Optimizing Heat Treatments of $Y/Yb_2Si_2O_7$ Environmental Barrier Coatings

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PRAXAIR SURFACE TECHNOLOGIES

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Ceramic Matrix Composites (CMCs) in gas turbine engines react with the atmosphere at high temperatures, and thus require protective Environmental Barrier Coatings (EBCs) to ensure a long service life. Yttrium-Doped Ytterbium Disilicate (Y/YbDS) EBCs are deposited with low crystallinity and must be heat treated to full crystallinity, during which thermal expansion generates cracks in the coating. In order to develop a heat treatment that minimizes stress generation, the effects of heat treatment time and temperature on cracking in Y/YbDS EBCs were investigated using X-Ray Diffraction and Indentation Fracture Toughness.

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Project Background

Silicon Carbide CMCs (SiC/SiC) enable higher operating temperatures in gas turbine engines, saving commercial airliners up to **$1,000,000 per year** and reducing environmentally harmful NO_X emissions by as much as 40% compared to traditional superalloy engines. However, SiC/SiC components are easily destroyed by water and Calcia-Magnesia-Alumina-Silicates (CMAS) at high temperatures, and require **Environmental Barrier Coatings** to prevent rapid degradation.

Yttrium-Doped Ytterbium Disilicate EBCs are commercially produced as coatings for SiC/SiC parts using Atmospheric Plasma Spray (APS), wherein constituent powders are vaporized and shot at a substrate. On contact with the substrate, the powders rapidly cool into a high density, amorphous "splat" microstructure containing both Yttrium-Doped Ytterbium Disilicate ($Y/Yb_2Si_2O_7$) and Monosilicate (Y/Yb_2SiO_5).

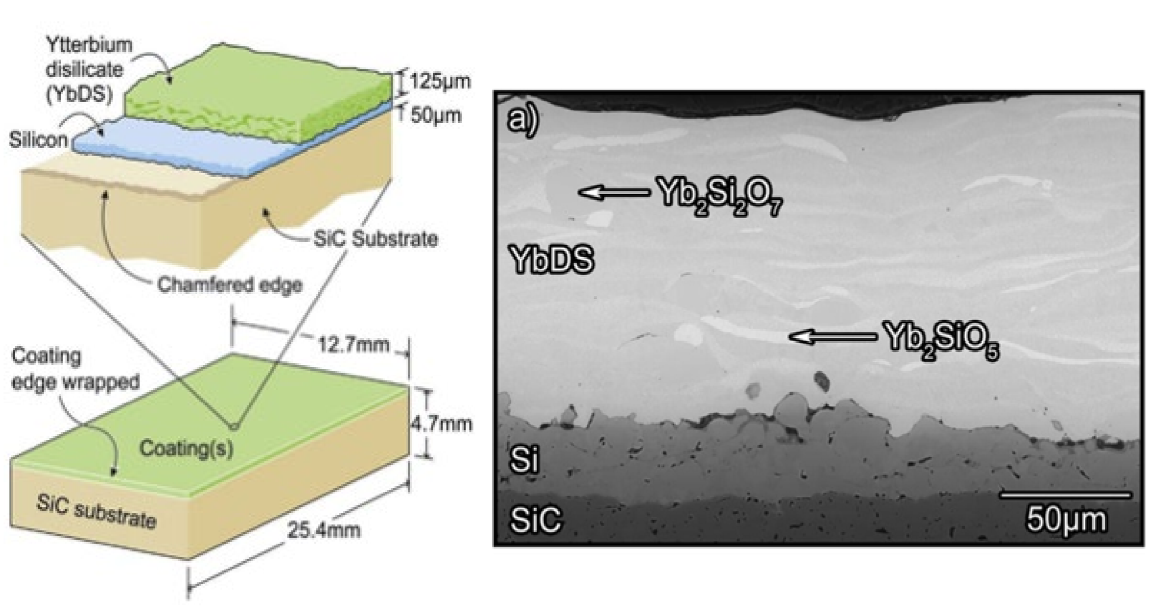

Figure 1: Diagram of an EBC bonding to a SiC/SiC substrate and the biphase splat microstructure of the coating [1].

