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TRISO COATING DEVELOPMENT PROGRESS FOR URANIUM NITRIDE KERNELS

Fuel Cycle Research & Development Advanced Fuels Campaign

Brian Jolly
Terrance Lindemer
Kurt Terrani

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TRISO COATING DEVELOPMENT PROGRESS FOR URANIUM NITRIDE KERNELS

Brian Jolly, Terrance Lindemer, Kurt Terrani
Oak Ridge National Laboratory, Oak Ridge, TN, 37831-6063
jollybc@ornl.gov

INTRODUCTION

In support of fully ceramic matrix (FCM) fuel development [1-2], coating development work is ongoing at the Oak Ridge National Laboratory (ORNL) to produce tri-structural isotropic (TRISO) coated fuel particles with UN kernels [3]. The nitride kernels are used to increase fissile density in these SiC-matrix fuel pellets with details described elsewhere [4]. The advanced gas reactor (AGR) program at ORNL used fluidized bed chemical vapor deposition (FBCVD) techniques for TRISO coating of UCO (two phase mixture of UO_2 and UC_x) kernels [5]. Similar techniques were employed for coating of the UN kernels, however significant changes in processing conditions were required to maintain acceptable coating properties due to physical property and dimensional differences between the UCO and UN kernels (Table 1).

Table 1: Comparison of UCO and UN kernel properties

	AGR-1 (UCO)	UN kernels
Nominal diameter (µm)	350	800
Density (g/cc)	10.92	14.4*
Specific heat [J/g-K]	>0.3	0.25
Thermal conductivity (W/mK)	<5	15.5*

*Literature values at 1000 °C. Actual values have not been measured.

Initial coating development work focused on re-establishing TRISO coating capability. Existing equipment was refurbished/recalibrated, and a demonstration experiment was successfully completed depositing all four coating layers on UN kernels. While functionality of the coating equipment was confirmed, none of the coating layers have been optimized to meet the property specifications. (Note that initial target specifications for coating properties will be similar to those of AGR-1 [6]) Also, the UN material available at the time of the initial TRISO coating experiment was only ~70% theoretical density and not suitable for repeatable coating development work due to inconsistencies. However, recent UN kernel fabrication has shown excellent batch to batch repeatability with good theoretical densities of 85-87%. With the new, higher quality, UN kernels and the coating equipment refurbished, coating development has begun in earnest.

Experimental Procedures

Equipment

The FBCVD coating furnace (figure 1) used consists of a conical graphite coating chamber surrounded by a resistively heated graphite element which are both housed within a water cooled shell. Process gases are delivered through a water cooled injector which supports the coating chamber. Kernels are loaded through the top of the furnace at room temperature with argon gas flowing through the injector to maintain fluidization during heat up. The furnace is brought to coating temperature and the individual layers are deposited by switching between precursor gasses. Once the coating experiment is complete, the furnace is brought back to room temperature and gas flow is shut off allowing the kernels to drain through the injector and into the catch cup fixed to the bottom of the injector.

