Integrated TBC/EBC for SiC Fiber Reinforced SiC Matrix Composites for Next Generation Gas Turbines

DoE UTSR DEFE0031281 (10/2017 – 09/2020)
Progress Review Meeting
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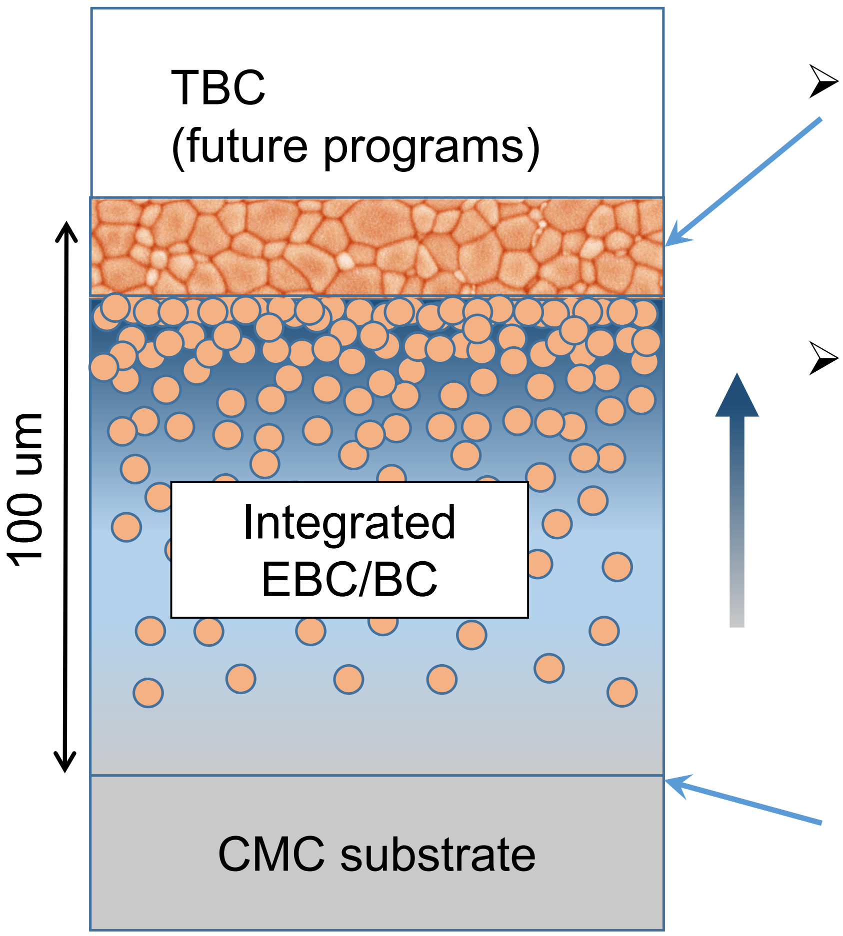
In Collaboration with GE Power Team led by John Delvaux

Outlines

1. The concept of integrated TBC/EBC/BC
2. Overall Goals
3. Objectives
4. Project Tasks
5. Project Progress
6. Project Short Term Plans

1. The concept of integrated TBC/EBC/BC



- Dense Y_2O_3 or Yb_2O_3 top layer ensures low oxygen diffusion and volatilization rates under high temperature high velocity steam environment.
- Graded composition allows smooth transition from SiC to SiBCNO with increasing amount of embedded Y_2O_3 particles.
  - Graded structure avoids sharp CTE mismatch
  - Tolerant to oxidation; further increase TBC lifetime
- PDC SiC bond coat/SiC substrate interface provides excellent adhesion

2. Overall Goals

- Develop an integrated and graded EBC/BC that is:
  - Good bonding with CMC;
  - Graded compositions without sharp interfaces to mitigate thermal stresses from CTE mismatch;
  - Low oxygen transport rate, low oxidation rate and low volatility in high temperature, high velocity steam environment;
  - Tolerant to certain degree of oxidation thereby preventing catastrophic failure;
  - Chemically stable and compatible with CMC and TBC

- Create a strong collaborative team with complementary expertise and state-of-the-art facilities
  - The Clemson University team of Drs. Bordia and Peng.
  - The GE team, led by John Delvaux

3. Objectives

- Investigate the effect of composite stoichiometry (*i.e.* Si/B/C/N ratio in the precursor and the ratio of the Si-based precursor to yttrium oxide (Y_2O_3) (or ytterbium oxide (Yb_2O_3)) particle filler and processing conditions on the size of the resultant phases and nanostructure of the composite ceramics.

- Investigate the effect of the composition and nanostructure on the thermal properties and oxidation and volatilization behavior in oxidizing and high velocity steam environments. The control parameters are the stoichiometry of the precursor (*e.g.* Si/B/C/N ratio) and the volume fraction of the oxide particles Y_2O_3 (or Yb_2O_3) and range of microstructures produced as part of the first objective

3. Objectives (contd.)

- Process the graded Y_2O_3 (or Yb_2O_3) particulate /silicon boron carbon nitride (SiBCN) matrix composite coating and investigate the phase and microstructure stability during high velocity steam exposure at temperatures up to 1500ºC.

- Develop a method to create Y_2O_3 (or Yb_2O_3) and SiBCN powders with predetermined compositions suitable for atmospheric plasma spraying (APS). The powders will be provided to the industrial collaborators for the fabrication of integrated environmental barrier coating/bond coating (EBC/BC) using APS.

- Evaluate the performance of integrated BC/EBCs from APS under high velocity steam environments at temperatures up to 1500ºC.

4. Project Tasks

- **Task 1:** Project management and planning
- **Task 2**: Processing and stability of Y_2O_3-Si-C-N and Yb_2O_3-Si-C-N composites
- **Task 3:** Thermal and oxidation response of Y_2O_3-Si-C-N and Yb_2O_3-Si-C-N composites
- **Task 4:** Processing and performance of graded coatings processed using cold spray and pyrolysis
- **Task 5:** Processing and performance of graded coatings processed using atmospheric plasma spraying (APS)

5. Project Progress-Task 1

Task 1.0: Project management and planning

- Kickoff meeting 10/27/17
- A poster at the 2017 UTSR review meeting
- Recruitment of UG students and post doc to work on the project
- Coordination with GE Team including scheduling review meetings
- Regular review meetings with DoE Program Manager and GE team
- A talk and a poster at the 2018 UTSR review meeting
- A talk at 2019 UTSR review meeting.

5. Project Progress-Task 2

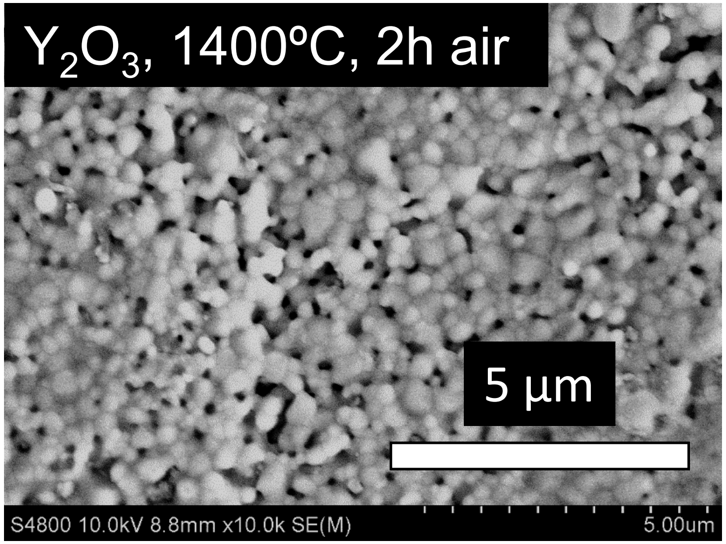
Task 2: Processing and stability of Y_2O_3-Si-C-N and Yb_2O_3-Si-C-N composites

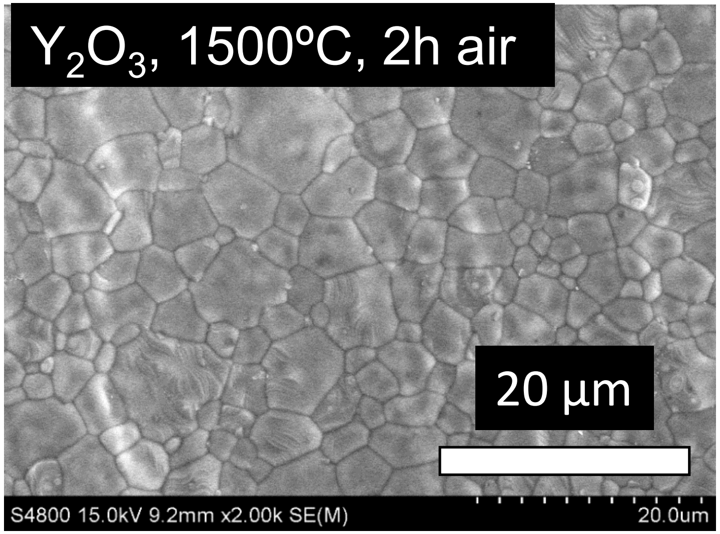
Completed:

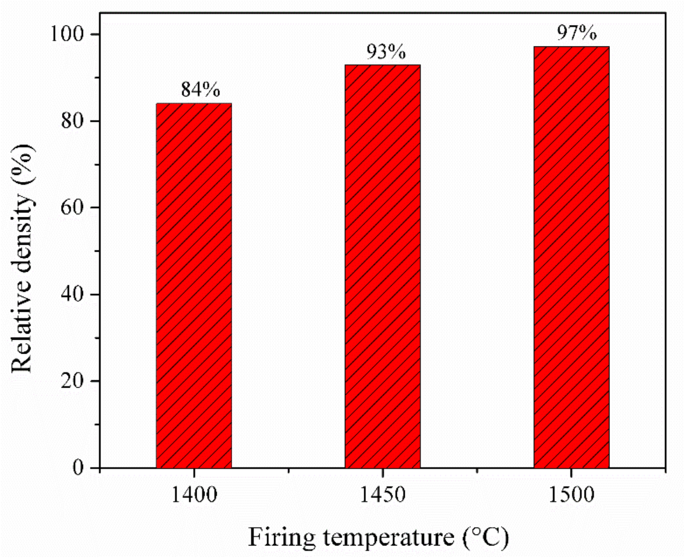
- ❖Sintered and characterized dense Y_2O_3 and Yb_2O_3 ceramics to be applied as the top layer.
- ❖Studied the effect of TiO_2 doping on the sintering of Y_2O_3 and Yb_2O_3.
- ❖Developed the processing protocols of Y_2O_3-SiCN and Yb_2O_3-SiCN composite submicrometer powder.
- ❖Studied the processing of composites in the Y_2O_3-SiCN and the Yb_2O_3-SiCN system. Investigated the effect of composition and processing temperature on density, porosity, and microstructure.
- ❖Characterized the crystalline phases of sintered Y_2O_3-SiCN and Yb_2O_3-SiCN composites.

