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Slurry Based Coatings on Silicon Based Ceramics

Sameer K. Khaja-Abdul

Cleveland State University

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SLURRY BASED COATINGS ON SILICON BASED CERAMICS

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Bachelor of Science in Chemical Engineering
Cleveland State University
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Submitted in partial fulfillment of requirements for the degree
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This thesis has been approved
for the Department of CHEMICAL AND BIOMEDICAL ENGINEERING
and the College of Graduate Studies by

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Thesis Committee Chairperson, Dr. Surendra N. Tewari

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Dr. Nolan B. Holland

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Department/Date
SLURRY BASED COATINGS ON SILICON BASED CERAMICS

SAMEER K KHAJA-ABDUL

ABSTRACT

The use of advanced structural ceramics, such as Si₃N₄ (SN282), SiC, and SiC/SiC composites, is a promising technology to accomplish the high performance goals for advanced turbine applications. These materials exhibit excellent corrosion resistance by forming a dense silica scale. However, severe degradation of the silica scale has been observed due to the presence of certain environmental species, like alkali salts and water vapors, limiting the long-term reliability of structural ceramics. Presence of water vapor in the turbine environment leads to the formation of a gaseous silicon hydroxide species (Si(OH)₄) via a reaction with silica layer, SiO₂(s) + 2H₂O(g) = Si(OH)₄(g). The Si(OH)₄ is swept away in turbine environments resulting in continued recession of the substrate.

Purpose of this research is to develop environmental barrier coatings (EBCs) for silicon nitride substrates (Si₃N₄ and SN282) that will protect them from water/salt vapor damage and enhance their usage. Slurry dip coating is chosen as the coating method in this research because it is easy, cost-efficient, and suitable to coat on complex shaped ceramic components because of its non-line-of-site nature. Mullite (3A1₂O₃.2SiO₂) is chosen as coating material because it has good mechanical properties at high temperatures, and a coefficient of thermal expansion (CTE) that is
very close to Si$_3$N$_4$ [SN282] (CTE of Mullite is $5.3 \times 10^{-6}$ °C$^{-1}$ and CTE of SiC is $4.7 \times 10^{-6}$ °C$^{-1}$ and Si$_3$N$_4$ [SN282] is $3-4 \times 10^{-6}$ °C$^{-1}$).

Mullite slurry is modified with sintering enhancing additives of rare earth silicates, such as gadolinium ($Gd_2SiO_5$), lutetium ($Lu_2SiO_5$), hafnium ($HfSiO_4$), and erbium ($Er_2SiO_5$). Mullite/rare earth silicate slurry, sintered at high temperatures ($1375°$C – $1450°$C) and coated on Si$_3$N$_4$ and SN282 substrates, did not show good bonding with the substrates. Yttria-alumina containing Si$_3$N$_4$ substrates coated with Mullite slurry and sintered at high temperatures ($1375°$C – $1430°$C) also showed lack of good bonding between the coat and substrate. 100% mullite coating sintered at $1475°$C and 50% mullite sol + 50% mullite powder coating sintered at $1450°$C on SN282 substrate showed good bonding of the coating to the substrate without any cracks at the interface.
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CHAPTER I
INTRODUCTION

Silicon-based ceramics materials, such as silicon carbide (SiC) and silicon nitride (Si₃N₄), are prime candidate materials for elevated temperature applications in advanced gas turbines, heat exchanger tubes, heating elements for industrial furnaces, and structural components in heat engines [1]. Silicon-based ceramics are primarily used in these applications due to their high-temperature mechanical strength, high thermal conductivity, low thermal expansion coefficient, high oxidation resistance, good thermal shock resistance because of their combined high mechanical strength and high thermal conductivity [2]. The excellent oxidation resistance of silicon carbide based ceramics in oxidative environments is due to the formation of a slow growing, dense, adherent silica scale which constitutes a barrier to further oxidation [1]. However, in applications where alkali salts and water vapors are present, severe degradation of the silica scale has been observed limiting the long-term reliability of
silicon based ceramics. Presence of water vapor in the turbine environment leads to the formation of a gaseous silicon hydroxide species (Si(OH)$_4$) via a reaction with silica layer, SiO$_2$(s) + 2H$_2$O(g) = Si(OH)$_4$(g). Figure 1 shows reaction mechanism of silicon component with water vapor in combustion environment. Si(OH)$_4$ is swept away by the exhaust gases in the turbine environments resulting in a continued recession of the silicon carbide or silicon nitride components. The continued silica scale formation and its volatilization lead to the degradation of their mechanical properties and to the loss of their component functions. Carbon present in the SiC composites further accentuates the damage because of the abundant formation of CO gas bubbles [3, 4].

![Figure 1: Reaction mechanism of Silicon component with water vapor in combustion environment. Courtesy: Kirby et al. (Oak Ridge National Laboratory)](image)

Therefore, silicon based ceramics need to be protected from moisture containing high temperature oxidative environments to allow their usage. One method would be to protect the component by applying suitable environmental barrier
coatings (EBC). The coating should not adversely affect the material’s microstructures and mechanical properties; and at the same time protect the substrate from water-vapor recession [5]. Selection of the coating material depends on factors such as their high environmental resistance, coefficient of thermal expansion (CTE) match between the coat and the substrate, chemical compatibility between them, and phase stability [6]. Mullite(3Al$_2$O$_3$.2SiO$_2$) has good resistance to oxidizing and reducing high temperature environments, good mechanical properties at high and low temperatures, and a coefficient of thermal expansion (CTE) close to that of SiC and Si$_3$N$_4$ [SN282] (CTE of Mullite is $5.3 \times 10^{-6} \, ^\circ\text{C}^{-1}$ and CTE of SiC is $4.7 \times 10^{-6} \, ^\circ\text{C}^{-1}$ and Si$_3$N$_4$ [SN282] is $3-4 \times 10^{-6} \, ^\circ\text{C}^{-1}$) [7,8]. The use of components and coatings with similar CTE minimizes thermal stresses generated during high temperature cycling.

There are several different methods to apply the coatings such as plasma-spraying, physical vapor deposition and chemical vapor deposition [9-11], solution-based coating techniques such as sol-gel and slurry-dip coatings. Figure 2 shows various coating techniques available for ceramics materials.
The most widely used process for EBC deposition is the Plasma spraying process. But it has many serious limitations. It is a very difficult and time-consuming to apply an EBC by plasma spray on a complex-shaped component, such as integrated turbine vanes and blades. Plasma-spraying process itself degrades the fracture strength of monolithic Si₃N₄ ceramics, lowering it by as much as 30%. Physical and chemical vapor depositions require high capital cost, and the rate of coating deposition is usually quite slow [12]. Solution-based coating techniques are simple, cost-effective, provide environmental resistance, and are suitable for depositing coatings with mixed compositions. Unlike plasma spraying, this technique allows a better uniform coating on complex shaped components [13-14]. Therefore, solution-based technique is used in this research to coat silicon based ceramic components.
CHAPTER II

PURPOSE

Silicon-based ceramic components have the high temperature strength required for the advanced supersonic cruise propulsion engines. However, their application is possible only if environmental barrier coatings (EBC) can be developed that can protect them from water/salt vapor damage. Two substrates are used in this research, namely: NT-154 Si$_3$N$_4$ (Saint Gobain) and SN282 (Kyocera Corp). Purpose of this research is aimed at developing EBCs for silicon nitride substrates (Si$_3$N$_4$ and SN282) that will provide significantly improved process flexibility and protect substrates from water/salt vapor damage to allow their usage in gas turbine engines. Slurry dip coating is chosen as the coating method in this research because it is easy, cost-efficient, and suitable to coat on complex shaped components.