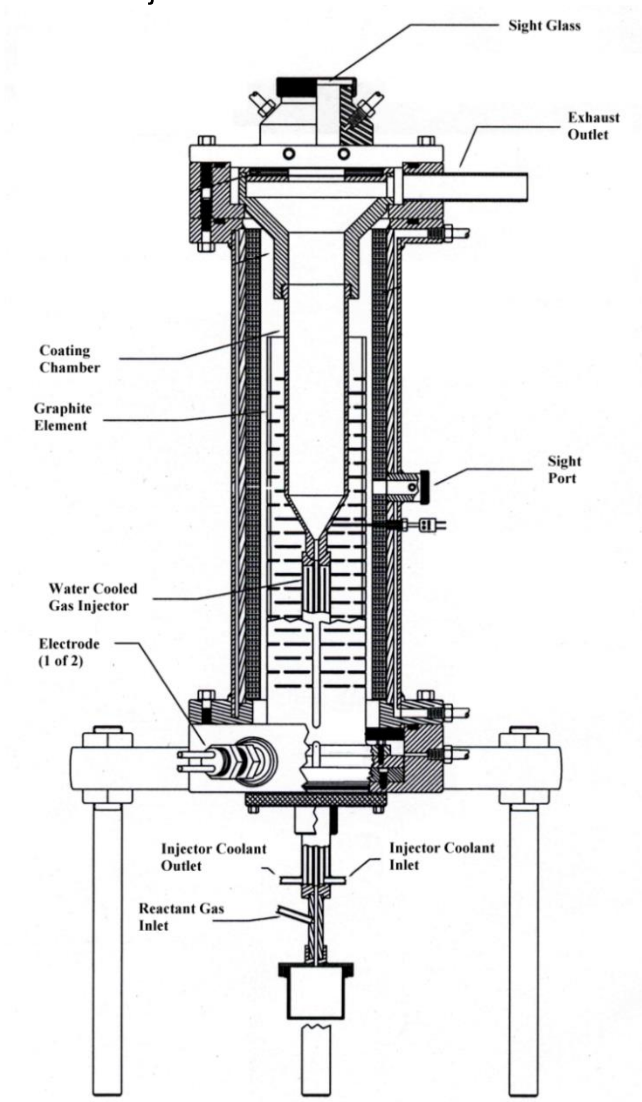

Figure 1: Schematic of FBCVD coating furnace used for TRISO coating of UN kernels

Temperature of the fluidized bed is measured directly with a hand held optical pyrometer which looks though the top of the furnace via a sight glass and prism. Signal attenuation from the sight glass/prism set is measured and the appropriate correction factor applied. Gas delivery is controlled using electronic mass flow controllers which are periodically calibrated using a wet test meter.

RESULTS AND DISCUSSION

The buffer layer is the first coating layer of a TRISO particle. It is a low density carbon layer (~1.0g/cm^{3,} ~50% TD) designed to provide room for fission gas buildup and allow compliance to mitigate stresses induced from thermal expansion mismatches between the kernel and other coating layers.

From visual comparison, the buffer layer deposited during the initial "shakedown" UN coating experiment had a higher density than desired (**Figure 2**). However, buffer density cannot be measured quantitatively with the subsequent TRISO layers deposited on top of it, so buffer only coating experiments are required.