5. Task 2-Sintering of Y_2O_3 Top Coat

Y_2O_3 ceramic pellets were fired at 1400, 1450, 1500ºC for 2h in air



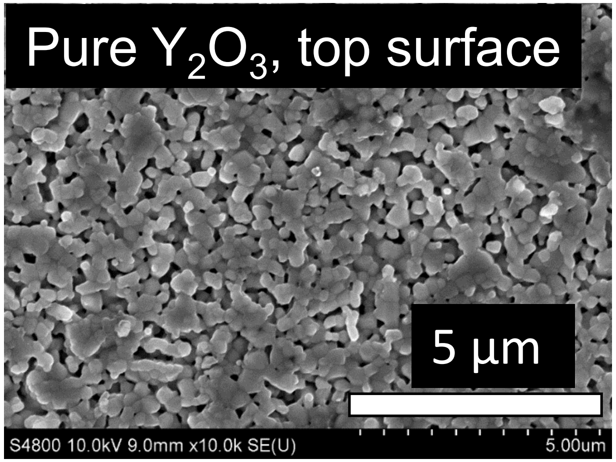


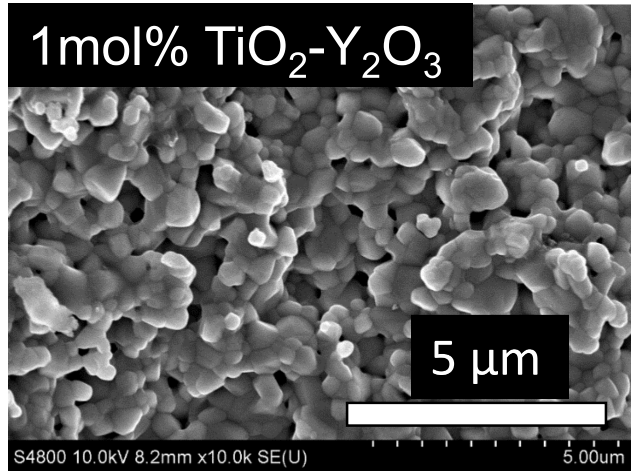


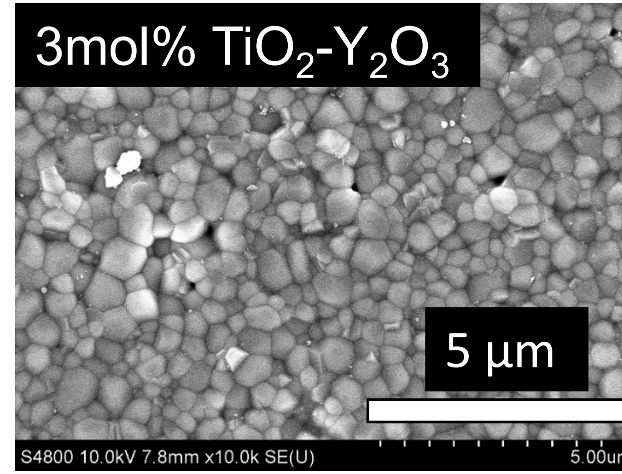
- At 1500ºC, the relative density reaches 97% with only 0.5% of open porosity
- Y_2O_3 fabricated at 1500ºC can be used at top EBC coating

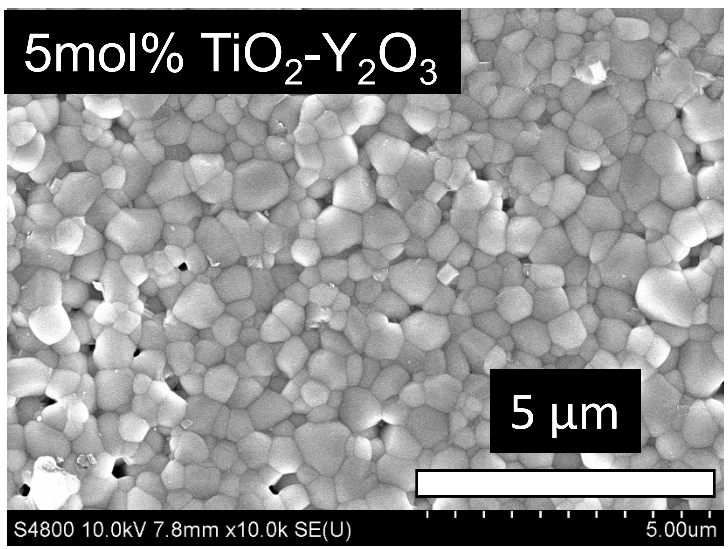
5. Task 2-Sintering of Y_2O_3 Top Coat

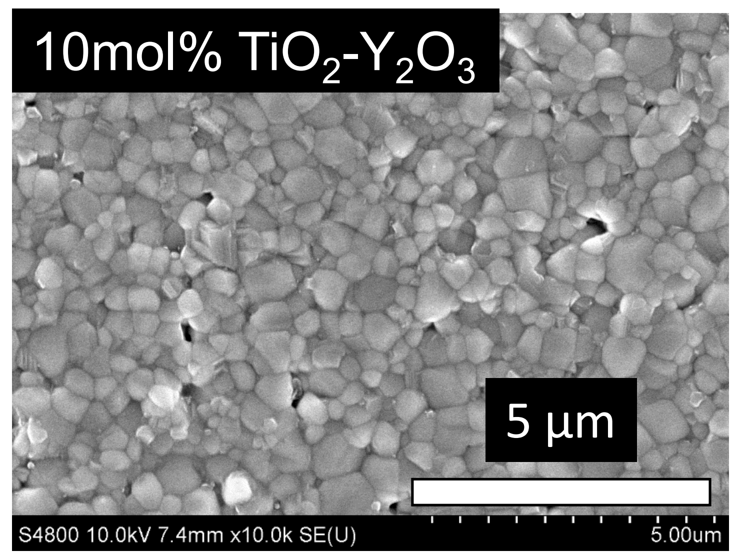
Y_2O_3 was doped with 1, 3, 5 and 10 mol.% TiO_2, and sintered at 1350 °C, air