The coatings must be **heat treated to full crystallinity** so that they prevent water vapor diffusion to the SiC/SiC part, withstand the intense operating conditions of the engine, and be phase-stable from rest to operating temperature. **The volumetric expansion due to crystallization generates stresses and cracks detrimental to the coating.** The objective of this project is to develop a heat treatment schedule that minimizes stress and crack development while achieving a fully crystalline Y/YbDS EBC.

Experimental Procedure

Heat Treatments: Eighteen coating samples were heat treated at hold temperatures ranging from 1100°C to 1300°C and hold times from 20 minutes to 20 hours. All samples were brought to temperature at a rate of 10°C/min and cooled by quenching in air.

XRD/Avrami Calculations: XRD spectra were collected with a Bruker D8 Focus and refined with MDI Jade 6 to determine each phase's crystallinity. The crystallinity, treatment time, and treatment temperature were used to derive each phase's Avrami constants, which allow estimation of crystallinity after a heat treatment.

Sample Preparation: Each sample was vacuum-mounted in epoxy and polished from 320 grit to 1µm. Samples were carbon coated (SPI Coater Module) and consequently imaged by SEM (Phenom ProX) and imageJ.

Indentation Fracture Toughness: Twenty indents were created on each sample using the LECO LM247 hardness tester to estimate cracking. A 500gF load was used on the as-deposited sample, and 1KgF load on the rest of the samples.

Experimental Procedure

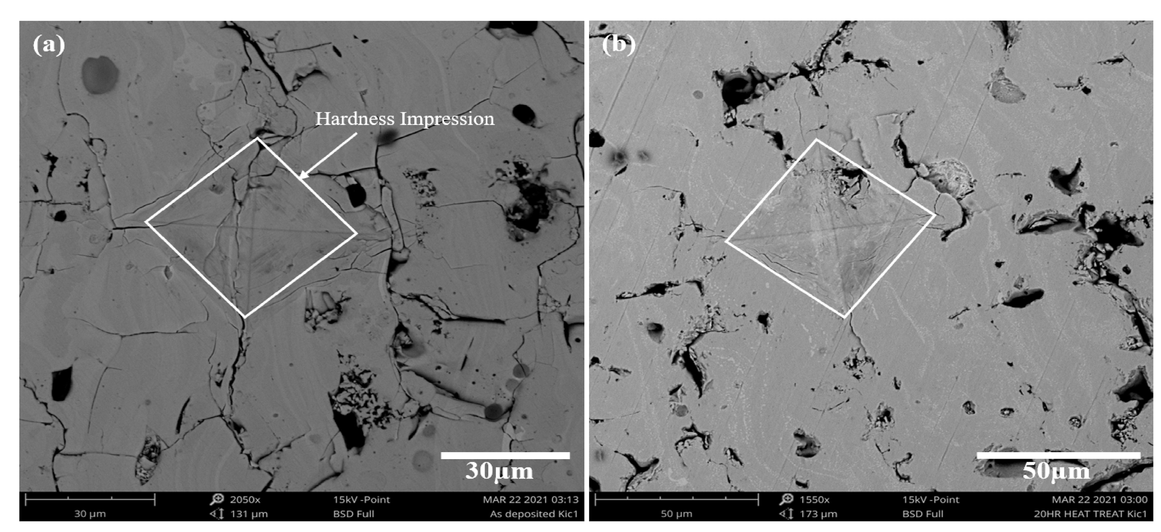

Figure 2: SEM Micrographs of the (a) as-deposited and (b) 20hr heat treated samples showing the hardness impression.

Hardness Analysis: SEM images of each indent were measured and used to approximate the coating's fracture toughness using the Shetty Relationship [2]. Vickers Hardness was determined using the hardness impression diagonal lengths.

