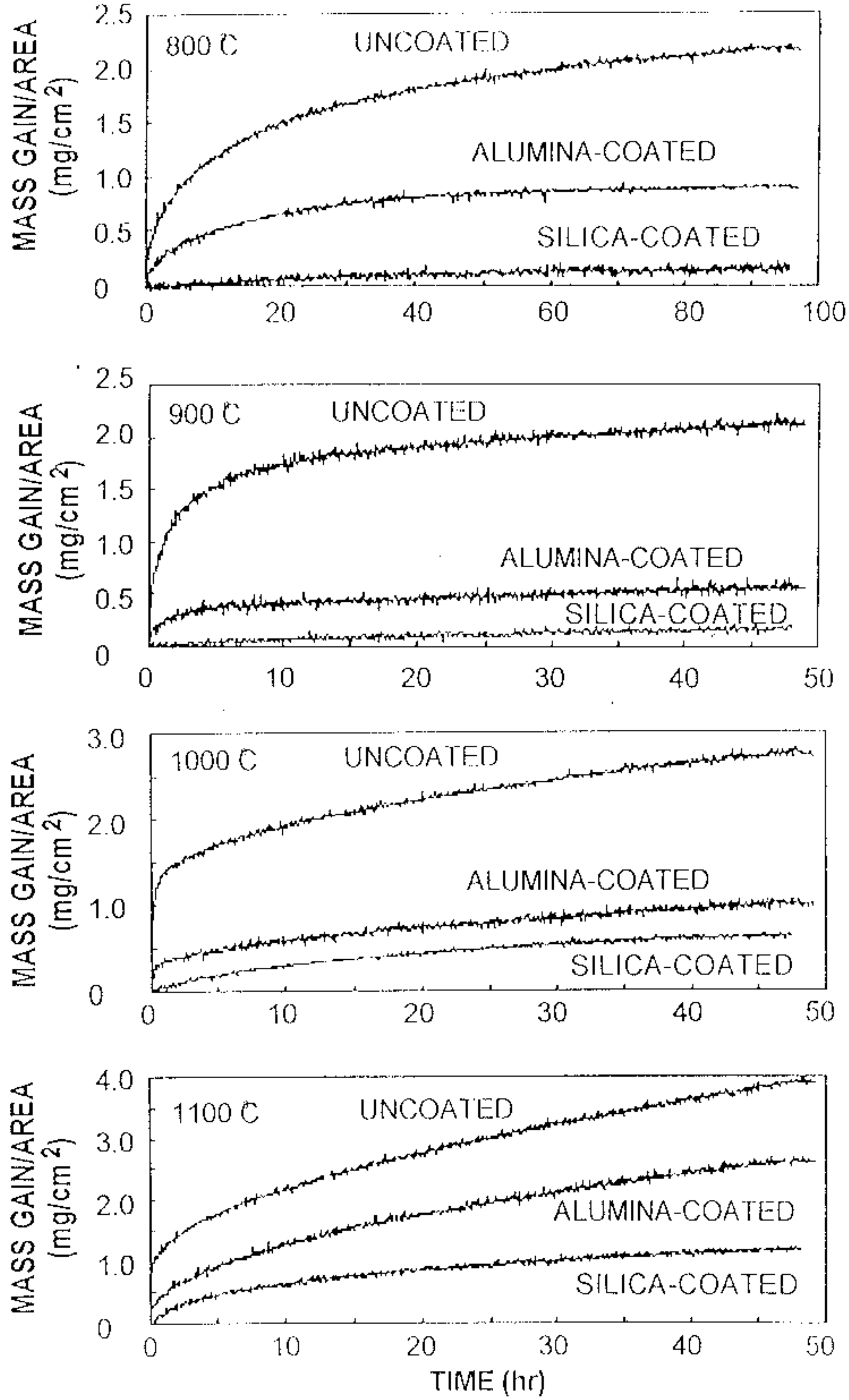
ALUMINA DEPOSITED VIA COMBUSTION-CVD



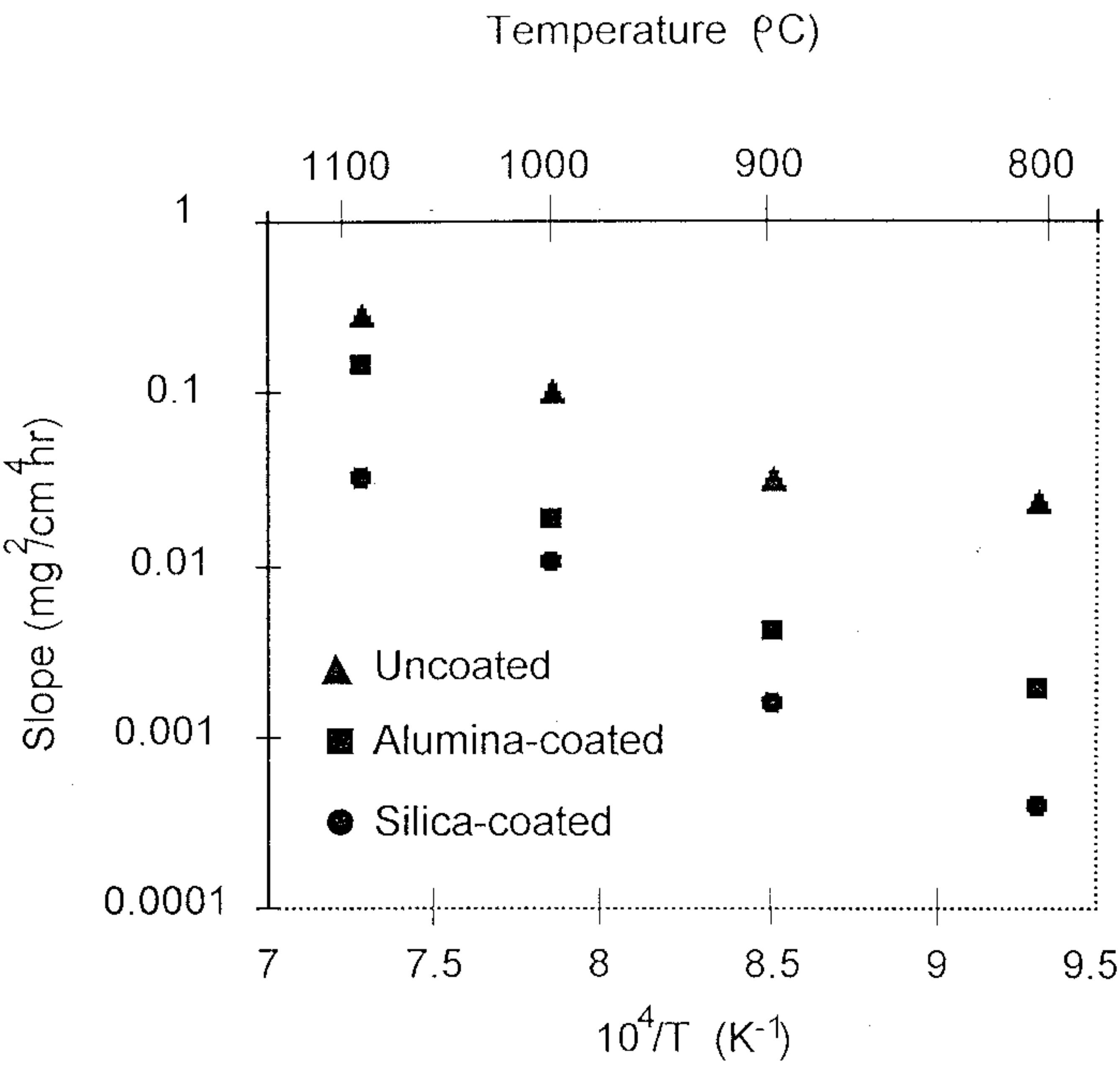
SILICA DEPOSITED VIA COMBUSTION-CVD

THERMOGRAVIMETRIC ANALYSIS

Uncoated, alumina- and silica-coated Ni-20Cr was oxidized in dry, high purity air and the rate of oxidation was characterized using thermogravimetry. The figure above summarizes the improvements is oxidation caused by the coatings. Below, the parabolic rate constants are summarized in an Arrhenius plot.