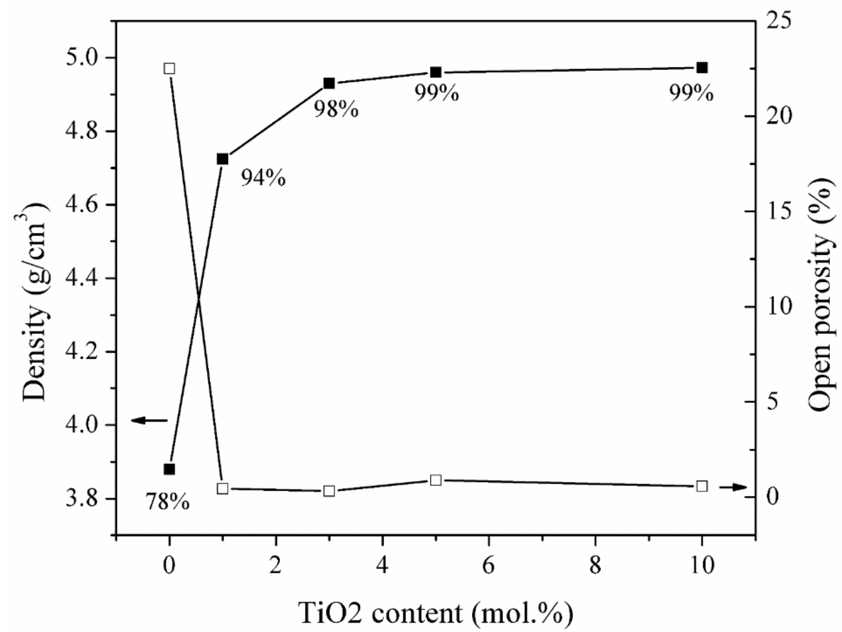


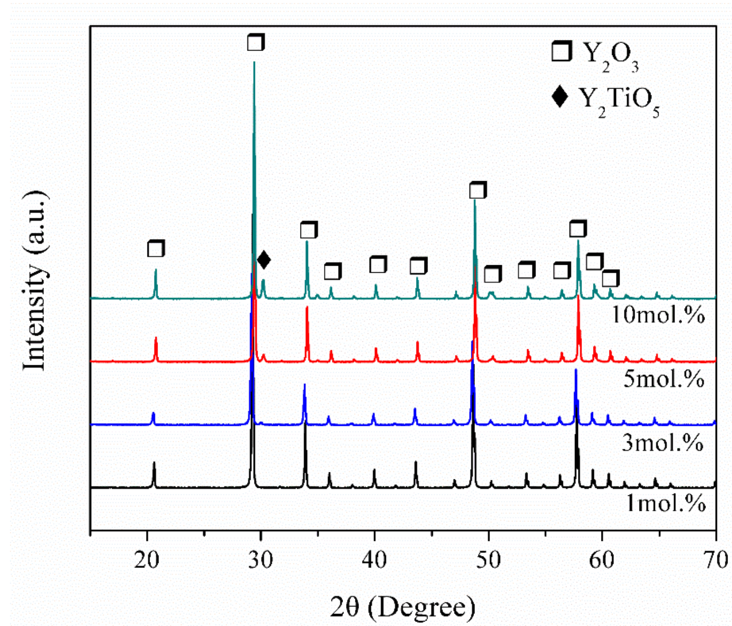


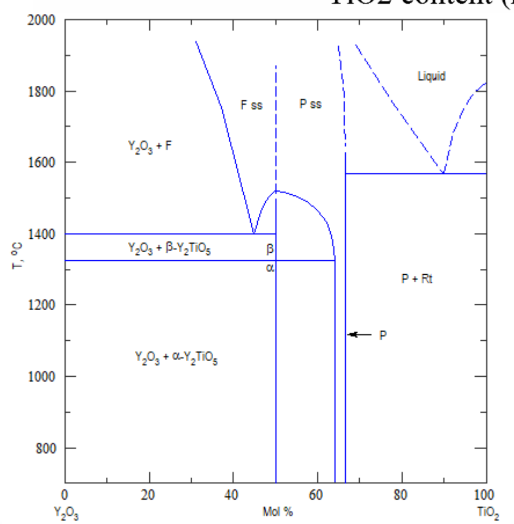


5. Task 2-Sintering of Y_2O_3 Top Coat

Y_2O_3 was doped with 1, 3, 5 and 10 mol.% TiO_2, and sintered at 1350 °C, air





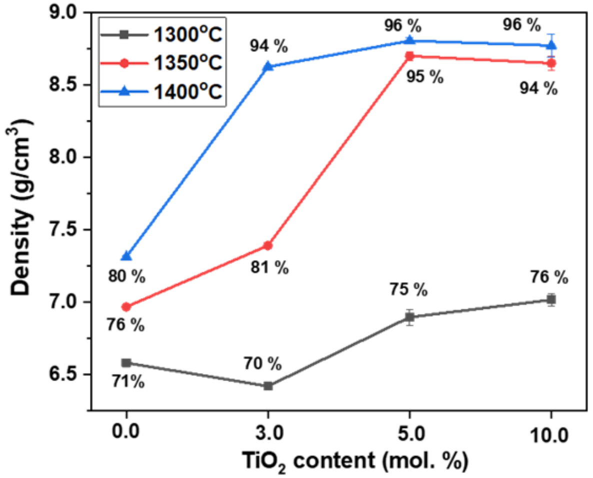


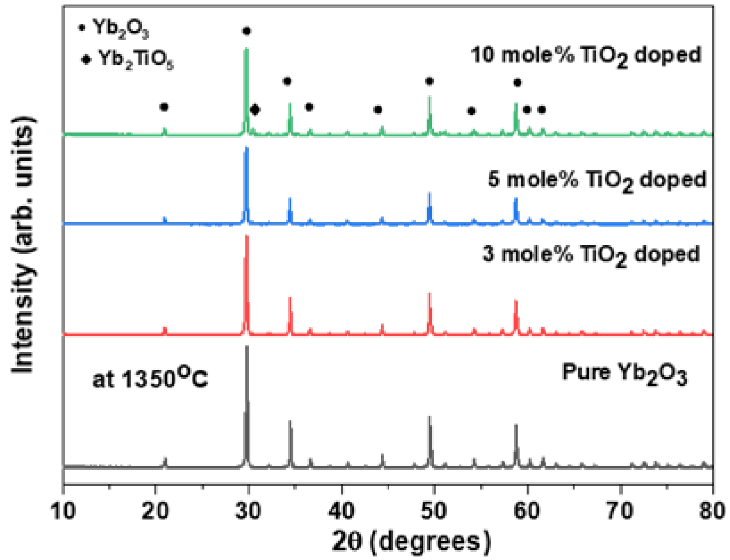
Density and open porosity

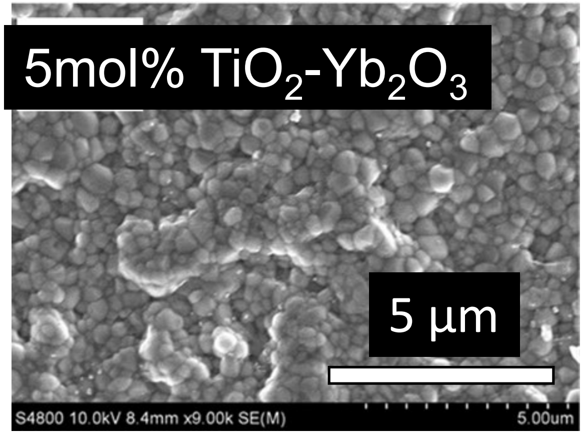
- ❖ As TiO_2 concentration increases, density increases and porosity decreased.
- ❖ 3-5 mol% TiO2 doping seems to be sufficient.
- ❖ Y_2TiO_5 minor phase formed. The results is in good agreement with Y_2O_3-TiO_2 phase diagram.

5. Task 2-Sintering of Yb_2O_3 Top Coat

Similar results were observed for Yb_2O_3 doped with TiO_2, sintered at 1350°C



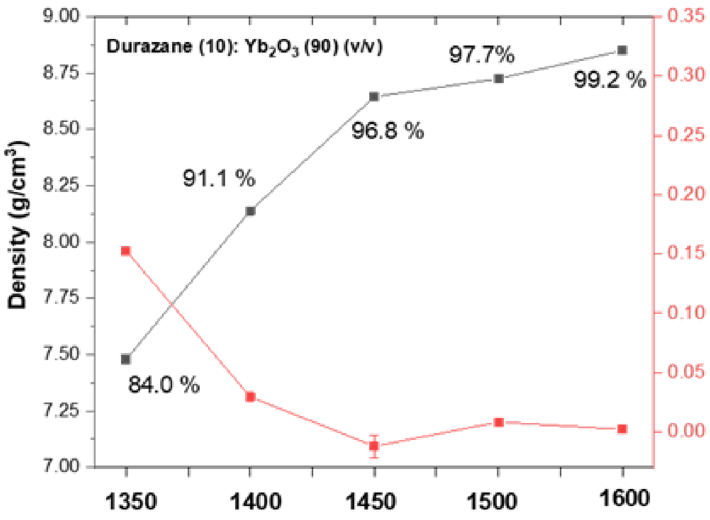


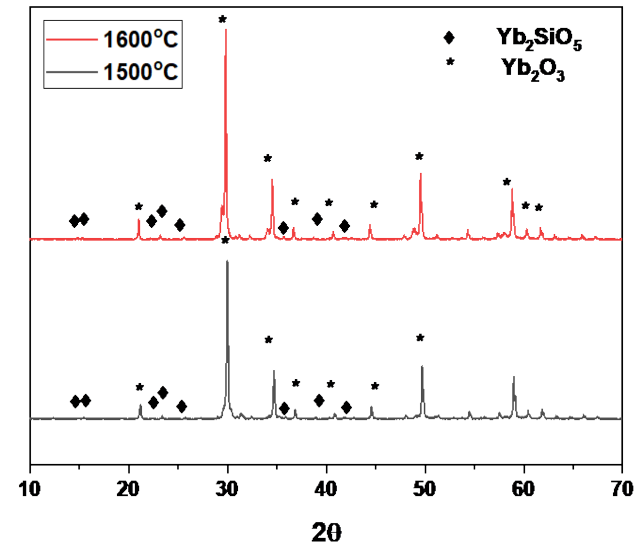


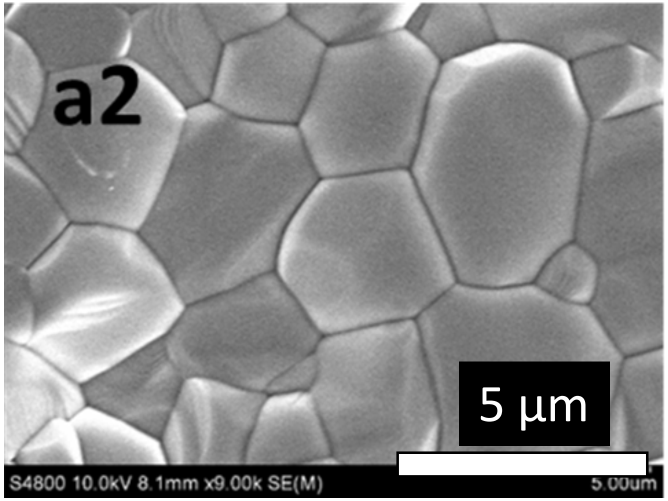
- Doping of TiO_2 significantly increase the density of Yb_2O_3 sintered at 1350°C.
- 3-5 mol% TiO_2 doping seems to be sufficient for 1350°C heat-treatment.
- Minor Yb2TiO5 formed, which is expected.

5. Task 2-Yb_2O_3/SiCN Composite Ceramic

90 vol%(5 mol%TiO_2-Yb_2O_3)-10 vol% SiCN







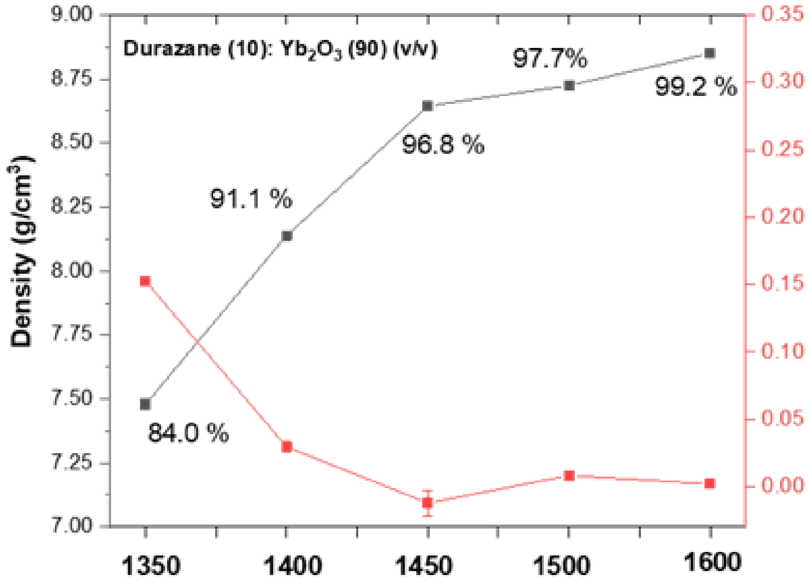
Yb_2SiO_5 forms due to the reaction between Yb_2O_3 and SiO_2.