The slurry preparation consists of ceramic powders; a variety of organic additives like solvents and binders, and sometimes particle dispersants. Due to high
surface area, ceramic particles have a strong tendency to agglomerate. This can be prevented by using dispersant which establishes well-dispersed and stable slurries of fine particles [15]. The main function of the binder is to establish strong bond of the ceramic material after the solvent has been evaporated, without leading to the formation of cracks and defects. Polyvinyl-butryral (PVB) is a commonly used binder, which is soluble in nonpolar media, and decomposes slowly during the burnout period [16]. Preliminary tests in the lab indicate that PVB, phosphate ester (PE), and ethyl alcohol as binder, dispersant, and solvent, respectively, are effective for the slurry dip process of various ceramic materials.

Slurry dip coating formulations aimed at environmental protection should match the coefficient of thermal expansion of the ceramic substrate. The sintered ceramic coat should not have interconnected pores which allow permeation of water vapor. Adequate fracture toughness is required in the coatings to resist crack propagation. Slurry dip coating formulations need an optimum viscosity level. A thin viscosity would not produce enough coverage around the substrate and a higher viscosity may cause nonuniform and thick coating. The viscosity of the slurry can be determined by the following parameters: the particle size/shape, the solid loading of ceramic powders [15].

The keys to successful preparation of slurry dip coatings are (a) well-dispersed particles in the suspension; and (b) avoiding cracking due to shrinkage during the consequent drying and sintering process. Sintering increases the density of the ceramic particles, and decreases the porosity of ceramic. Since the porosity is inversely proportional to mechanical properties, reducing the defect concentration in
a ceramic is required [17]. During the sintering process pores in the ceramic close up as seen in the Fig. 3(a) and (b).

![Fig. 3. Ceramic particles (a) before sintering (b) after sintering [16]](image)

The porosity decreases during high temperature sintering because the grain boundaries are areas of high energy and the ceramic material by getting rid of all its grain boundaries attempts to reach its lowest possible energy state. The thermal energy the furnace gives to the ceramic material allows it to change the size of its grain boundaries; as a result the porosity is decreased [17]. A critical test of EBC is its resistance to degradation during thermal cycling in a moisture containing environment. EBCs that can withstand the water vapor damage can then be further examined for their potential future applications.
CHAPTER III
EXPERIMENTAL PROCEDURE

3.1 - Materials

Materials used for the experimental work include: Mullite, 3Al₂O₃.2SiO₂ (Baikowski International Corporation, $D_{21} = 1 \, \mu m$, $D_{22} = 3 \, \mu m$, surface area: 2.6 $m^2/g$ ), $Gd_2SiO_5$ (Praxair Surface Technologies, $D_{50} = 0.9 \, \mu m$, surface area: 4.86 $m^2/g$ ), $Lu_2SiO_5$ (Praxair Surface Technologies, $D_{50} = 1 \, \mu m$, surface area: 3.34 $m^2/g$ ), $HfSiO_4$ (Praxair Surface Technologies, $D_{50} = 0.7 \, \mu m$, surface area: 4.48 $m^2/g$ ), $Er_2SiO_5$ (Praxair Surface Technologies, $D_{50} = 0.8 \, \mu m$, surface area: 5.25 $m^2/g$ ), ethyl alcohol (Fischer Scientific), polyvinyl-butyral (Sigma, Butvar B-98), and phosphate Ester (Sigma, Triton QS-44 solution).

3.2 - Process

The quality of mixing and uniformity in the initial powder material is crucial in generating high quality ceramic slurry. Here the ceramic powders such as Mullite,
$Gd_2SiO_5$, $Lu_2SiO_5$, $HfSiO_4$, and $Er_2SiO_5$ were ball-milled in a planetary ball-mill. A 20g charge of ceramic powder was ball milled, using two zirconia balls, in two 15 minute cycles. Different ceramic powder combinations used for the slurry preparation in this research are indicated in the table below.

<table>
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<th>Slurry 2</th>
<th>Slurry 3</th>
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<th>Slurry 5</th>
<th>Slurry 6</th>
<th>Slurry 7</th>
<th>Slurry 8</th>
</tr>
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<tr>
<td>94% Mullite pwdr + 6% Er$_2$SiO$_5$</td>
<td>40 g</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>94% Mullite pwdr + 6% Lu$_2$SiO$_5$</td>
<td>-</td>
<td>40 g</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>94% Mullite pwdr + 6% HfSiO$_4$</td>
<td>-</td>
<td>-</td>
<td>40 g</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>88% Mullite pwdr + 12% Gd$_2$SiO$_5$</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>20 g</td>
<td>40 g</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>94% Mullite pwdr + 6% Gd$_2$SiO$_5$</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>40 g</td>
<td>-</td>
</tr>
<tr>
<td>100% Mullite pwdr</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>40 g</td>
</tr>
<tr>
<td>50% Mullite sol [29.5% Al$_2$O$_3$ + 70.5% (CH$_3$)$_4$NOH, 2SiO$_2$] + 50% Mullite pwdr</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>60 g</td>
</tr>
<tr>
<td>Ethyl alcohol</td>
<td>20 g</td>
<td>20 g</td>
<td>20 g</td>
<td>20 g</td>
<td>20 g</td>
<td>20 g</td>
<td>-</td>
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<tr>
<td>PVB</td>
<td>0.8 g</td>
<td>0.8 g</td>
<td>0.8 g</td>
<td>1.2 g</td>
<td>0.8 g</td>
<td>0.8 g</td>
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<td>PE</td>
<td>0.12 g</td>
<td>0.12 g</td>
<td>0.12 g</td>
<td>0.12 g</td>
<td>0.12 g</td>
<td>0.12 g</td>
<td>0.12 g</td>
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Table 1: Slurries prepared for deposition of coatings on Si$_3$N$_4$ and SN282

The slurries were prepared in a 250 ml glass jar as follows: Slurry 1,2,3,5 and 6 were prepared by adding 20 g of ethyl alcohol and 0.12 g of PE. This solution was stirred for one hour using a magnetic stirrer. 40 g of (mullite powder + rare earth...
Silicate powder was added to the solution and stirred for 5 hours. Finally, 0.8 g of PVB was added to the mixture and stirred for 24 hours.

Slurry 4 was prepared by adding 20 g of ethyl alcohol and 0.12 g of PE. This solution was stirred for 1 hour. 20 g of 88% mullite powder + 12% rare earth Silicate powder was added to the solution and stirred for 5 hours. Finally, 1.2 g of PVB was added to the mixture and stirred for 24 hours.

Slurry 6 was prepared by adding 20 g of ethyl alcohol and 0.12 g of PE. This solution was stirred for 1 hour. 40 g of 100% mullite powder was added to the solution and stirred for 5 hours. Finally, 0.8 g of PVB was added to the mixture and stirred for 24 hours.

Slurry 7 was prepared by adding 30 g of mullite sol [29.5% Al₂O₃ + 70.5% (CH₃). 4 NOH. 2SiO₂] and was stirred for 24 hours. 30 g of mullite powder was then added to the sol and stirred for another 24 hours.