Results

Analyzing the dependance of fracture toughness on the time and temperature of a heat treatment requires the **Larson Miller Parameter (LMP)**. The LMP differentiates between samples with different heat treatments that resulted in equal crystallinities - a higher LMP means a more heat treated sample.

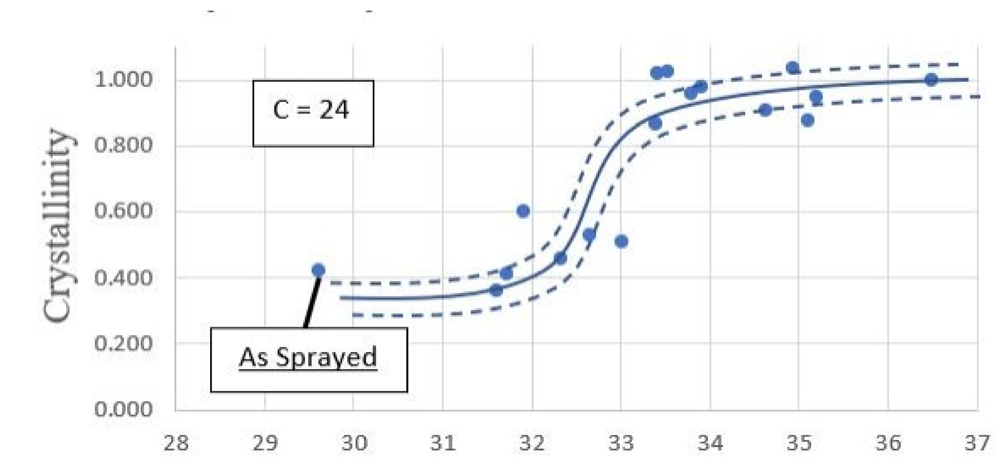

Figure 3: The LMP's and crystallinities of heat treated samples. Rapid crystallization occurs between LMP's of 75 and 76.

A phase's **Avrami parameters** give insight on crystallinity as a result of time and temperature. Once a treatment temperature is decided, these equations can be used to minimize the treatment time until full crystallinity, resulting in greater processing speeds.

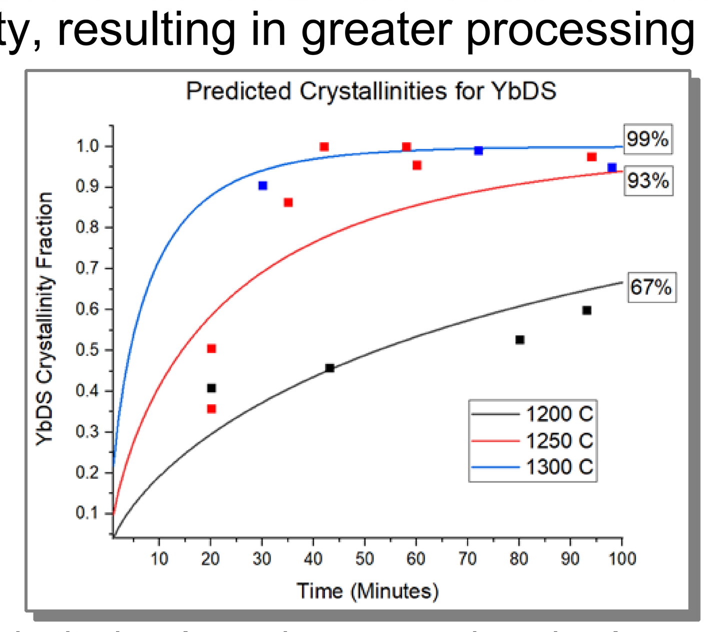

Figure 4: Substituting Avrami constants into the Avrami equation [3] estimates a phase's crystallinity after a heat treatment.