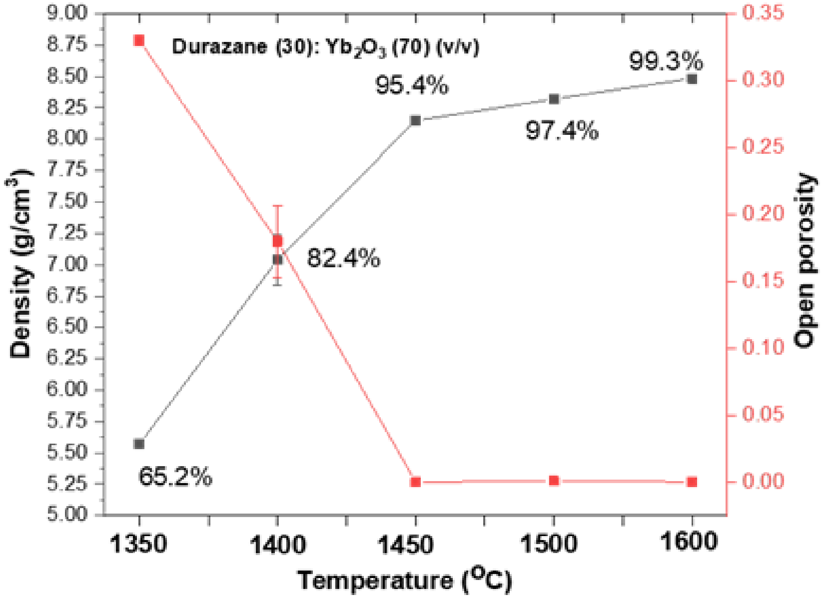
Sintered at 1600°C

5. Task 2-Yb_2O_3/SiCN Composite Ceramic

90 vol%(5 mol%TiO_2-Yb_2O_3)-10 vol% SiCN

70 vol%(5 mol%TiO_2-Yb_2O_3)-30 vol% SiCN

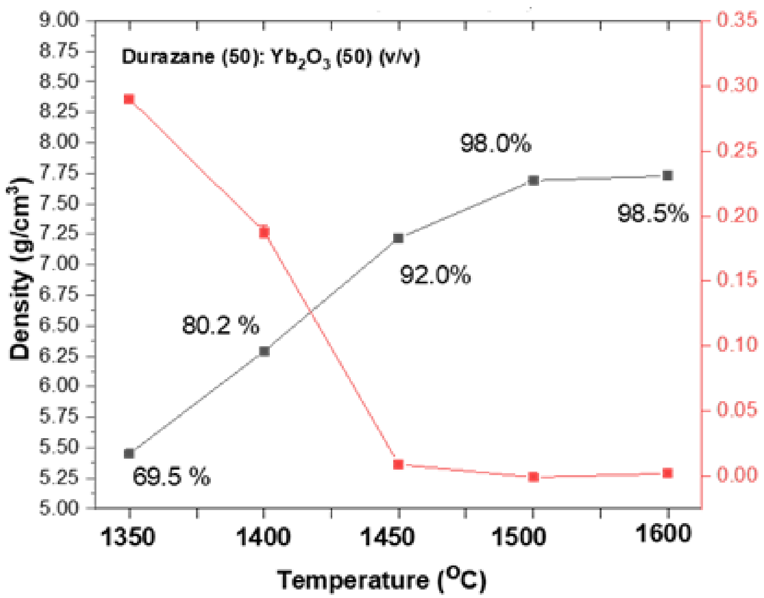


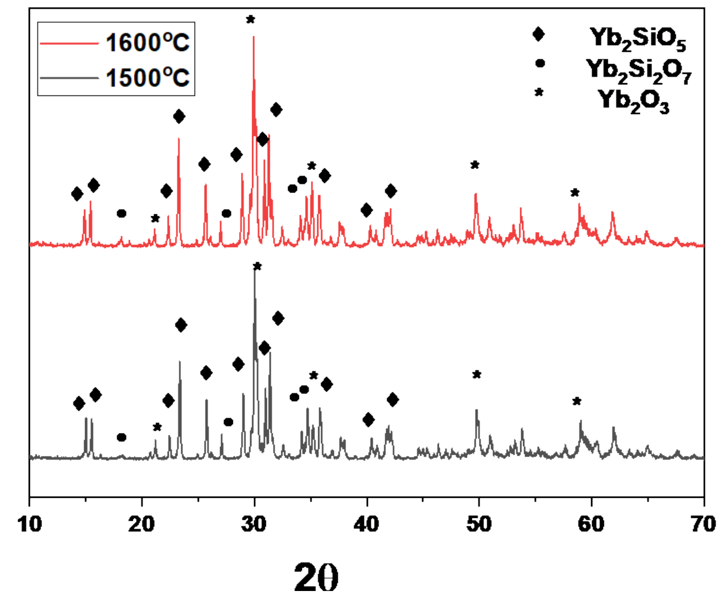


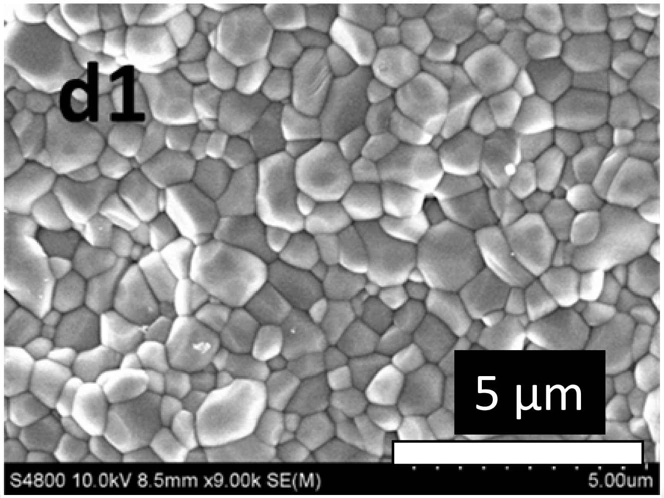
A lower density was observed at 1300-1400°C with a higher SiCN content.

5. Task 2-Yb_2O_3/SiCN Composite Ceramic

50 vol%(5 mol%TiO_2-Yb_2O_3)-70 vol% SiCN





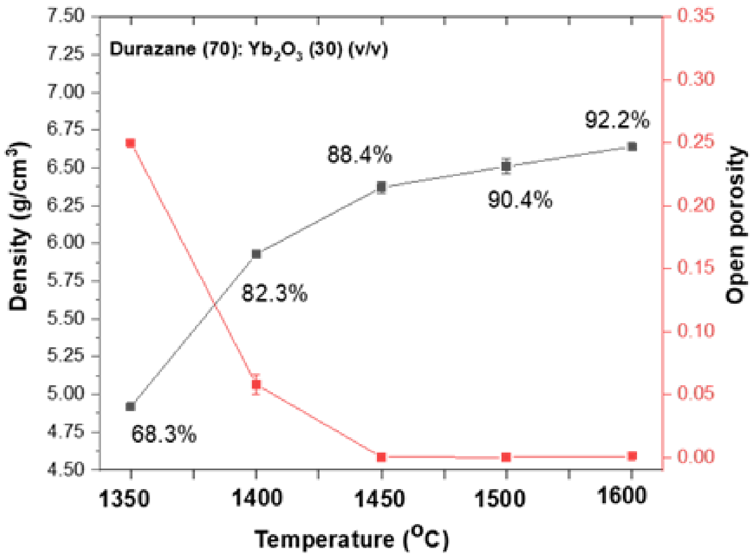


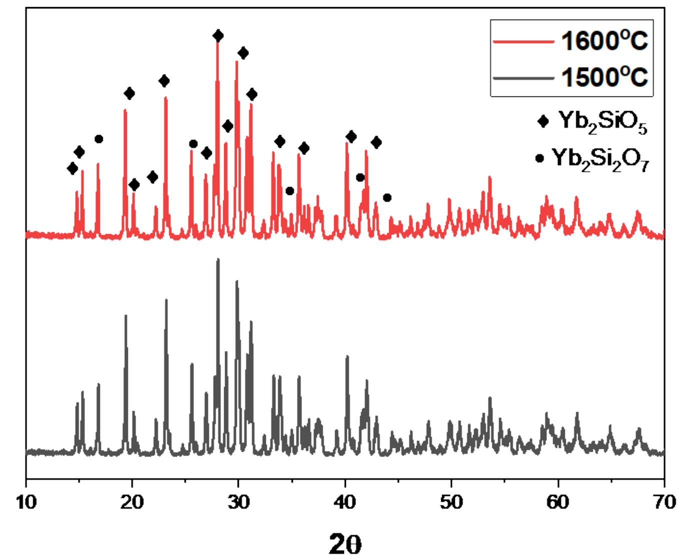
A mixture of Yb_2SiO_5 and $Yb_2Si_20_7$ was observed.

Sintered at 1600°C

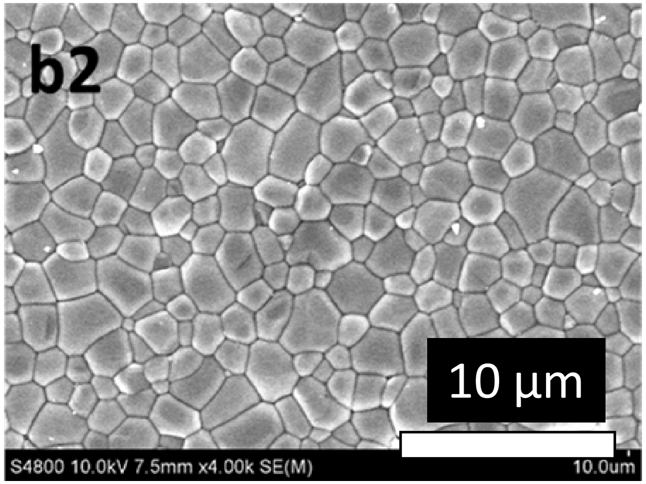
5. Task 2-Yb_2O_3/SiCN Composite Ceramic

30 vol%(5 mol%TiO_2-Yb_2O_3) - 70 vol% SiCN





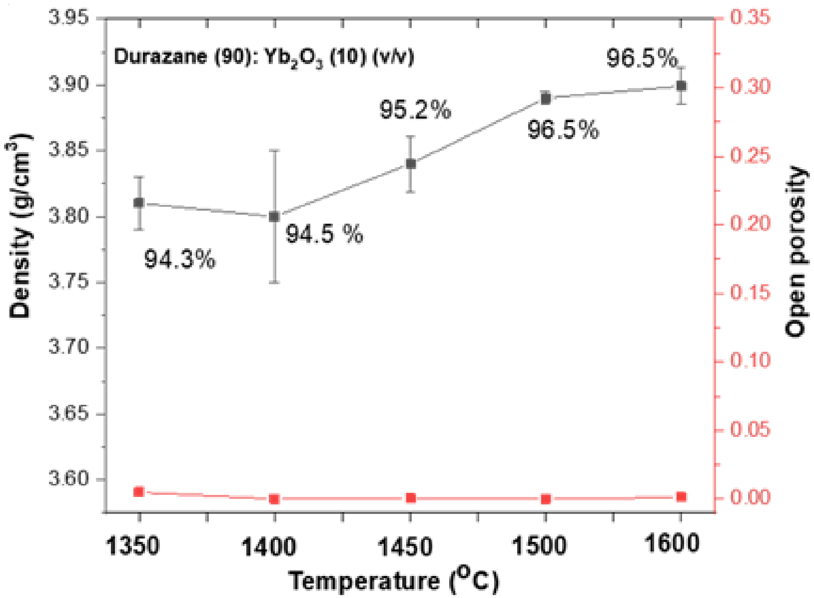
A mixture of Yb_2SiO_5 and $Yb_2Si_20_7$ was observed

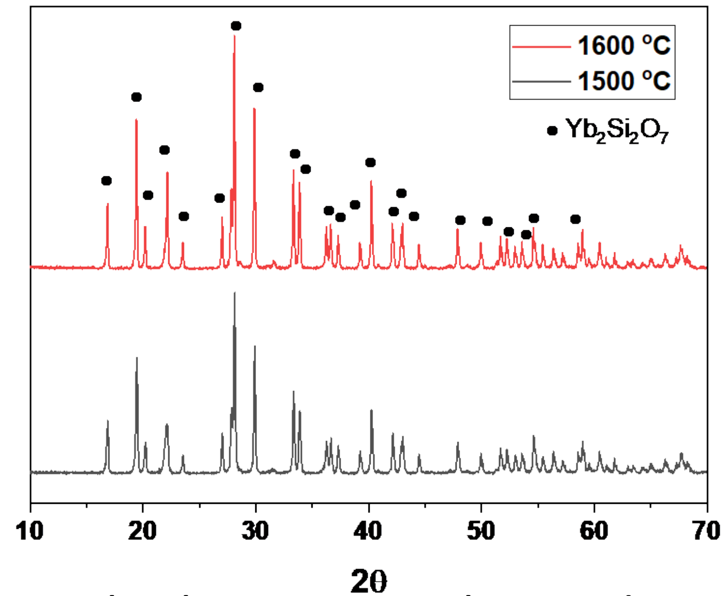


Sintered at 1600°C

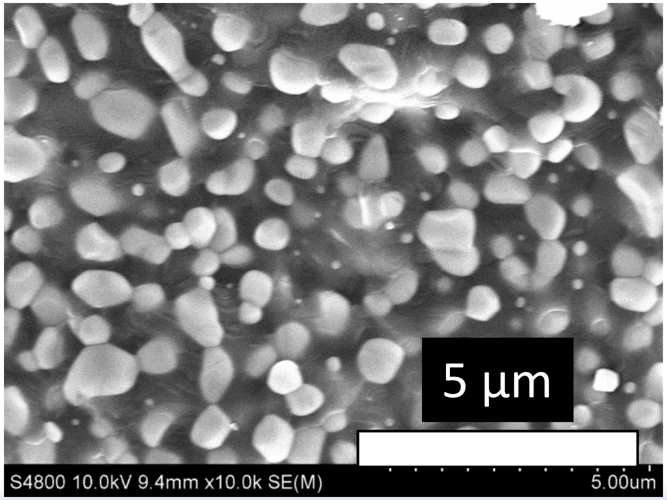
5. Task 2-Yb_2O_3/SiCN Composite Ceramic

10 vol%(5 mol%TiO_2-Yb_2O_3)-90 vol% SiCN





Only $Yb_2Si_2O_7$ was observed at high temperatures.