After slurry preparation, coating was performed on Si₃N₄ (0.94” × 0.125” × 0.09”) and SN282 (0.94” × 0.125” × 0.09”) substrates. The slurry dip coating process is as follows: the Si₃N₄ and SN282 were vertically dipped in the well-stirred slurry and rotated clockwise and anti-clockwise for uniform coating. The substrates were then withdrawn after an approximate time of 30 seconds.
After the substrates were coated, they were held vertically and dried in a vacuum chamber at room temperature for 3 hours. The vacuum environment helps remove entrapped air bubbles from the coating. The vacuum dried coated substrates were then sintered in the furnace (Deltech inc.) with maximum temperature capability of 1600°C. Sintering transforms the porous coating layer into a strong and dense coating. The sintering schedule with different heating and cooling rates are indicated in Fig. 5.

Fig. 5. Temperature and time profile for the furnace.
The heating and cooling schedule is divided into three regions. In Region 1, the heating was started from 25 °C to 500 °C, and the temperature stayed constant at 500 °C for 3h. Binders and dispersants mixed in the slurry will evaporate near 500 °C. Region 2 represents sintering zone where the temperature was increased from 500 °C to 1375-1550 °C at a rate of 10 °C/min, and the temperature stayed constant at 1375-1550 °C for 3h. Region 3 represents the cooling zone where the temperature decreases from 1375-1550 °C to 40 °C at a rate of 5 °C/min.

3.3 - Characterization

To maintain edge retention and flatness, the sintered substrates were cut and mounted in epoxy resin (Buehler). The mounted samples were polished as per the procedure indicated in Table 2. The microstructure of the coating is then characterized by scanning electron microscope (SEM).

<table>
<thead>
<tr>
<th>Surface</th>
<th>Abrasive / Size</th>
<th>Load (Lb/sample)</th>
<th>Base speed / rotation</th>
<th>Time (min:sec)</th>
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</thead>
<tbody>
<tr>
<td>Ultra-prep diamond grinding disc</td>
<td>75-μm diamond disc</td>
<td>5</td>
<td>120*</td>
<td>40:00</td>
</tr>
<tr>
<td>Ultra-prep diamond grinding disc</td>
<td>35-μm diamond disc</td>
<td>5</td>
<td>120*</td>
<td>20:00</td>
</tr>
<tr>
<td>Buehler consumables, 8” UltraPol cloths</td>
<td>9-μm Struers diamond suspension</td>
<td>5</td>
<td>100*</td>
<td>5:00</td>
</tr>
<tr>
<td>Buehler consumables, 8” Trident polishing cloths</td>
<td>3-μm Struers diamond suspension</td>
<td>5</td>
<td>100*</td>
<td>5:00</td>
</tr>
<tr>
<td>Buehler consumables, 8” Apex DGD cloths</td>
<td>0.05-μm Buehler Masterprep diamond suspension</td>
<td>5</td>
<td>100*</td>
<td>5:00</td>
</tr>
</tbody>
</table>

Table 2: Five-step Procedure for polishing ceramic materials
* Platen and specimen holder both rotate in the opposite direction
After SEM characterization, the substrates were thermally cycled in an experimental set-up shown in Figure 6. This is a simulated combustion environment of 90% H$_2$O vapor- 10% O$_2$. A total of 100 cycles (1 h cycle) were performed where the sample was kept at 1350 °C for one hour and then was quickly moved to room temperature during each cycle. After thermal cycling, the substrates and coatings were characterized by SEM.

Fig.6. Thermal Cycling Set up
CHAPTER IV
RESULTS

Si$_3$N$_4$ (Y$_2$O$_3$, Al$_2$O$_3$) and SN282 (Lu$_2$O$_3$) substrates were coated with mullite based slurries. Compositions of these slurries were varied with the addition of silicates like $Gd_2SiO_5$, $Lu_2SiO_5$ $HfSiO_4$ and, $Er_2SiO_5$ by weight percent. Table 1 lists the slurries prepared for coating purposes.

4.1 - Si$_3$N$_4$ (Y$_2$O$_3$, Al$_2$O$_3$)

Si$_3$N$_4$ coated with slurry containing 94% Mullite powder - 6% Er$_2$SiO$_5$ (Slurry 1)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% Er$_2$SiO$_5$), and 0.8g of PVB. Si$_3$N$_4$ substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C.
Fig. 7: Optical observation of Si$_3$N$_4$ substrate coated with 94% Mullite + 6% Er$_2$SiO$_5$.

(a) Sintered at 1375 °C - 1400 °C. (b) Sintered at 1375 °C - 1400 °C - 1430 °C.

Figures 7(a) and (b) show bubbles on the surface at 1400 °C causing a complete peel of the coating from the substrate at 1430 °C. Therefore, 1400 °C and 1430 °C are too hot for Si$_3$N$_4$ coated with 94% Mullite powder + 6% Er$_2$SiO$_5$.

Si$_3$N$_4$ coated with slurry containing 94% Mullite powder - 6% Lu$_2$SiO$_5$: (Slurry 2)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% Lu$_2$SiO$_5$) and, 0.8g of PVB. Si$_3$N$_4$ substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C.
Fig. 8: SEM observation of Si$_3$N$_4$ substrate coated with 94% Mullite + 6% Lu$_2$SiO$_5$ (a) and (b) Sintered at 1375 °C - 1400 °C - 1430°C.

Figures 8(a) and (b) show that the coating has no contact at the ceramic-coat interface. This indicates that 1430 °C is too hot for Si$_3$N$_4$ coated with 94% Mullite + 6% Lu$_2$SiO$_5$.

Si$_3$N$_4$ coated with slurry containing 94% Mullite powder - 6% HfSiO$_4$. (Slurry 3)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% HfSiO$_4$) and, 0.8g of PVB. Si$_3$N$_4$ substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C to 1450 °C.
Fig. 9: Optical observation of Si$_3$N$_4$ substrate coated with 94% Mullite+ 6% HfSiO$_4$ (a) and (b) Sintered at 1375 °C - 1400 °C - 1430°C.

Fig. 10: SEM observation of Si$_3$N$_4$ substrate coated with 94% Mullite+ 6% HfSiO$_4$ (a) and (b) Sintered at 1375 °C - 1400 °C - 1430°C.

Figures 9 (a) and (b) show that there are no sintering cracks present on the coated surface. However, there are a few cracks present on the beveled region. There are no bubbles present on the surface either. Figures 10 (a) and (b) show that the
coating has no contact with the interface. Also there is no contact between the layers of the coating.

Fig.11 shows that the coating has bubbles present on the chamfered region. Therefore, 1430°C & 1450 °C is too hot for Si₃N₄ coated with 94% Mullite powder + 6% HfSiO₄.

![Fig.11: Optical observation of Si₃N₄ substrate coated with 94% Mullite+ 6% HfSiO₄. Sintered at 1375 °C -1400 °C -1430°C-1450°C](image)

**Si₃N₄ coated with slurry containing 88% Mullite powder - 12% Gd₂SiO₅** *(Slurry 4)*

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 20g of ball milled powder (88% Mullite powder + 12% Gd₂SiO₅) and, 0.8g of PVB. Si₃N₄ substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C. Samples were then thermally cycled at 1350°C in 90% water vapor – 10% O₂ environment for 100 h (1 h cycle).
Figures 12 (a) and (b) show that the coating has elongated pores present near the interface. Since the coating itself is dense, these samples were thermally cycled.