The LMP is a better descriptor of a heat treatment than crystallinity, and can be plotted with fracture toughness to determine heat treatment effects on the stresses and cracks developed in coating

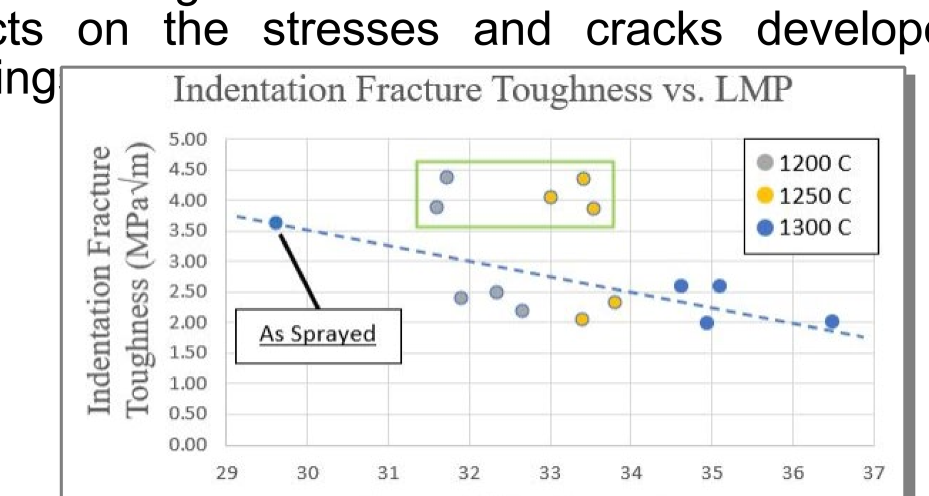

Figure 5: Indentation fracture toughness displays negative correlation with the Larson Miller Parameter. Low crystallinity samples (green) had higher fracture toughness.

Discussion

The Avrami parameters gave moderately accurate estimates of crystallinity as a result of heat treatment time and temperature. The model may be improved by more accurately refining XRD spectra and improving the accuracy of crystallinity calculations. This accuracy can be improved by normalizing all integrated peak areas with the peak area of a known amount of standard material placed in the XRD with the coating.

Fracture toughness and Vicker's Hardness were both **negatively correlated with the Larson Miller Parameter**. Interestingly, low crystallinity coatings generally showed higher fracture toughness. This is likely because low crystallinity coating phases had not yet been thermally cracked. That said, the splat microstructure of the coatings greatly complicated indentation fracture toughness measurements - it is possible that our methods produced inaccurate results.

A more complete understanding of how heat treatments impact cracking in a coating can be built by studying the effects of heating rate, cooling rate, cyclic treatments, and long-treatment sintering on the fracture toughness of Y/YbDS coatings. It is expected that lower heating rates will result in higher fracture toughness by easing the rate of thermal expansion, and that treatment at high temperatures for long times would increase the fracture toughness once microcracks in the coatings began to heal through sintering.

Recommendations

Our studies have shown that heat treatments resulting in a lower LMP improve the fracture toughness of coatings. We suggest that Rolls Royce and Praxair Surface Technologies heat treat Y/YbDS EBC's at 1250°C, for 150 minutes in order to efficiently achieve the toughest full crystallinity coatings. Crack sintering in high LMP heat treatments, stress relaxation holds, and heating and cooling rate are important steps of heat treatments and deserve further research investment.

References

[1] Richards, B. T., Young, K. A., Francqueville, F. D., Sehr, S., Begley, M. R., & Wadley, H. N. (2016). Response of ytterbium disilicate–silicon environmental barrier coatings to thermal cycling in water vapor. Acta Materialia,106,1-14.doi:10.1016/j.actamat.2015.12.053

[2] GmbH, Fritz Mueller. (2007). "Software Validation Certificate." *PRECIDUR V3.89 2007-JAN-08*, Fritz Mueller GmbH, Muenchen, Deutschland, files.engineering.com/download.aspx?folder=b2d8ff40-ecbd-49e7-9e55-655dbcfeab2e&file=Fracture_Toughness_Calcs.pdf.

[3] Ohring, M., & Kasprzak, L. (2015). The Mathematics of Failure and Reliability. *Reliability and Failure of Electronic Materials and Devices*, 181–248. https://doi.org/10.1016/b978-0-12-088574-9.00004-5

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