Sintered at 1600°C
$Yb_2Si_2O_7$ islands in a SiCN matrix

5. Task 2-Y_2O_3/SiCN Composite Ceramic

- Y_2O_3 or Yb_2O_3 reacts with SiO_2 to form yttrium silicates (Y_2SiO_5 and $Y_2Si_2O_7$) or ytterbium silicates (Yb_2SiO_5 and $Yb_2Si_2O_7$).

- TiO_2 facilitates Y_2O_3 but not predominant for Y_2O_3-SiCN composites.

- When Y_2O_3 or Yb_2O_3 content was high (*e.g.* 90 vol% and 70 vol% Y_2O_3), Y_2SiO_5 (or Yb_2O_3) was the dominant phase. The dominant phases of 50 vol% and 30 vol% Y_2O_3 (or Yb_2O_3) were a mixture of the two types of yttrium silicates or (ytterbium silicates). When SiCN content was high e.g. 90 vol%, $Y_2Si_2O_7$ ($Yb_2Si_2O_7$)was the dominant phases.

5. Project Progress-Task 2

Task 2: Processing and stability of Y_2O_3-Si-C-N and Yb_2O_3-Si-C-N composites (90% complete)

In the future:

❖Complete the microstructure characterization of all the sintered samples.

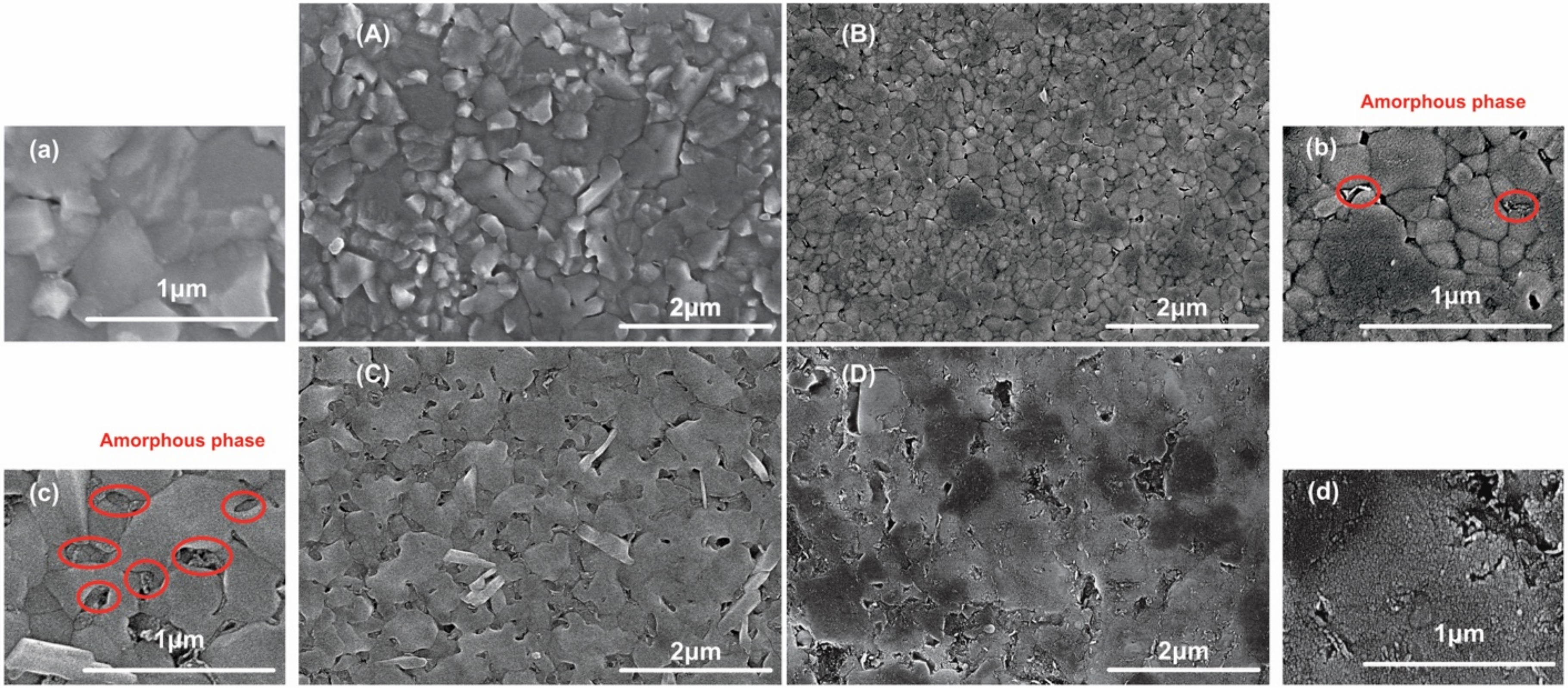
5. Project Progress-Task 3

Task 3: Thermal and oxidation response of Y_2O_3-Si-C-N and Yb_2O_3-Si-C-N composites (60% complete)

Completed:

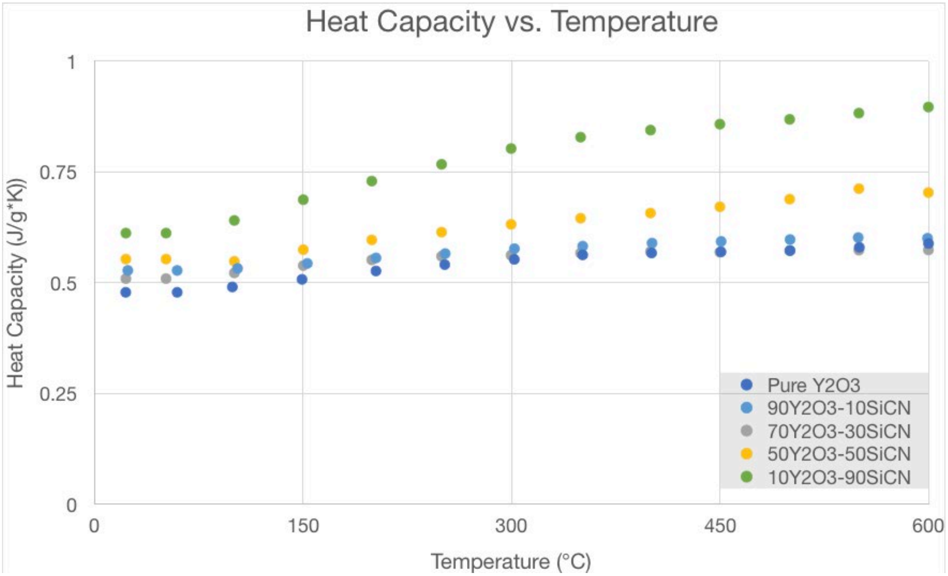
- ❖Fabricated fully dense Y_2O_3-SiCN and Yb_2O_3-SiCN composite using field-assisted sintering (FAST) at 1350°C as the model system for thermal conductivity measureament.
- ❖Measured the thermal conductivities of Y_2O_3-SiCN composites
- ❖Developed the protocol of accurately measure the Young's modulus for small sample at high temperature.
- ❖Characterized the oxidation stability of Yb_2O_3-SiCN composite ceramic powders
- ❖Studied the stability of Yb_2O_3-SiCN composite under high speed steam jet at 1300°C.

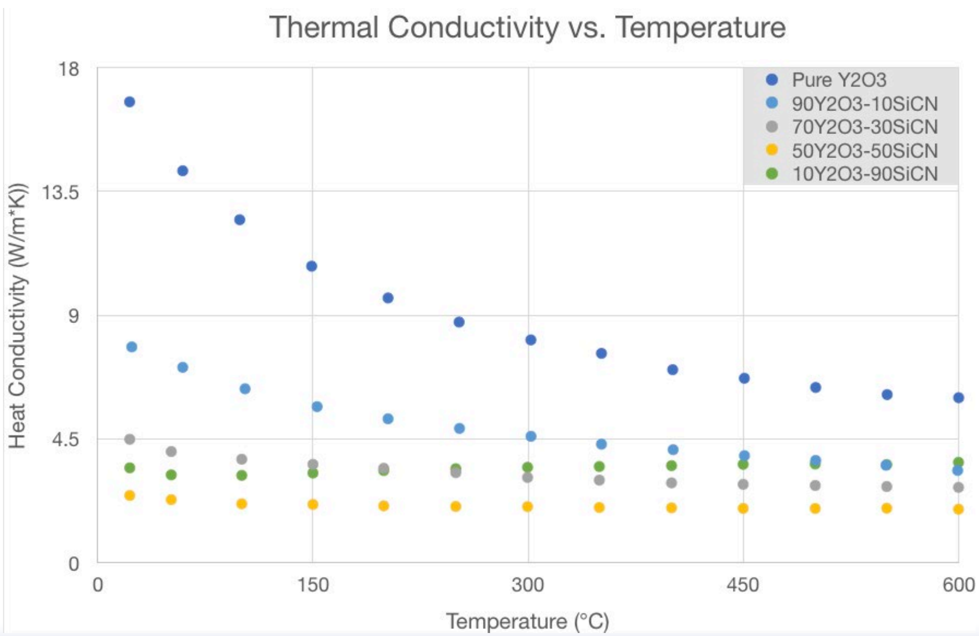
5. Task 3-Process fully dense Y_2O_3/SiCN Composite Ceramic



Microstructure of fully dense (Archimedes' method) FAST sintered at 1350°C for 5 min under vacuum. (A),(a) 90-Y_2O_3/10-SiCN; (B),(b) 70-Y_2O_3/30-SiCN; (C),(c) 50-Y_2O_3/50-SiCN; (D),(d) 10-Y_2O_3/90-SiCN.