Figures 13 (a) and (b) shows that the coating is debonded and has no contact with the interface. Therefore, 1350 °C is too hot for Si$_3$N$_4$ coated with 88% Mullite powder + 12% Gd$_2$SiO$_5$. 
Si$_3$N$_4$ coated with slurry containing 88% Mullite powder-12% Gd$_2$SiO$_5$. *Pre-oxidized (Slurry 5)*

Si$_3$N$_4$ substrate was pre-oxidized in air at 1300 °C for 5h. This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (88% Mullite powder + 12% Gd$_2$SiO$_5$) and, 0.8g of PVB. Si$_3$N$_4$ substrates were then coated and sintered. The sintering was done twice at 1375 °C.

Fig.14: Optical observation of pre-oxidized Si$_3$N$_4$ substrate coated with 88% Mullite + 12% Gd$_2$SiO$_5$ (a) and (b) Sintered at 1375 °C -1375 °C.

Figures 14 (a) and (b) show that the coating has lots of sintering cracks and bubbles present on the surface and the beveled region. This implies that 1375 °C is too hot for pre-oxidized Si$_3$N$_4$ coated with 88% Mullite powder + 12% Gd$_2$SiO$_5$.

Si$_3$N$_4$ coated with slurry containing 94% Mullite powder-6% Gd$_2$SiO$_5$. *Pre-oxidized (Slurry 6)*

Si$_3$N$_4$ substrate was pre-oxidized in air at 1400 °C for 5h. This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% Gd$_2$SiO$_5$) and, 0.8g of PVB. Si$_3$N$_4$ substrates were then
coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1415 °C.

Figures 15(a) and (b) show that the coating has lots of sintering cracks and bubbles present on the surface and the chamfered region. This implies that 1415 °C is too hot for pre-oxidized Si$_3$N$_4$ coated with 94% Mullite powder + 6% Gd$_2$SiO$_5$.

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% Gd$_2$SiO$_5$) and, 0.8g of PVB. Si$_3$N$_4$ substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1415 °C.
Fig. 16: Optical observation of Si$_3$N$_4$ substrate coated with 94% Mullite+ 6% Gd$_2$SiO$_5$ (a) and (b) Sintered at 1375 °C -1400 °C-1415 °C.

Fig. 17: SEM observation of Si$_3$N$_4$ substrate coated with 94% Mullite+ 6% Gd$_2$SiO$_5$ (a) and (b) Sintered at 1375 °C -1400 °C-1415 °C.

Figures 16 (a) and (b) show that the coating has sintering cracks present on the surface. There are few bubbles present on the chamfered region. Figures 17 (a) and (b) shows that the coating has no contact with the interface. This indicates that 1415°C is too hot for Si$_3$N$_4$ coated with 94% Mullite powder + 6% Gd$_2$SiO$_5$.
**Si₃N₄ coated with slurry containing 100% Mullite powder: (Slurry 7)**

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled Mullite powder and, 0.8g of PVB. Si₃N₄ substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C to 1450 °C.

![Fig.18: Optical observation of Si₃N₄ substrate coated with 100% Mullite (a) Sintered at 1375 °C - 1400 °C. (b) Sintered at 1375 °C - 1400 °C - 1430 °C.](image)

![Fig.19: SEM observation of Si₃N₄ substrate coated with 100% Mullite (a) and (b) Sintered at 1375 °C - 1400 °C - 1430 °C.](image)
Figures 18(a) and (b) show that the coating surface was smooth until 1400 °C but at 1430 °C there were cracks present on the surface. One of the reasons for these sintering cracks present on the surface is the high viscosity of the slurry. This high viscous nature of the slurry caused an increase in the thickness of the coating. Figures 19(a) and (b) shows that the coating has no contact at the interface.

Figure 20 shows that the coating has cracks present on the surface. Therefore, 1430°C and 1450 °C is too hot for Si₃N₄ coated with 100% Mullite powder.

Si₃N₄ coated with slurry containing 50% Mullite sol and 50% Mullite powder:

*(Slurry 8)*

This slurry was prepared by adding 30 g of Mullite sol [29.5 % Al₂O₃ + 70.5% (CH₃). 4 NOH. 2SiO₂] and 30 g of ball milled Mullite powder. Si₃N₄ substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1415 °C to 1430 °C.
Based on the optical observation, Fig. 21, the coating looked smooth. There were few hairlines cracks and few bubbles on the coated surface.

Fig. 21: Optical observation of Si$_3$N$_4$ substrate coated with 50% Mullite sol & 50% Mullite powder. Sintered at 1375 °C - 1400 °C - 1415 °C.

Based on the SEM observations, Figures 22(a) and (b), there are fine pores present throughout the coating. Most areas of the coating appear to have contact at the interface. Also there are few cracks present at the interface. This microstructure looks attractive for thermal cycling and fracture toughness.

Fig. 22: SEM observation of Si$_3$N$_4$ substrate coated with 50% Mullite sol & 50% Mullite powder (a) and (b) Sintered at 1375 °C - 1400 °C - 1415 °C.
However after 1430°C, the coating showed many bubbles present on the surface and the chamfered region as shown in Fig.23.

Fig. 23: Optical observation of Si$_3$N$_4$ substrate coated with 50% Mullite sol & 50% Mullite powder. Sintered at 1375°C -1400°C -1415°C-1430°C.

Figures 24 (a) and (b) show that the coating has no contact at the interface. This implies that 1430 °C is too hot for Si$_3$N$_4$ coated with 50% mullite sol and 50% mullite powder. Therefore, the maximum sintering temperature for Si$_3$N$_4$ coated with this slurry to have a good bonding at the interface is 1415 °C.
4.2 - SN282 (Lu$_2$O$_3$)

SN282 coated with slurry containing 94% Mullite powder - 6% Er$_2$SiO$_5$ (Slurry 1)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% Er$_2$SiO$_5$) and, 0.8g of PVB. SN282 substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C.

Figure 25 shows that the coating has peeled off the surface. Therefore, 1430 °C is too hot for SN282 coated with 94% Mullite powder + 6% Er$_2$SiO$_5$.

![Optical observation of SN282 substrate coated 94% Mullite+ 6% Er$_2$SiO$_5$ Sintered at 1375 °C -1400 °C -1430°C.](image)

SN282 coated with slurry containing 94% Mullite powder - 6% Lu$_2$SiO$_5$ (Slurry 2)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% Lu$_2$SiO$_5$) and, 0.8g of PVB. SN282 substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C.
Fig. 26: Optical and SEM observation of SN282 substrate coated with 94% Mullite + 6% Lu₂SiO₅ (a) and (b) Sintered at 1375 °C -1400 °C-1430°C. 

Figure 26(a) shows that the coating is smooth. There are few cracks present on the chamfered region and on the surface. Figure 26(b) shows that the coating itself was very porous. Most areas of the coating appear to have good contact at the interface. Figure 27 shows that the coating has sintering cracks present on the surface.

Fig 27: Optical observation of SN282 substrate coated 94% Mullite + 6% Lu₂SiO₅ Sintered at 1375 °C -1400 °C -1430°C-1450°C.
Figures 28(a) and (b) show that the coating has no contact at the interface. This implies that 1450 °C is too hot for SN282 coated with 94% Mullite powder + 6% $Lu_2SiO_5$. Therefore, the maximum sintering temperature for SN282 coated with this slurry to have a good bonding at the interface is 1430 °C.

SN282 coated with slurry containing 94% Mullite powder - 6% $HfSiO_4$ (Slurry 3)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% $HfSiO_4$) and, 0.8g of PVB. SN282 substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C.
Fig. 29: Optical and SEM observation of SN282 substrate coated with 94% Mullite + 6% HfSiO₄ (a) and (b) Sintered at 1375 °C - 1400 °C - 1430°C.