MASS GAIN PER UNIT AREA VS. TIME DATA FOR OXIDATION AT 800,900,1000, AND 1100°C FOR UNCOATED, ALUMINA-COATED, AND SILICA-COATED Ni-20Cr.



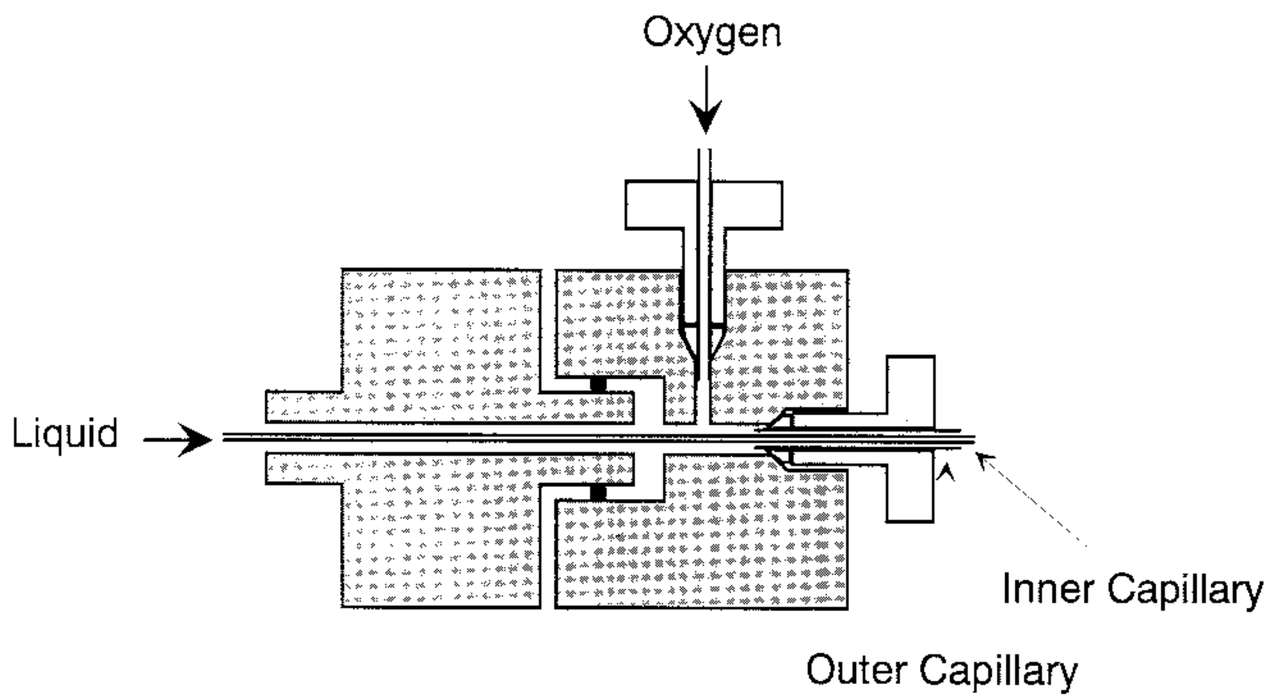
OXIDATION RATE CONSTANTS VERSUS RECIPROCAL TEMPERATURE FOR UNCOATED, SILICA-COATED, AND ALUMINA-COATED Ni-20Cr.