5. Task 3-Themal conductivity of Y_2O_3-SiCN composites

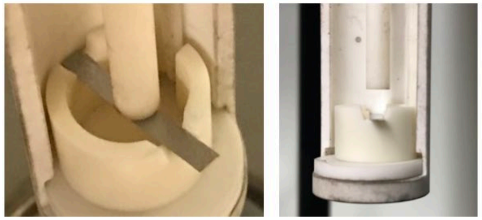


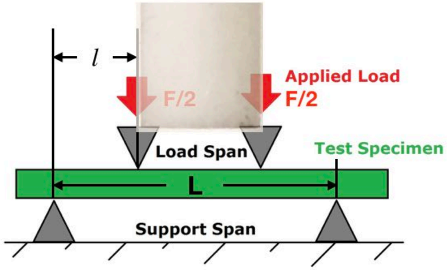


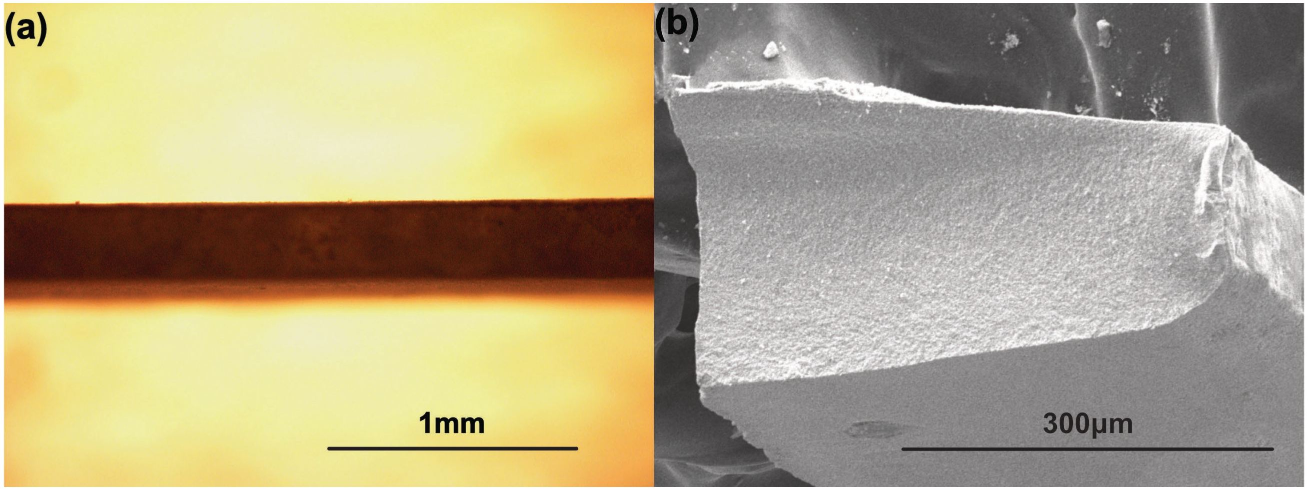
- ❖ The higher SiCN content, the higher heat capacity.
- ❖ The higher Y_2O_3 content, the higher thermal conductivity

5. Task 3-Young's Modulus of Y_2O_3-SiCN composites

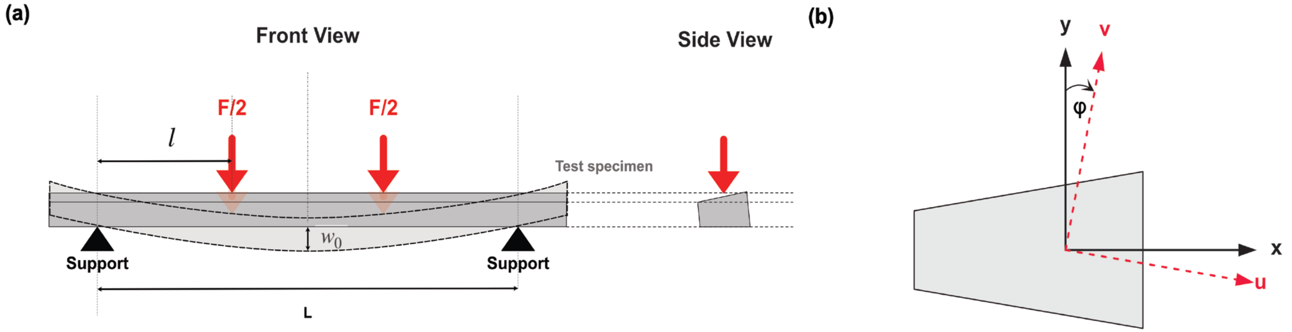
Thermo Mechanical Analyzer (SS6000)







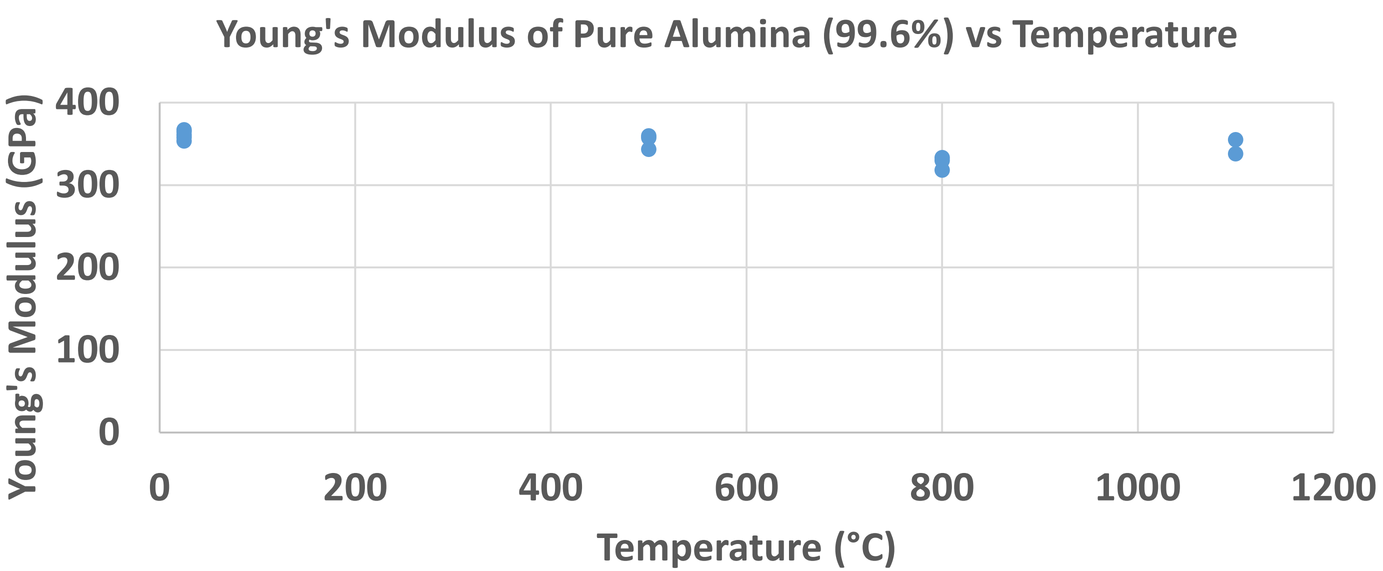
5. Task 3-Young's Modulus of Y_2O_3-SiCN composites



$$E = \frac{Fl(3L^2 - 4l^2)}{48 w_0 I}$$

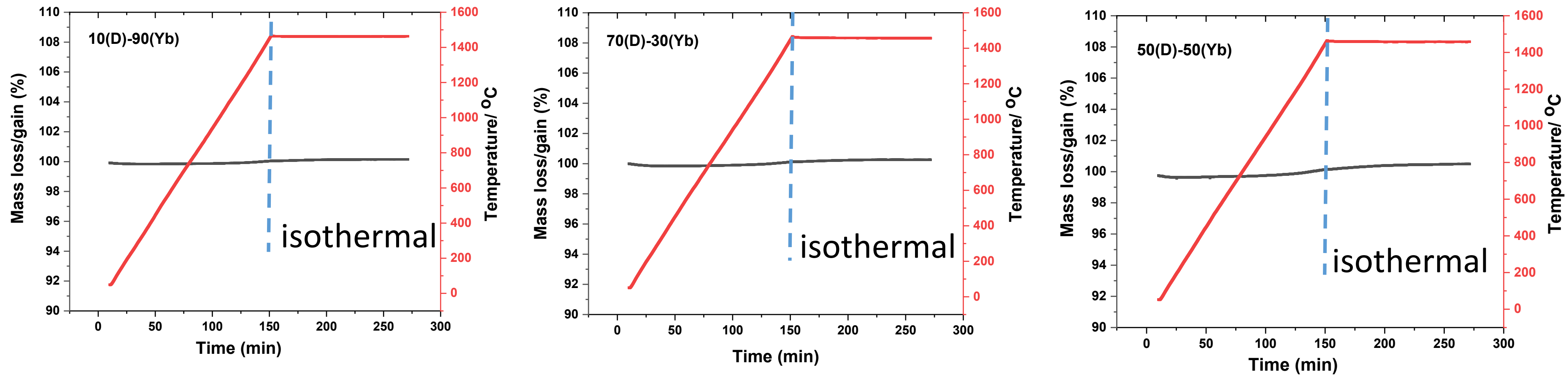
second moment of inertia

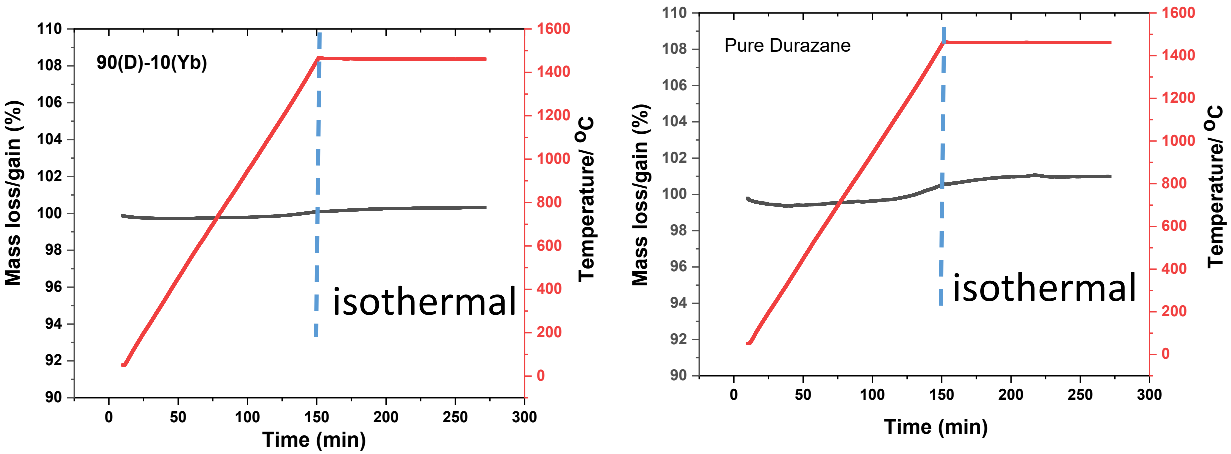
$$I_v = \frac{I_x + I_y}{2} - \frac{I_x - I_y}{2}\cos 2\varphi + I_{xy}\sin 2\varphi$$



This measured value is very close to the reported value of the Young's modulus of pure alumina(99.6%) (380~400GPa).