Figure 29(a) shows that the coating surface is smooth. Figure 29(b) shows that the coating has no contact at the interface. This implies that 1430 °C is too hot for SN282 coated with 94% Mullite powder + 6% HfSiO₄.

SN282 coated with slurry containing 88% Mullite powder - 12% Gd₂SiO₅ (Slurry 4)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 20g of ball milled powder (88% Mullite powder + 12% Gd₂SiO₅) and, 0.8g of PVB. SN282 substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C. Samples were then thermally cycled at 1350°C in 90% water vapor – 10% O₂ environment for 100 h (1 h cycle).
Fig. 30: SEM observation of SN282 substrate coated with 88% Mullite + 12% Gd$_2$SiO$_5$ (a) and (b) Sintered at 1375 °C - 1400 °C.

Fig. 31: SEM observation of SN282 substrate coated with 88% Mullite + 12% Gd$_2$SiO$_5$ (a) and (b) Thermally cycled at 1350 °C.

Figures 30 (a) and (b) show that the coating is porous near the interface and dense at the top coat. The coating bond to the substrate before thermal cycling has discontinued traces of hair line cracks seen in the interface region. Figures 31 (a) and
(b) shows that the coating has no contact with the interface. Therefore, 1350 °C is too hot for SN282 coated with 88% Mullite powder + 12% $Gd_2SiO_5$.

**SN282 coated with slurry containing 94% Mullite powder - 6% $Gd_2SiO_5$ (Slurry 6)**

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 40g of ball milled powder (94% Mullite powder + 6% $Gd_2SiO_5$) and, 0.8g of PVB. SN282 substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C.

![Fig.32](image)

Fig.32: Optical and SEM observation of SN282 substrate coated with 94% Mullite+ 6% $Gd_2SiO_5$ (a) and (b) Sintered at 1375 °C - 1400 °C-1430 °C.

Figure 32 (a) shows that the coating surface is smooth. Figure 32 (b) shows that the coating has no contact at the interface. This implies that 1430 °C is too hot for SN282 coated with 94% Mullite powder + 6% $Gd_2SiO_5$. 
SN282 coated with slurry containing 100% Mullite powder: (Slurry 7)

This slurry was prepared by adding 20g of Ethyl alcohol, 0.12g of PE, 35g of ball milled Mullite powder and, 0.8g of PVB. SN282 substrates were then coated and sintered. The sintering temperature was increased in steps from 1450 °C to 1475 °C to 1500 °C.

Figure 33 shows that the coating surface is smooth. In addition there are no sintering cracks present on the coating. Figures 34 (a) and (b) shows that the coating is quite dense. Most areas of the coating appear to have good contact at the interface.

Fig 33: Optical observation of SN282 substrate coated with 100% Mullite powder Sintered at 1450 °C-1475 °C.

Fig.34: SEM observation of SN282 substrate coated with 100% Mullite powder (a) and (b) Sintered at 1450 °C -1475 °C.
Fig. 35: Optical and SEM observation of SN282 substrate coated with 100% Mullite powder (a) and (b) Sintered at 1450 °C - 1475 °C-1500 °C.

Figure 35(a) shows that the coating has some bubbles in the chamfered region and sintering cracks present on the surface. Fig 35(b) shows that some regions of the coating have no contact at the interface. Also there are cracks present inside the coating. This implies that 1500 °C is too hot for SN282 coated with 100% mullite powder. Therefore, the maximum sintering temperature for SN282 coated with this slurry to have a good bonding at the interface is 1475 °C.

SN282 coated with slurry containing 50% Mullite sol and 50% Mullite powder: (Slurry 8)

This slurry was prepared by adding 30 g of Mullite sol [29.5 % Al₂O₃ + 70.5% (CH₃). 4 NOH. 2SiO₂] and 30 g of ball milled Mullite powder. SN282 substrates were then coated and sintered. The sintering temperature was increased in steps from 1375 °C to 1400 °C to 1430 °C to 1450 °C to 1475 °C.
Fig. 36: Optical and SEM observation of SN282 substrate coated with 50% Mullite sol & 50% Mullite powder. (a) and (b) Sintered at 1375 °C - 1400 °C - 1430 °C.

Figure 36(a) shows that there are sintering cracks present on the coating.

Figure 36(b) shows that there are fine pores present throughout the coating. Most areas of the coating appear to have contact at the interface.

Fig. 37: Optical and SEM observation of SN282 substrate coated with 50% Mullite sol & 50% Mullite powder. (a) and (b) Sintered at 1375 °C - 1400 °C - 1430 °C - 1450 °C.
Figure 37(a) shows that the coating surface is smooth. There are few sintering cracks present on the coating. Figure 37(b) shows that the coating is denser compared to figure 36(b). Most areas of the coating appear to have good contact at the interface.

Figure 38(a) shows that the coating has some bubbles and sintering cracks present on the coating. Figure 38(b) shows that the coating does not have a good bonding at the interface compared to 1450 °C. This implies that 1475 °C is too hot for SN282 coated with 50% mullite powder and 50% mullite sol. Therefore, the maximum sintering temperature for SN282 coated with this slurry to have a good bonding at the interface is 1450 °C.
Mullite is selected as the main coating material because of its coefficient of thermal expansion match with silicon nitride substrates. However, it requires very high temperature to sinter and become dense and impermeable for water-vapor transport. To densify the coat at low sintering temperatures and become impervious, rare earth monosilicates are added. These silicates are chosen based on their CTE match with silicon nitride substrates and their own moisture resistance. Schematic diagram of the silicon nitride substrate (Si₃N₄ [Y₂O₃, Al₂O₃] and SN282 [Lu₂O₃]) and a coating made up of rare earth silicate containing mullite particles is shown in Figure 39; the images shown in (a), (b) and (c) correspond to sintering at increasingly higher temperatures.
At high temperatures, ceramic particles sinter and this increases coating density and decreases its porosity. Figure 39(a) shows that at lower sintering temperatures continuous porosity and cracks are present throughout the coating. This microstructure makes it easier for oxygen present in the environment to reach the substrate and form silica scale. Silica scale then reacts with impurities present in silicon nitride substrates such as yttria (Y$_2$O$_3$), alumina (Al$_2$O$_3$) and lutetium oxide (Lu$_2$O$_3$) and forms low melting point phases. Such coating with continuous porosity is undesirable for moisture resistance and fracture toughness. If the sintering temperature is increased significantly in order to densify the coating and make it impervious, then NO$_2$ and SiO$_2$ gases present in the silicon nitride substrate cannot escape through the dense coating and tend to form bubbles. These bubbles causes
coating lift-off from the substrate and interface debonding as shown in Fig. 38(c). Dense coating also forms thermal expansion related cracks at the substrate-coat interface. Figure 39(b) shows that at some intermediate sintering temperature the coating would have good bonding at the interface, it would have uniformly distributed, but isolated, pores in its microstructure resulting in improved fracture toughness and crack resistance, and it would not have bubble formation. The aim of this research has been to find an optimum combination of coating composition (ratio and type of rare earth silicate in the mullite-silicate slurry), sintering temperatures and times that would yield a sintered coating microstructure similar to that shown in Fig. 39(b).