Ceria and YSZ

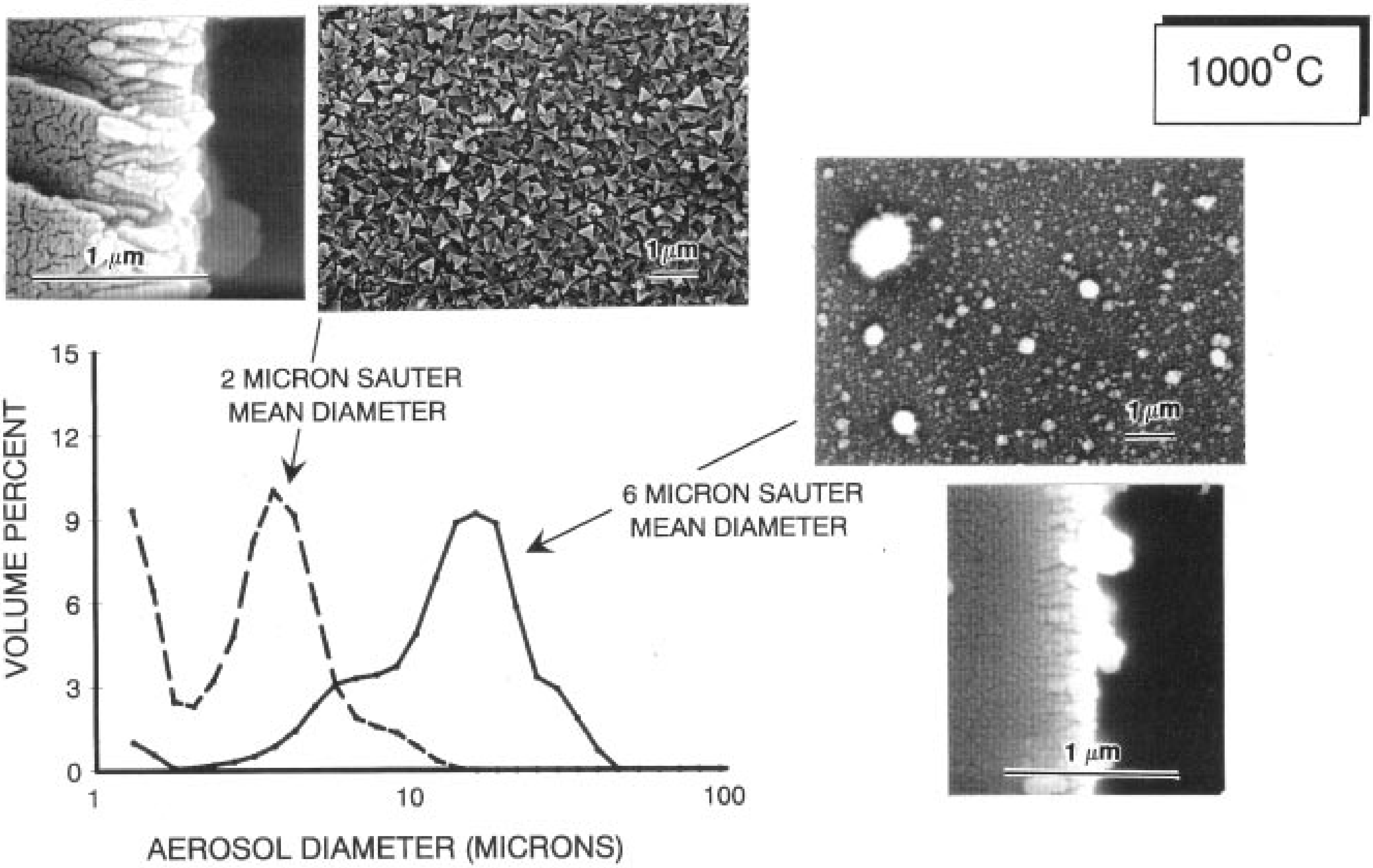
OSCILLATING CAPILLARY NEBULIZER (OCN)