5. Task 3-Oxidation behaviors of Yb_2O_3-SiCN composites

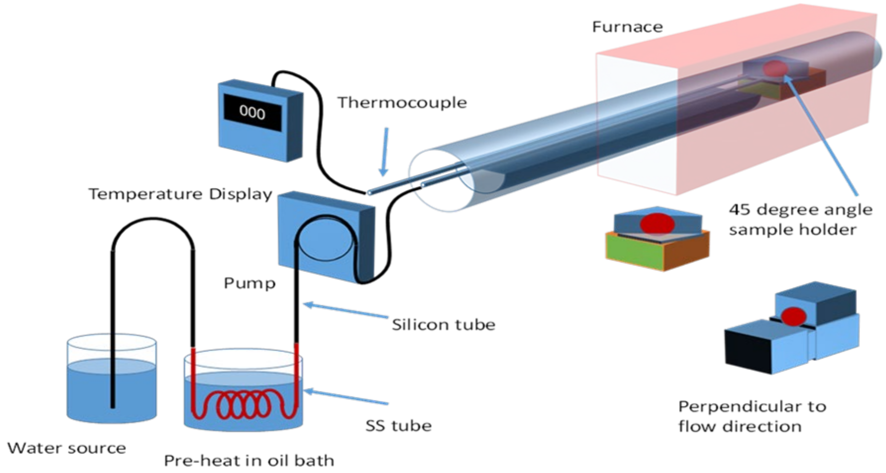




- Maximum weigh gain is about 1% after 2 h in 1450°C.
- With a higher SiCN content, a higher weight gain was observed.

Powder samples were heat-treated using the same procedure (1450°C in air for 2 h) as for the sintered samples prior the TGA.

5. Task 3-Stability under high velocity steam



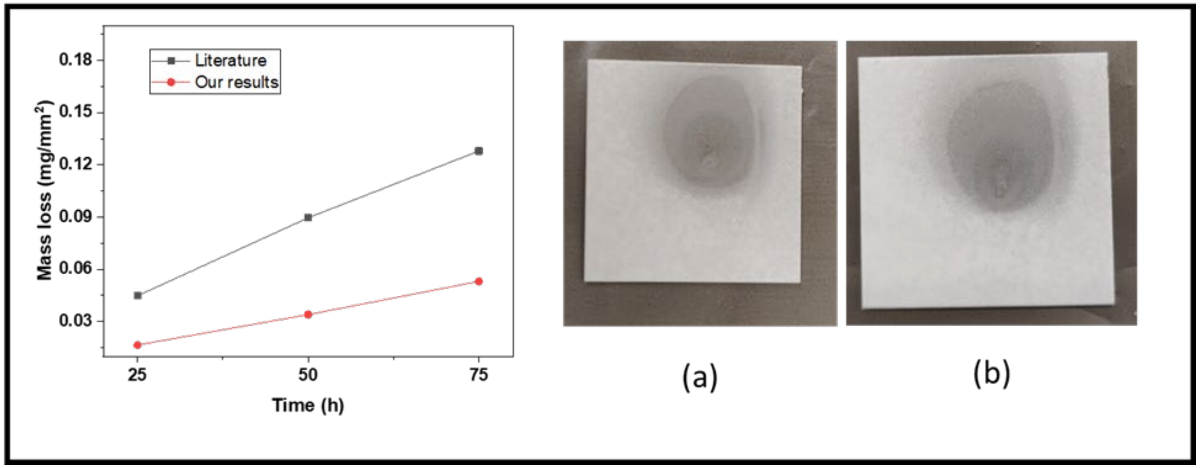


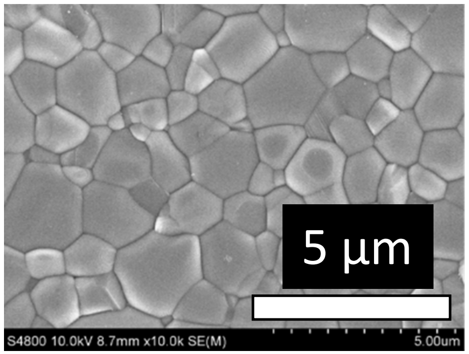
Image of steam exposed quartz. (a) after 50 h (b) after 75 h

Dimension: 25 x 25 x 1 mm
Water flowrate: 1.7- 1.8 ml/min
Temp: 1300°C
Distance between specimen and steam impingement: 0.75 cm

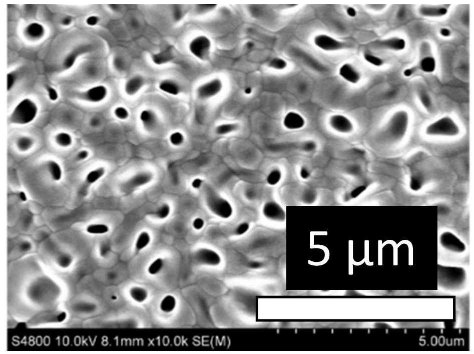
The deviation is due to rate of volatilization is not uniform throughout the surface.

5. Task 3-Stability of Yb_2O_3 under high speed steam

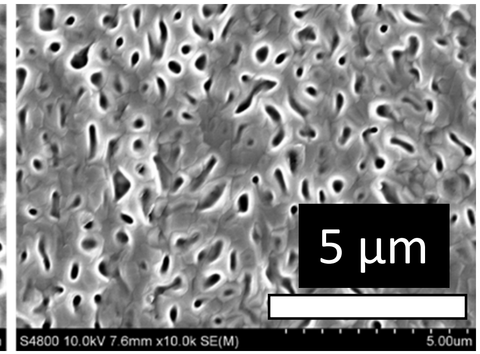
High velocity steam impact at 1300°C



Fully dense Yb_2O_3



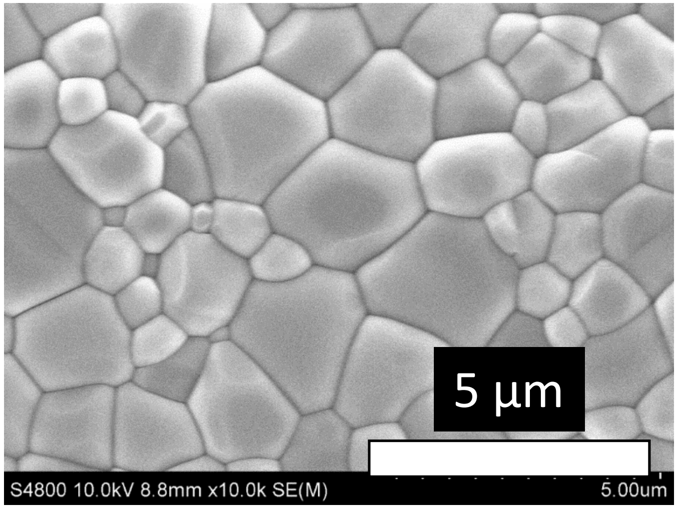
After 25 h



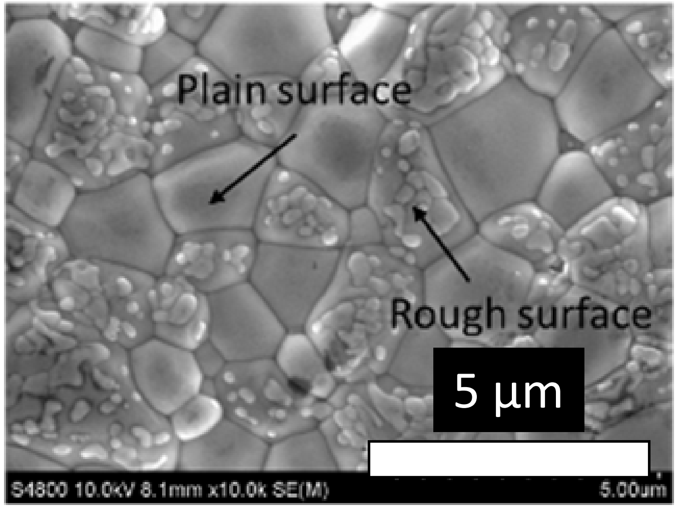
After 75 h

- EDX showed heavy presence of Al element on the surface.
- Porous microstructure is probably due to the reaction between Yb_2O_3 and aluminum hydroxide.
- Sapphire tube will be used instead of alumina tube.

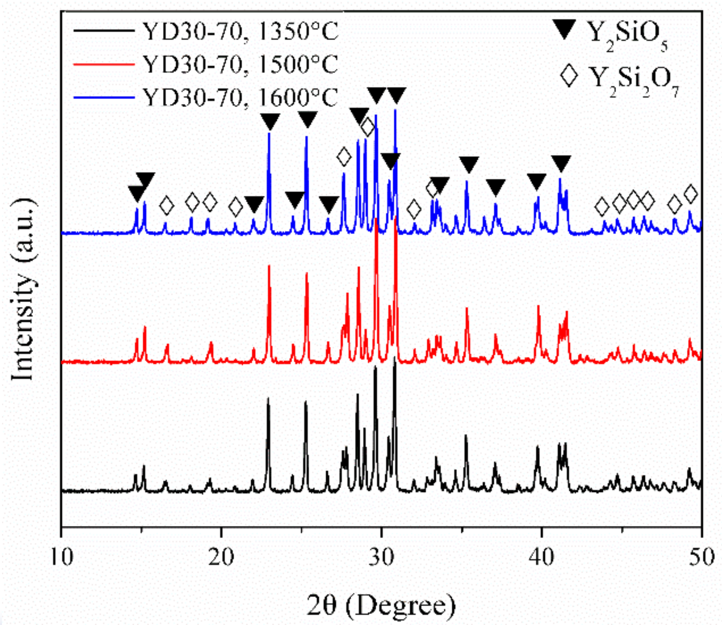
5. Task 3-Stability of Yb_2O_3–SiCN composite under high speed steam



$30\%Yb_2O_3$-$70\%SiCN$

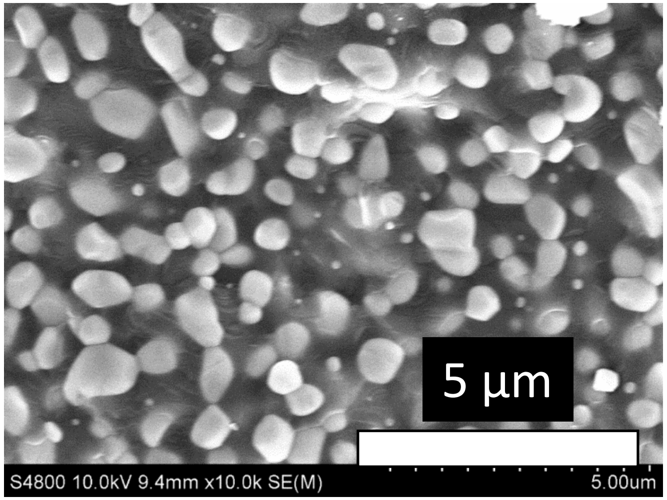


After 25 h under steam jet at 1300°C

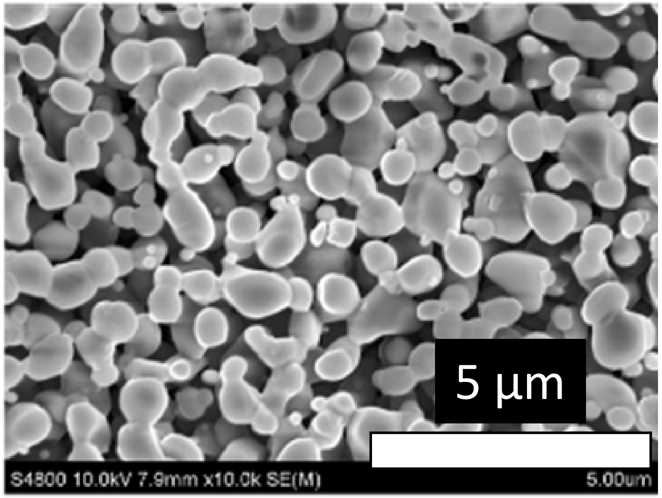


- This composition showed exceptional stability under high velocity steam jet.
- We hypothesize that one of the yttrium silicate is more stable than the other.