Table 3 presents a summary of the results obtained using various mullite/rare earth silicate coatings on two silicon nitride substrates examined in this study: the alumina-yttria containing silicon nitride (Si₃N₄) and lutetia containg silicon nitride (SN282).
<table>
<thead>
<tr>
<th>Slurry</th>
<th>Substrate</th>
<th>Temperature (°C)</th>
<th>Results</th>
</tr>
</thead>
<tbody>
<tr>
<td>94% Mullite pwdr + 6% $Er_2SiO_5$</td>
<td>$Si_3N_4$</td>
<td>1400</td>
<td>Coating is smooth*</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1430</td>
<td>Coating peels off the substrate*</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1430</td>
<td>Coating completely peels off the substrate**</td>
</tr>
<tr>
<td>94% Mullite pwdr + 6% $Lu_2SiO_5$</td>
<td>$Si_3N_4$</td>
<td>1430</td>
<td>Dense coating but has no contact at the interface**</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1430</td>
<td>Porous coating w/ less contact at the interface***</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1450</td>
<td>Dense coating but has no contact at the interface***</td>
</tr>
<tr>
<td>94% Mullite pwdr + 6% $HfSiO_4$</td>
<td>$Si_3N_4$</td>
<td>1430</td>
<td>Coating has no contact at the interface**</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1430</td>
<td>Porous coating w/ no contact at the interface***</td>
</tr>
<tr>
<td>88% Mullite pwdr + 12% $Gd_2SiO_5$</td>
<td>(-)$Si_3N_4$</td>
<td>1350</td>
<td>Dense coating with elongated pores but has no contact at the interface***</td>
</tr>
<tr>
<td></td>
<td>(-)SN282</td>
<td>1350</td>
<td>Porous coating w/ no contact at the interface**</td>
</tr>
<tr>
<td>94% Mullite pwdr + 6% $Gd_2SiO_5$</td>
<td>$Si_3N_4$</td>
<td>1415</td>
<td>Dense coating but has no contact at the interface***</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1430</td>
<td>Porous coating w/ no contact at the interface**</td>
</tr>
<tr>
<td>100 % Mullite pwdr</td>
<td>$Si_3N_4$</td>
<td>1430</td>
<td>Porous coating but no contact at the interface**</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1475</td>
<td>Dense coating w/ good contact at the interface***</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1500</td>
<td>Dense coating but has no contact at the interface***</td>
</tr>
<tr>
<td>50 % Mullite sol [29.5 % $Al_2O_3$ + 70.5% (CH$_3$). 4 NOH. 2SiO$_2$ ] + 50 % Mullite pwdr</td>
<td>$Si_3N_4$</td>
<td>1415</td>
<td>Continuous pores in the coating</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1430</td>
<td>Dense coating but has no contact at the interface***</td>
</tr>
<tr>
<td></td>
<td>SN282</td>
<td>1450</td>
<td>Dense coating w/ good contact at the interface***</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1475</td>
<td>Dense coating but has no contact at the interface***</td>
</tr>
</tbody>
</table>

Table 3: Mullite/rare earth silicates coating results on silicon nitride substrates.

* Bubbles observed  
** Cracks observed  
*** Bubbles & Cracks observed  
- Thermally cycled
5.1 - 94% Mullite powder + 6% Er₂SiO₅ (Slurry 1)

Rare earth monosilicates with relatively low CTE also happen to show high temperature phase stability [14]. Erbium silicate (Er₂SiO₅) was selected because of its low CTE (5-7 ×10⁻⁶ °C⁻¹) as compared to Si₃N₄ (3-4 ×10⁻⁶ °C⁻¹). At 1400 °C (1 h cycles) mullite/Er₂SiO₅ coating on silicon carbide (SiC) substrate after 46 h in 90% H₂O–balance O₂ turned into a layer of erbium-aluminum-silicate bubbles making it unsuitable for temperatures around 1400 °C or higher as reported in Ref [14]. Addition of erbium silicate (Er₂SiO₅) to mullite is expected to reduce the sintering temperature of mullite. Figure 7(a) showed that the coated surface is smooth with few bubbles on the chamfered sides when Si₃N₄ was sintered at 1400 °C. Bubbles are formed due to nitrogen oxide gas forming during high temperature sintering in oxygen environment at the interface. Since NO₂ cannot escape through the glassy viscous coating layer on top, it produces entrapped bubbles at temperature as low as 1400 °C. When the sintering temperature was increased to 1430 °C, the coating peeled off the substrate due to thermal mismatch as shown in Fig. 7(b). The inadequate CTE matching with the substrate is responsible for completely peeled off coating from the SN282 substrate at temperature as low as 1430 °C as shown in Figure 25. Erbium silicate was added to the mullite slurry to densify coating and become impervious at low sintering temperature. But erbium silicate combined with the yttria-alumina (Y₂O₃, Al₂O₃) and lutetium oxide (Lu₂O₃) impurities present in the silicon nitride substrate to form lower melting point glassy layer covering the substrate. This layer did not allow the gases to escape and yielded bubbling and lack of coat bonding with the substrate. Therefore, to prevent bubble formation and to
establish good bonding of the coating with the substrate, another silicate was considered for coating silicon nitride substrates.

**5.2 - 94% Mullite powder + 6% Lu₂SiO₅ (Slurry 2)**

Lutetium silicate (Lu₂SiO₅) was selected as rare earth silicate because its CTE is very close to that of silicon nitride ceramics [18]. At 1300 °C Lu₂SiO₅/ Lu₂SiO₇ coating on SN282 substrate after 500 hours in steam jet resulted in the oxidation of the silicon nitride substrate and the corrosion and decomposition of the Lu₂SiO₇ phase during the test[19]. Figures 8(a) and (b) showed that the coating is dense with uniform thickness but for this combination also there was no bonding of the coating to the Si₃N₄ substrate after 1430 °C sintering. Dense coating caused the thermal expansion related cracks at the substrate-coat interface.

The optical view (Figure 26(a)) of SN282 substrate showed the coating to be smooth with some cracks on the surface at 1430 °C. However, cross-sectional SEM observation revealed coating to be very porous with lots of microcracks as seen in Fig.26 (b). Such microstructure would make it easier for oxygen to reach the substrate to form silica scale and further accentuate damage to the substrate. Since the coating had good contact at the interface, the sintering temperature was increased to 1450 °C to densify the coating further. After this treatment the optical view did not show any problem (Fig.27). However, SEM observations revealed that coating was dense but it had no bonding with the substrate. Since the sintering temperature was increased to make the coating dense, the NO₂ formed in the silicon nitride substrate cannot escape forming bubbles; these bubbles cause coating lift-off from the substrate as seen in Figs. 28 (a) and (b). The coating had longitudinal cracks and did not have a good
bonding to the substrate, which must be due to the addition of Lu$_2$SiO$_5$ as it acts as sintering aid and lowers the sintering temperature.

5.3 - 94% Mullite powder + 6% HfSiO$_4$ (Slurry 3)

It has been reported that after 300 cycles in 90% H$_2$O – balance O$_2$ from 1380°C (1 h cycle) the mullite/yttria-stabilized HfO$_2$ coating on Si$_3$N$_4$ substrate showed severe delaminated at the interface. It was suggested that it was due to thermal mismatch between yttria-stabilized HfO$_2$ and the substrate leading to no contact at the interface [14]. Hafnium silicate (HfSiO$_4$) was chosen as an additive with mullite as coating material because it has low thermal conductivity and CTE (~ 10×10^{-6} °C) close to Si$_3$N$_4$ [14]. Si$_3$N$_4$ substrate was coated with slurry 3 and sintered at 1430°C. Optical view of the coated substrate (Fig. 9(a)) showed the coating to be smooth with cracks present, but the SEM view (Fig. 9(b)) again showed lack of chemical bonding between the coating and the substrate. Due to the thermal mismatch between the substrate and the coating debonding occurs at the interface. Longitudinal cracks at the interface would cause substrate oxidation and spallation during thermal cycling.