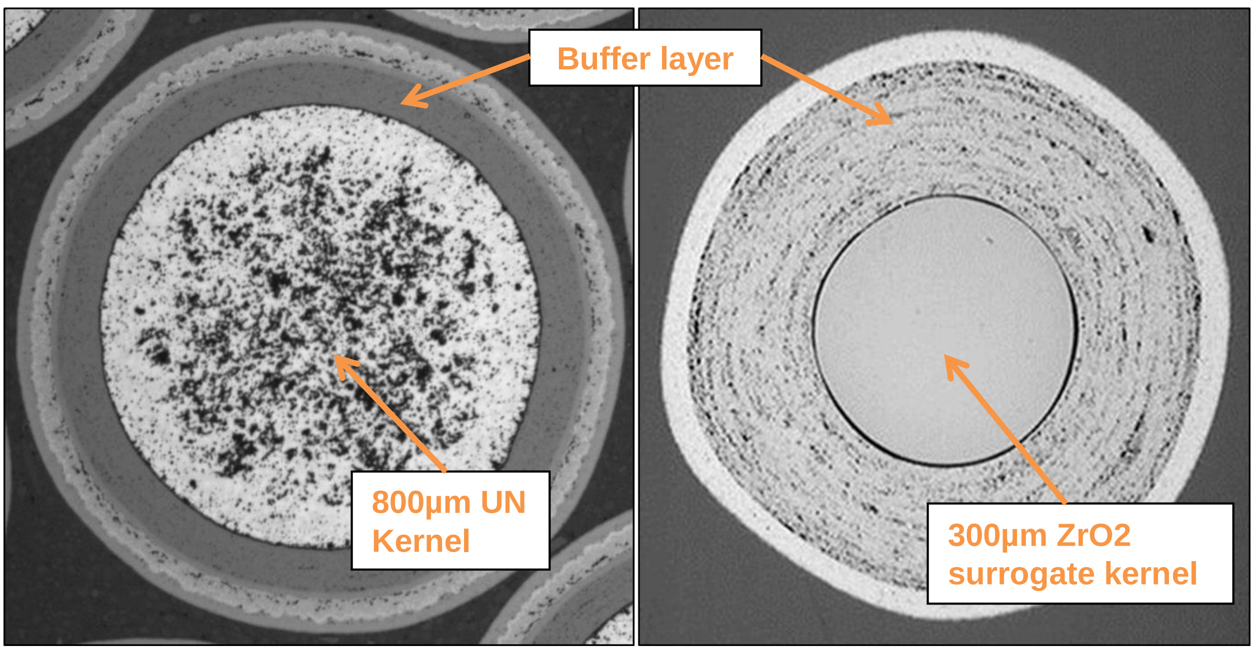

Figure 2: visual comparison of buffer density from initial UN TRISO coating run (left) and archived particle with buffer density within specification (right)

Initial buffer coating experiment using UN kernels

As buffer density was likely too high for the initial UN TRISO experiment, deposition temperature was increased slightly from 1450°C to 1500°C for the first buffer only coating experiment (FCM-3A-UN18K-01B) in an attempt to decrease density. **Figure 3** shows an example of a cross sectioned UN particle from the first buffer only coating experiment. Visually the buffer density still appears too high, and this is confirmed using mercury porosimetry techniques for quantitative analysis measuring an average density value of 1.3g/cm^3 compared to the target density of 1.0g/cm^3.

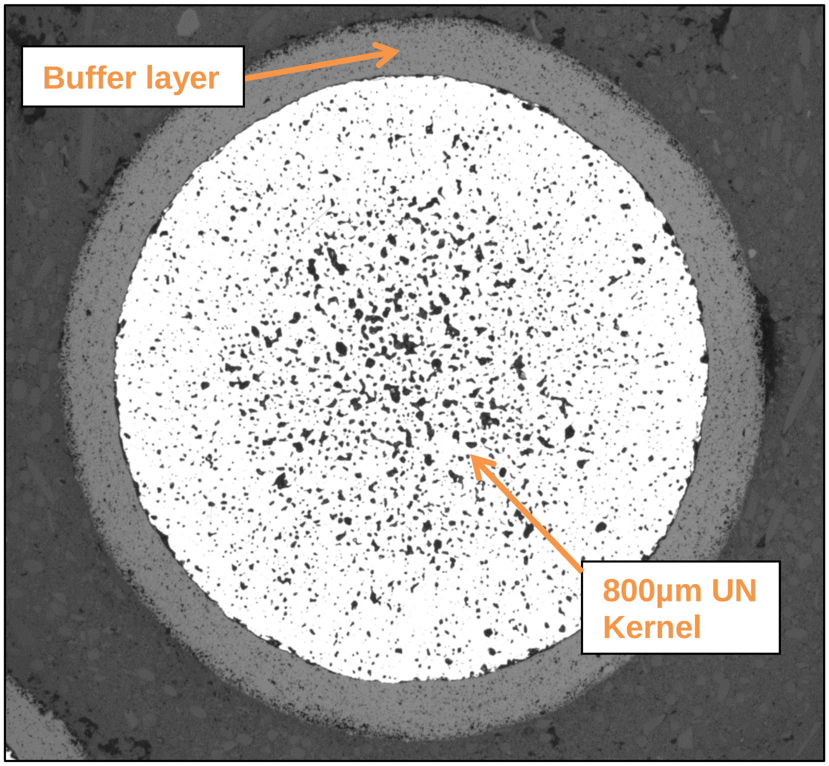

Figure 3: Cross sectioned UN particle from buffer only coating experiment FCM-3A-UN18K-01B. Buffer density 1.3g/cm^3

Buffer density study using ZrO_2 surrogate kernels

In the beginning of the coating development process, it was planned to use temperature as the primary tool to adjust coating properties. Temperature affects the kinetics of the CVD reaction and provides a very fine adjustment allowing precise control of the coating properties, such as density. For buffer depositions, typically higher temperatures result in lower density coatings, and lower temperatures result in higher density coatings. When reviewing data developed during the earlier AGR work regarding buffer density as a function of temperature (**Figure 4**), it soon became evident temperature alone wouldn't be sufficient for adjusting density as temperature required would exceed the capabilities of the coating furnace considering the first buffer only experiment using UN kernels had a buffer density of 1.3g/cm^3 at a deposition temperature of 1500°C. Therefore, a more coarse density adjustment is necessary.