5. Task 3-Stability of Yb_2O_3–SiCN composite under high speed steam



10%Yb_2O_3-90%SiCN



After 25 h under steam jet at 1300°C

Silica and SiCN phase was removed, as we expected

$$SiC_{(s)} + 1.5\ O_{2\ (g)} \rightarrow SiO_{2\ (l)} + CO_{(g)}$$

$$SiO_{2\ (l)} + 2H_2O_{(g)} \rightarrow Si(OH)_{4\ (g)}$$

5. Project Progress-Task 3

Task 3: Thermal and oxidation response of Y_2O_3-Si-C-N and Yb_2O_3-Si-C-N composites (60% complete)

Future plan:

❖Complete the thermal and oxidation response study for all compositions.

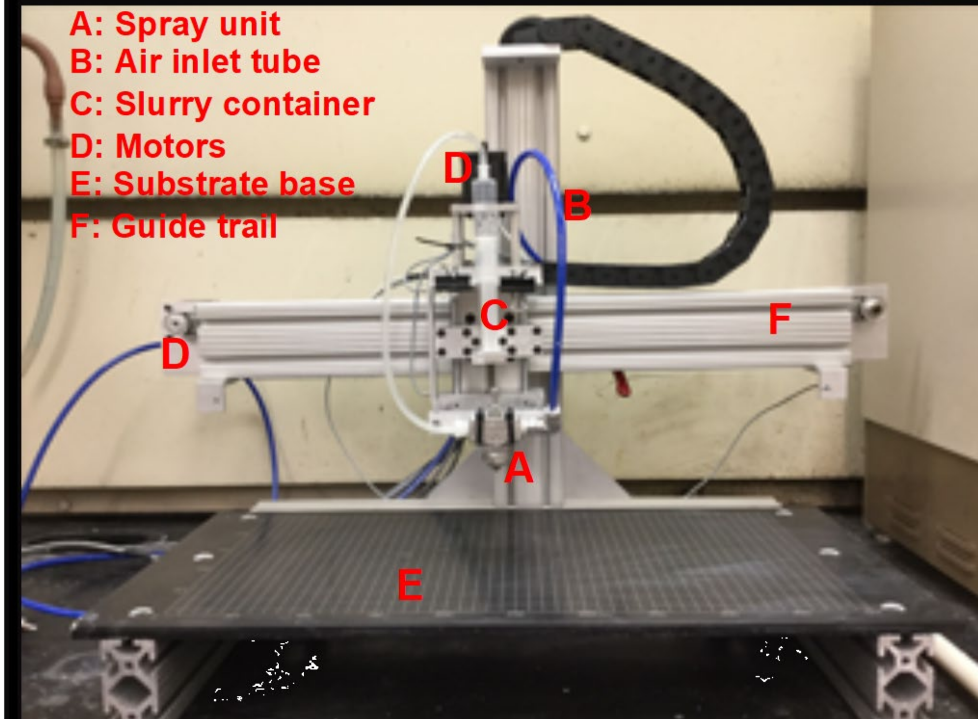
5. Project Progress-Task 4

Task 4: Processing and performance of graded coatings processed using cold spray and pyrolysis (30% complete)

Completed:

- The cold spray system to make coatings has been assembled
- Purchased the tube furnace for the investigation of oxidation response of the composites
- Developed the protocol of the Y_2O_3-SiCN submicrometer suspension
- Studied the microstructure uniformity of Y_2O_3-SiCN composite film from dip-coating

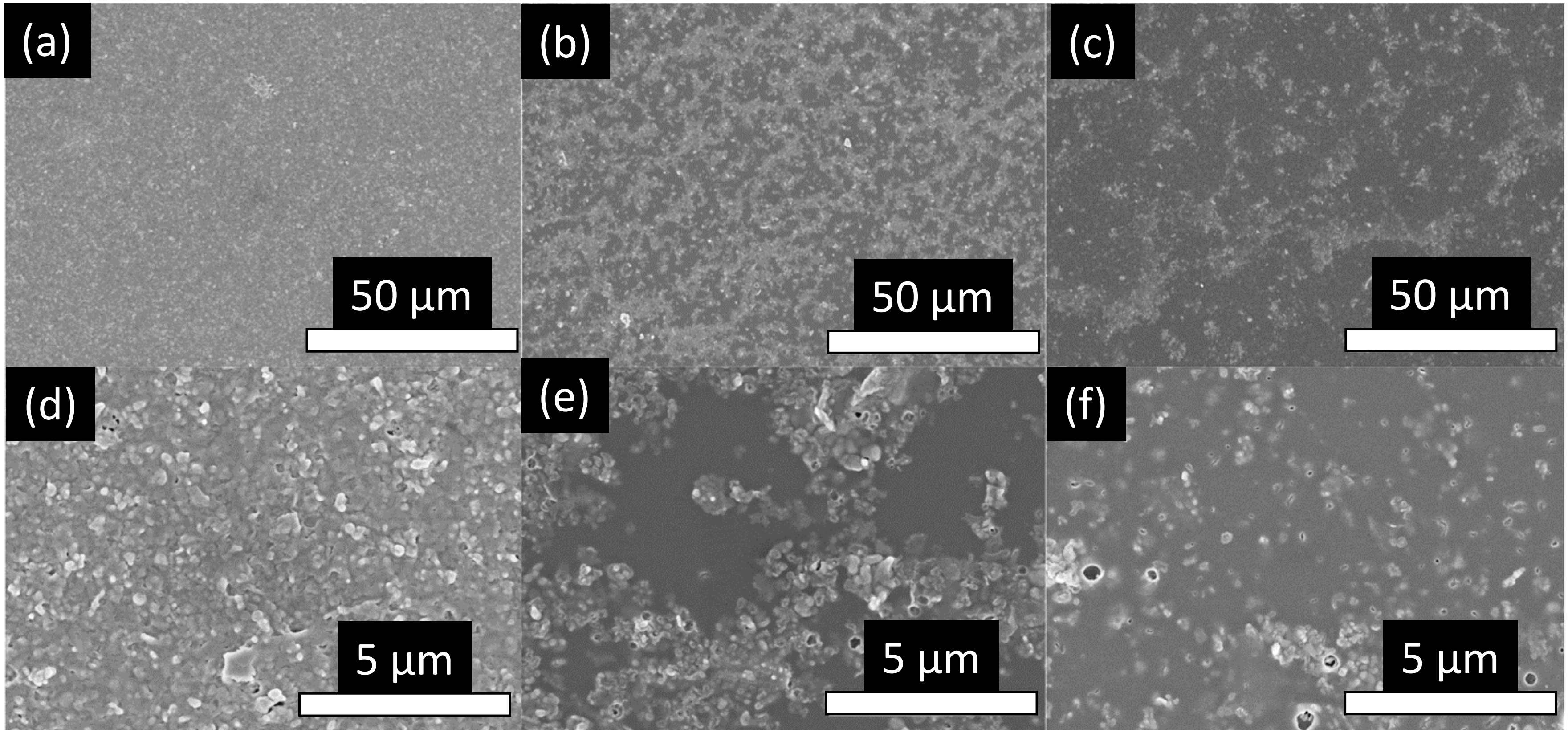
5. Task 4-Cold spray device



A controlled spray system to make coatings from liquid precursors or slurries

- Y_2O_3-SiCN coatings of all compositions (from 90%Y_2O_3-10%SiCN to 10%Y_2O_3-90%SiCN) have been fabricated

5. Task 4-Microstructure of sintered Y_2O_3-SiCN coatings



Y_2O_3-SiCN composite coating (dip-coated) after heat treatment at 1100°C. *80%Y_2O_3-20%SiC*: (a), (d); *50%Y_2O_3-50%SiC*: (b), (e); *20%Y_2O_3-80%SiC*: (c), (f).

5. Project Progress-Task 4

Task 4: Processing and performance of graded coatings processed using cold spray and pyrolysis (30% complete)

Future Plan:

- Fabricate graded coating using the cold spray method and characterize the microstructure

5. Project Progress-Task 5

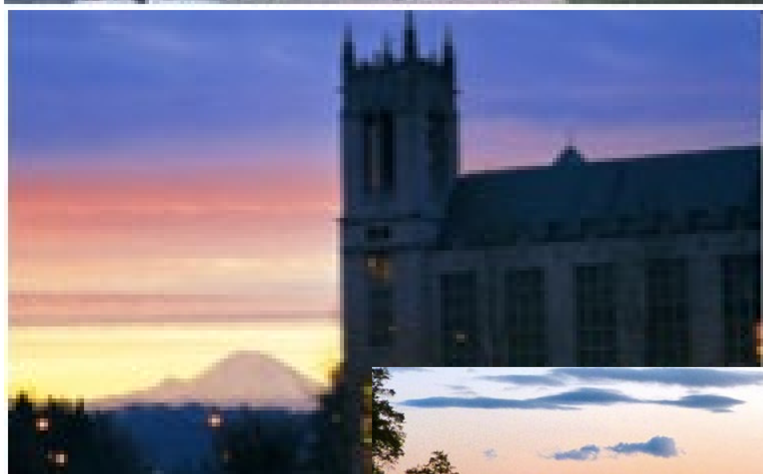
Task 5: Processing and performance of graded coatings processed using atmospheric plasma spraying (APS) (10% complete)

Current effort:

- We are preparing Y_2O_3-SiCN and Yb_2O_3-SiCN powder to be sent to GE for the initial APS trial. (5 lb each composition).

6. Project Short Term Plans

- Complete the microstructure characterization of all the sintered samples.
- Complete the thermal and oxidation response study for all compositions.
- Fabricate graded coating using the cold spray method and characterize the microstructure.
- We are preparing Y_2O_3-SiCN and Yb_2O_3-SiCN powder to be sent to GE for the initial APS trial. (5 lb each composition).



Thank you very much for your attention and support