Optical view of the same coating on the SN282 substrate did not show any problems after sintering at 1430°C as the coating was smooth with no cracks (Fig. 29(a)). But again the SEM observation (Fig. 29(b)) revealed a lack of bonding between the substrate and the coating. This makes addition of HfSiO$_4$ to the mullite also an ineffective EBC formulation.
5.4 - 88% Mullite powder + 12% Gd$_2$SiO$_5$ (Slurry 4 & 5) and 94% Mullite powder + 6% Gd$_2$SiO$_5$ (Slurry 6)

Gadolinium silicate (Gd$_2$SiO$_5$) was selected as the coating material because of its low CTE [15]. GdSiO$_4$ and mullite slurry with ethanol, and PVB resulted in good coating on SiC [15]. At 1400 °C, Si$_3$N$_4$ substrate coated with slurry 4 showed that there were elongated pores/openings at the substrate-coating interface (Figs. 12(a) and (b)). The coating was dense with minimal porosities and it might be due to reaction of slurry with the Y$_2$O$_3$/Al$_2$O$_3$ dopants present in the substrate that reduce the sintering temperature. Since the coating was dense, this substrate was examined further for its response to thermal cycling in a moisture containing environment. SEM observation of the sample after one hundred thermal cycles (one hour at 1350 °C and ten minutes at room temperature in a 90% oxygen-10% water vapor environment) showed that there was no contact between the coating and the substrate (Figs.13 (a) and (b)). In addition cracks were present at the coating-substrate interface, possibly due to thermal expansion mismatch. Accumulated/segregated voids were present in the damage zone – coating interface region leading to the failure of coating adhesion to the substrate.

To get a better contact of the coating with the interface, the Si$_3$N$_4$ substrate was then pre-oxidized in air for 5 h at 1300 °C and coated with slurry 5. The substrate was supposed to form an oxide layer consisting of yttria/silica/alumina layer. The oxide layer was supposed to have a good reaction with the coating to eliminate the coating-interface bonding problems. The optical view (Figs.14 (a) and (b)) of the Si$_3$N$_4$ substrate showed that bubbles were formed on the surface at 1375 °C. This
must be due the reaction of the gadolinium silicate in the slurry with the oxide layer to form a very low melting point constituent that caused the bubbles to form.

To eliminate the bubbles on the surface, the gadolinium silicate content was decreased from 12% to 6% and the Si₃N₄ substrate was again pre-oxidized in air for 5 h at 1400 °C and coated with slurry 6. It did allow one to use higher sintering temperatures without bubble formation; bubbles now formed on the surface at 1415°C as seen in Figs.15 (a) and (b).

These observations suggest that regardless of decreasing the gadolinium silicate content, bubbles still form on the surface and pre-oxidization of the substrate to improve coating substrate bonding will not work for Si₃N₄ substrate.

The, Si₃N₄ substrates were then coated with 94% Mullite powder + 6% Gd₂SiO₅ (slurry 6). In the optical view (Figs.16 (a) and (b)), sintering cracks, a sure sign of significant coating densification were observed. The SEM observation (Figs.17 (a) and (b)), showed the coating to be dense but it had no contact with the substrate. It is likely that a reaction between the ceramics in the slurry with the yttria-alumina additives in the substrate is responsible to allow the bubble formation; leading to coating to lift-off the interface.

At 1400°C, similar coating experiments were performed on SN282 substrate with slurry 4 as with the Si₃N₄ substrates. SEM micrographs (Figs. 30(a) and (b)) showed discontinued traces of hair line cracks at the interface region. Since the coating was dense with uniform pores, the samples were again examined for thermal cycling at 1350°C. SEM pictures show that there was a coating-substrate de-bonding after thermal cycling for this SN282 substrate, similar to the Si₃N₄ discussed earlier.
The damages for SN282 substrate were more severe than for the Si₃N₄ substrate and most of the coating had peeled off after thermal cycling (Figures 31(a) and (b)). A reaction between lutetium oxide additives in the substrate with the slurry must be responsible for this coating debonding. The coating portion that remained bonded to the substrate also had frequent cracks; accumulated/segregated voids as seen for the Si₃N₄ substrate were also seen for SN282.

To eliminate the coating-interface problem and to increase porosity, the gadolinium silicate content was decreased from 12% to 6%. However, the SEM observation (Fig.32 (b)) showed that the interface debonding still occurs at 1430 °C. Regardless of decreasing the gadolinium silicate content, coating-interface problems still occur. This makes addition of Gd₂SiO₅ to the mullite as an unsuitable for a successful EBC.

The above observations showed that addition of rare earth silicates like Gd₂SiO₅, Lu₂SiO₅, HfSiO₄, and Er₂SiO₅ to the mullite slurry allowed the coat to densify and become resistant to water vapor damage at low sintering temperatures. However, the mullite/silicate coatings on the silicon nitride substrates showed thermal expansion related micro-cracks and bubbles at the substrate-coat interface resulting in coating de-bonding. In order to eliminate this coating-interface de-bonding with a hope to increase the sintering temperature and promote good chemical bonding at the interface, silicon nitride substrates were then coated with mullite slurries only without any rare earth silicates.
5.5 - 100% Mullite (Slurry 7)

Mullite (3Al₂O₃·2SiO₂) has attracted the most interest as an EBC candidate for silicon nitride ceramics because of its low CTE (5.3×10⁻⁶ °C), chemical compatibility with Si-based ceramics, and phase stability [20]. Plasma sprayed mullite coating on SiC substrate after 200 thermal cycles (one hour at 1300 °C -10 minute at room temperature in 90% H₂O – balance O₂) showed through-thickness cracks, which provided the path for water vapor to reach the mullite/SiC interface. The water vapor attacked the SiC substrate causing the formation of a thick porous silica scale [21].

The optical view (Figs.18 (a) and (b)) of the substrate showed that the slurry processed coating is smooth with few cracks present on the surface, a sign of densification of the coating. The cross-sectional SEM view (Figs.19 (a) and (b)) revealed that the coating is dense. But the bonding of coating to the substrate was very poor with cracks all over the coating-substrate interface. Cracks in the mullite layer would affect the durability in long-term exposures (over hundreds of hours) as the water vapor eventually will find its way through the cracks in the mullite coating and reach the Si₃N₄ substrate. Even though mullite sinters at high temperatures but it might be the reaction of slurry with the Y₂O₃/Al₂O₃ additives present in the Si₃N₄ substrate that causes coating-interface debonding at sintering temperature as low as 1430 °C.

To eliminate coating-interface problems, similar coating experiments were performed on SN282 substrates. The optical view (Fig.33) of the substrate showed the coating to be smooth with no sintering cracks on the surface. The cross-sectional
SEM view (Figs.34 (a) and (b)) of the substrate showed the coating to be nice and dense without any cracks at the interface. The coating near the interface was denser compared to the top coat which is desirable from the point of view of its moisture transport resistance. The coating was thicker compared to other mullite/silicate coatings because this slurry was more viscous compared to other slurries. Since the coating had a good contact at the interface and did not show any problems after sintering at 1475 °C, the sintering temperature was increased to 1500 °C to further densify the coat. After this treatment an optical view (Fig.35 (a)) of the substrate showed coating to contain extensive bubbling in the chamfered region of the sample. In addition some sintering cracks were also present on the surface. The cross-sectional SEM view (Fig.35 (b)) of the substrate showed presence of hair line cracks at the interface region. These are signs that 1500 °C is too hot a sintering temperature for this coating.