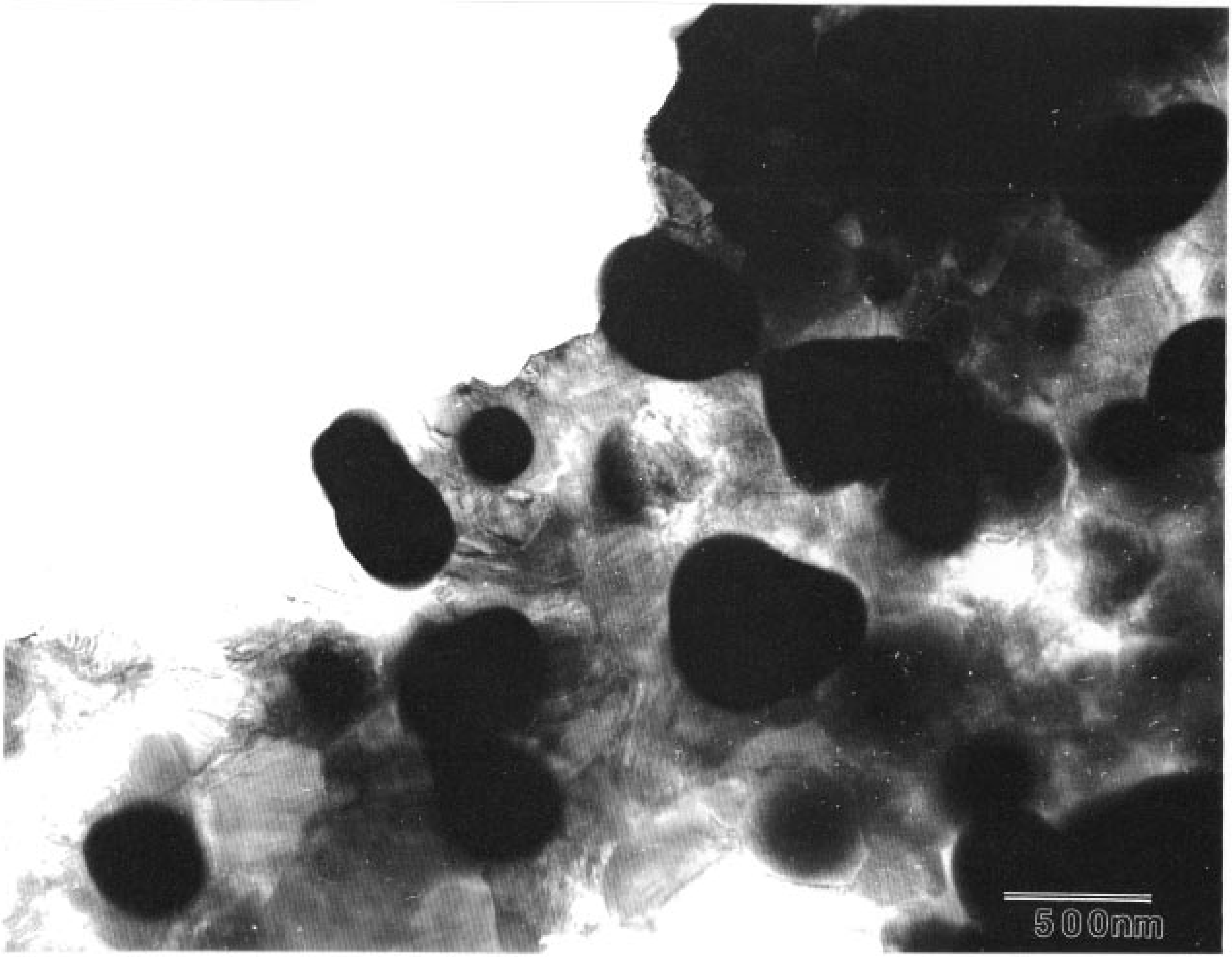
A newly developed nozzle assembly, the OCN, allows control over the droplet size distribution produced, as shown below for two nozzle configurations. The OCN consists of two concentric silica capillary tubes; the inner tube is used to transport liquid to the flame front while the outer tube is used to transmit the oxidizing gas. The coating produced with the smaller size aerosol is dense, nodular and adherent; whereas the coating produced with the larger size aerosol is powdery and non-adherent. Thus, by reducing the overall droplet size distribution, the temperature necessary for high quality films is reduced.