Examination of 100% Mullite powder coating on SN282 substrate sintered at 1475 °C has shown good contact at the interface without any cracks and this substrate can be further examined for thermal cycling resistance.

5.6 - 50% Mullite sol + 50% Mullite powder (Slurry 8)

Mullite slurries can also be prepared by adding 50% Mullite sol [29.5 % Al₂O₃ + 70.5% (CH₃)₄ NOH. 2SiO₂] + 50% Mullite powder. This mullite slurry contains no PVB as compared to the previously discussed 100% mullite slurry. Since coating densification is achieved by inter-particle sintering a finer starting particle size distribution is expected to enhance sintering kinetics. The alumina and silica particles contained in the initial sol mixture are of nanometer size and are expected to
fill the gap between larger micron-size mullite particles added to the slurry and lead to a denser coating. The optical view (Fig. 21) of the coating prepared by the mullite sol process on the substrate showed the coating to be smooth with few hair line cracks. However, bubbles present on the surface indicated that 1415 °C sintering temperature itself was too hot for the Si$_3$N$_4$ substrate coated by this process. Figures 22(a) and (b) show presence of hairline cracks and elongated pores/openings present at the substrate-coating interface. The coating was still porous but it had good bonding at the interface. The sintering temperature was then increased to 1430 °C with an aim to decrease the porosity of the coating in order to prevent oxygen from reaching the substrate. However, after this treatment many bubbles were seen on the flat-surface and also on the side edges when viewed optically (Fig. 23). The SEM view (Figs. 24(a) and (b)) showed lack of contact between the coating and the interface.

As mentioned above the mullite-sol based slurry results in denser coating with more uniformly distributed fine pores in the coating because of the fine size of the starting mullite particles formed from the alumina sol (Al$_2$O$_3$) and silica sol (SiO$_2$). However, the yttria-alumina additives present in the Si$_3$N$_4$ substrate and the free silica present in the mullite-sol slurry accentuate bubble formation and it occurs at temperatures that are lower than those for the PVB based slurries. Therefore, the 50% mullite sol + 50% mullite powder coating is not suitable for Si$_3$N$_4$ substrate.

Since the mullite sol route did not work for the Si$_3$N$_4$ substrate, similar experiments were also carried out for the SN282 substrates. The coating was smooth but some sintering cracks were seen on the coating surface in the optical view...
(Fig.36 (a)). The coating had good contact at the interface at low sintering temperatures, such as 1430 °C (Fig.36 (b)). However, the coating remained unsintered and porous. This was expected because the mullite particle loading of this slurry was less compared to the 100% mullite slurry. In order to further densify the coating the sintering temperature was therefore increased to 1450 °C. After this treatment the optical view (Fig.37 (a)) revealed the coating to be smooth, but there were sintering related hairline cracks on the surface. The SEM observation (Fig.37 (b)) showed an increase in the densification of the coating, as expected. This mullite-sol coating on the SN282 substrate sintered at 1450 °C was denser as compared with the 100% mullite powder PVB coating on the SN282 for the same sintering temperature (Figs.34 (a) and (b)). The coating near the interface was also denser compared to the rest of the coat. Presence of some white looking phases near the interface implies a transport of Lu₂O₃ from the substrate towards the coating. The coating had very good bonding at the interface. Now, in order to further increase the coating density, the substrate was sintered to 1475 °C. Optical view (Fig.38 (a)) showed bubbles in the coating and sintering cracks on its surface. The SEM view (Fig.38 (b)) showed a very dense coating but a lack of bonding with the substrate. This indicated that 1475 °C was too hot a sintering temperature for the SN282 substrate coated with mullite particle containing mullite sol slurry.

A 50% mullite sol + 50% mullite powder coating on SN282 substrate at 1450°C showed good contact at the interface without any cracks. This coating-substrate combination can therefore be examined for its thermal cycling resistance.
CHAPTER VI
CONCLUSIONS AND RECOMMENDATIONS

Silicon based ceramics, such as silicon-carbide, silicon nitride or SiC-SiC composites have very attractive high temperature mechanical properties for their potential application as ultrahigh temperature gas turbine engine components in hypersonic transport. However, their degradation in moisture containing environment due to the continued silica formation and its evaporation demands availability of an environmental barrier coating without which they just can not be used for these applications. This research evaluated mullite based coatings on silicon nitride substrates (Si₃N₄ and SN282). Compositions of the mullite coatings were varied with the addition of four rare earth silicates, gadolinium (Gd₂SiO₅), lutetium (Lu₂SiO₅), hafnium (HfSiO₄), and erbium (Er₂SiO₅) to the slurry.

Mullite / rare earth silicate coatings on Si₃N₄ and SN282 substrates, sintered between 1350°C -1450 °C, showed bubbles and cracks on the coated surface and there was lack of bonding between the coating and the interface.
94% mullite-6% Er$_2$SiO$_5$ coating on Si$_3$N$_4$ and SN282 substrates at 1430 °C showed coating peeled off the substrate due to CTE mismatch.

94%Mullite-6% Lu$_2$SiO$_5$ coating on Si$_3$N$_4$ substrate at 1430 °C and SN282 substrate at 1450 °C showed coating was dense but had no contact with the substrates.

94%Mullite-6% HfSiO$_4$ coating on Si$_3$N$_4$ and SN282 substrates at 1430 °C showed de-bonding occurs at the interface due to thermal mismatch between the substrate and the coating.

Thermal cycling tests in simulated moisture containing environment of the coated samples (between 1350 °C to room temperature) showed that 88% Mullite-12% Gd$_2$SiO$_5$ (slurry 4) coating on Si$_3$N$_4$ and SN282 substrates developed extensive cracks and de-bonding between the coating and the interface. The Si$_3$N$_4$ substrates were pre-oxidized in air for 5 h at 1300 °C and 1400 °C respectively and coated with 88% Mullite-12% Gd$_2$SiO$_5$ (slurry 5) and 94% Mullite powder - 6% Gd$_2$SiO$_5$. Bubbles still formed on the surface and pre-oxidization of the Si$_3$N$_4$ substrate to improve coating substrate bonding did not work. 94% Mullite powder - 6% Gd$_2$SiO$_5$ coating on Si$_3$N$_4$ and SN282 substrates at 1430 °C showed that the coating de-bonding still occurs at the interface. This shows that regardless of decreasing the gadolinium silicate content, coating-interface problems still occur.

Si$_3$N$_4$ coated with 100% Mullite (slurry 7) and 50% mullite sol + 50% mullite powder (slurry 8) also showed lack of good bonding to the substrate.

Only the 100% mullite (slurry 7) coating on SN282 substrate sintered at 1475°C and 50% mullite sol + 50% mullite powder (slurry 8) coating on SN282 substrate sintered at 1450°C have shown good bonding of coating to the substrate.
without any cracks at the interface. It is therefore recommended that these EBC coatings be further evaluated by thermal cycling in the moisture containing environment at high temperatures for the SN282 substrates.

The lack of available physical properties and chemical thermodynamics data requires an empirical experimental approach to develop an EBC. More tests should be done to identify new materials candidates for water vapor resistance. Thermodynamic modeling should be done to study the effect of the environment on the coating and to study the effect of the interface of the coating on silicon nitride substrates. Therefore, if these steps are taken then it would be easier to identify best EBC coating on silicon nitride substrates.
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