OSCILLATING CAPILLARY NEBULIZER ENABLES CONTROL OF PRIMARY AEROSOL DIAMETER



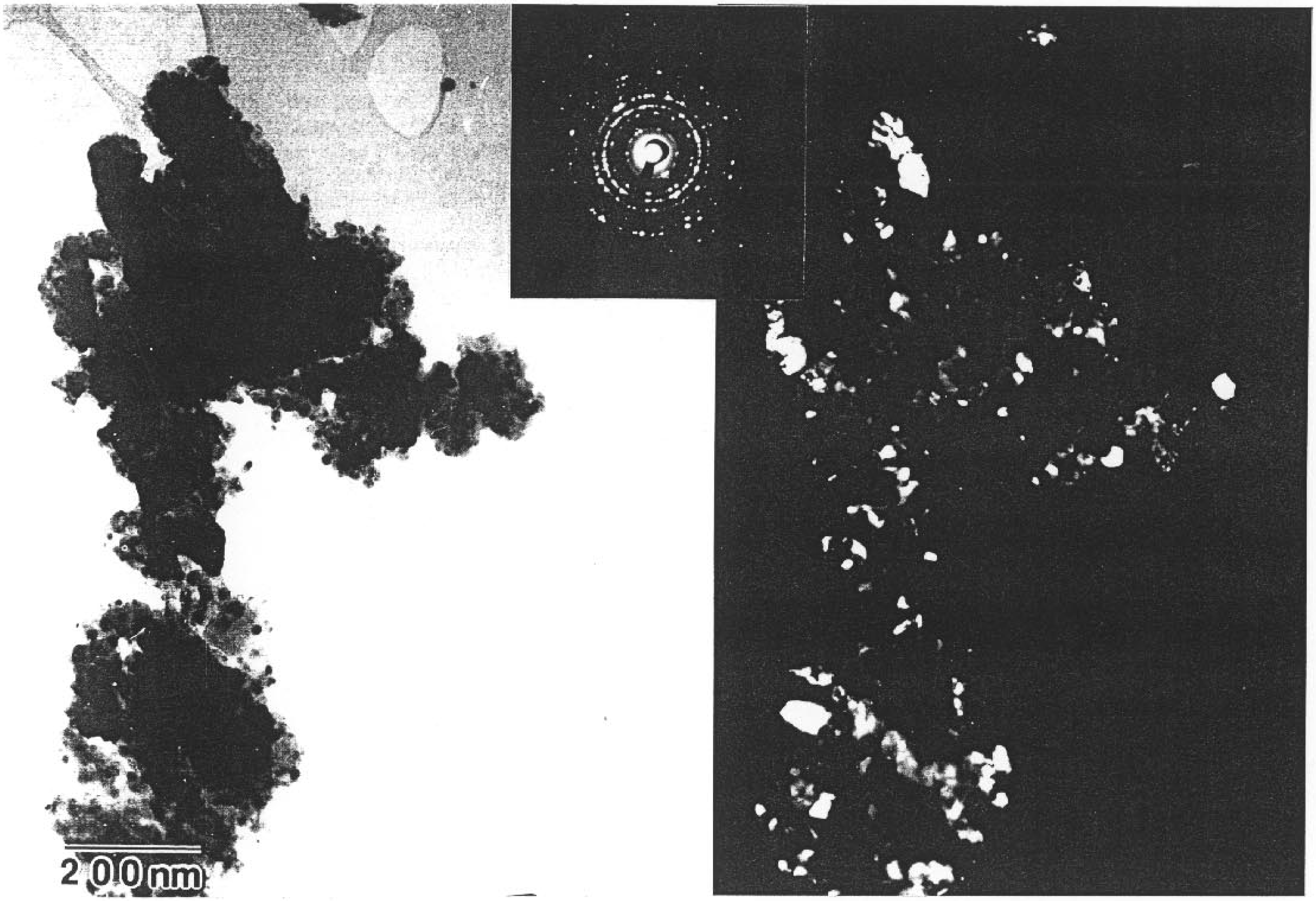
CERIA DEPOSITED ON ALUMINA WITH DIFFERENT AEROSOL SIZE DISTRIBUTIONS





YSZ DEPOSITED VIA COMBUSTION-CVD

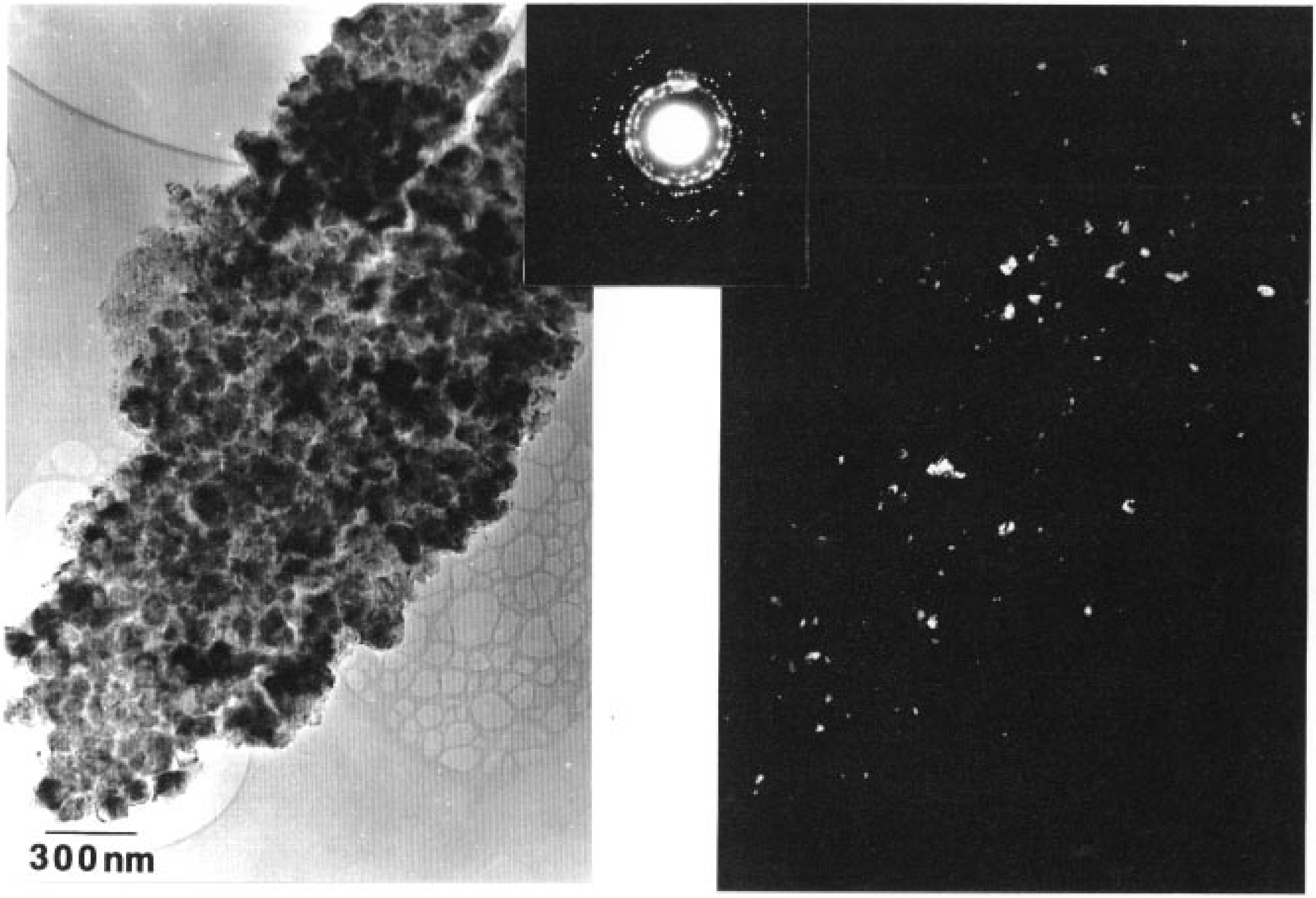
$NiAl_2O_4$ and $MgAl_2O_4$



BRIGHT FIELD DARK FIELD

NICKEL SPINEL DEPOSITED VIA COMBUSTION-CVD

1a



BRIGHT FIELD DARK FIELD

MAGNESIUM SPINEL DEPOSITED VIA COMBUSTION-CVD

Testing of Materials

- Experiments completed
  - furnace cycle testing (FCT) of alumina, ceria and annealed alumina specimens; annealed alumina showed 63% improvement over control samples, microstructural investigation
- Experiments in progress
  - repeat FCT of alumina with and without anneal, to be contrasted with GE anneal and GT anneal; microstructural investigation
- Experiments remaining
  - FCT of YSZ and Ni spinel; corrosion/erosion testing of alumina, Mg-Al spinel, ceria and YSZ overlayer coatings

Summary

All materials which have been targeted in this program for testing as innerlayer and overlayer coatings have been successfully deposited onto test materials using the new technology, combustion CVD. The phases and microstructures of the coatings are reported here, which include nanostructured theta-phase alumina, ceria with columnar morphology, silica in an amorphous state, equiaxed grains of YSZ and nanostructural spinel. The final stage of the research program, now in progress, is to complete the testing of these materials and report the findings.

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