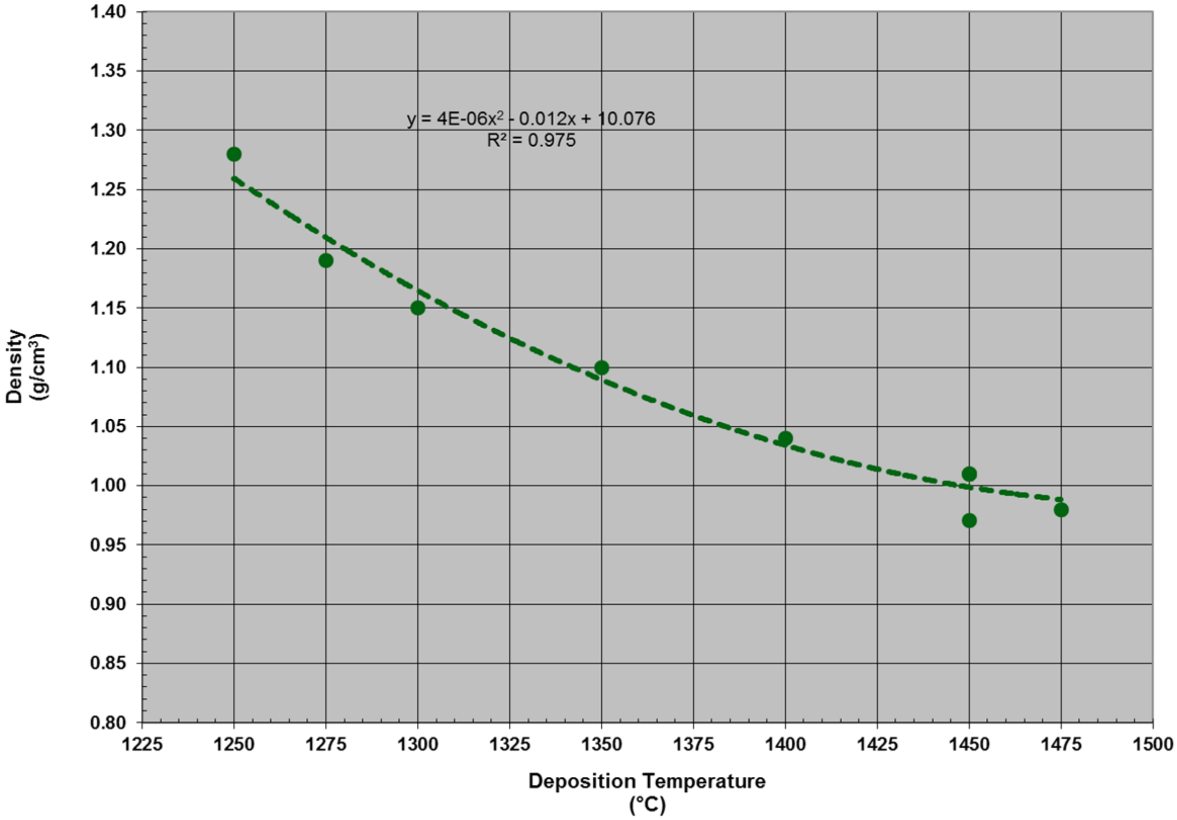

Figure 4: Buffer density trends as a function of temperature

Coating gas fraction (CGF), defined as precursor gas flow divided by total gas flow, has a strong influence on buffer density. A study was conducted to quantify the effect of CGF on buffer density. As multiple experiments were necessary for this study, 800µm diameter ZrO_2 microspheres were used as a surrogate for the UN kernels in order to conserve the available UN material. While coating properties as a function of processing conditions can't be duplicated perfectly when using ZrO_2 as a surrogate, due to thermo-physical property differences, trends can be established and used to guide the UN coating. **Figure 5** shows the results from the surrogate buffer study as well as the buffer density when using UN kernels for reference. Visual comparisons of the buffer densities from the surrogate runs can be seen in **Figure 6**. From this surrogate study, it appears the target buffer density of 1.0g/cm^3 can be met by increasing the CGF to .58 and making a slight temperature adjustment. Future UN kernels coating experiments will use this technique to achieve the desired buffer density.

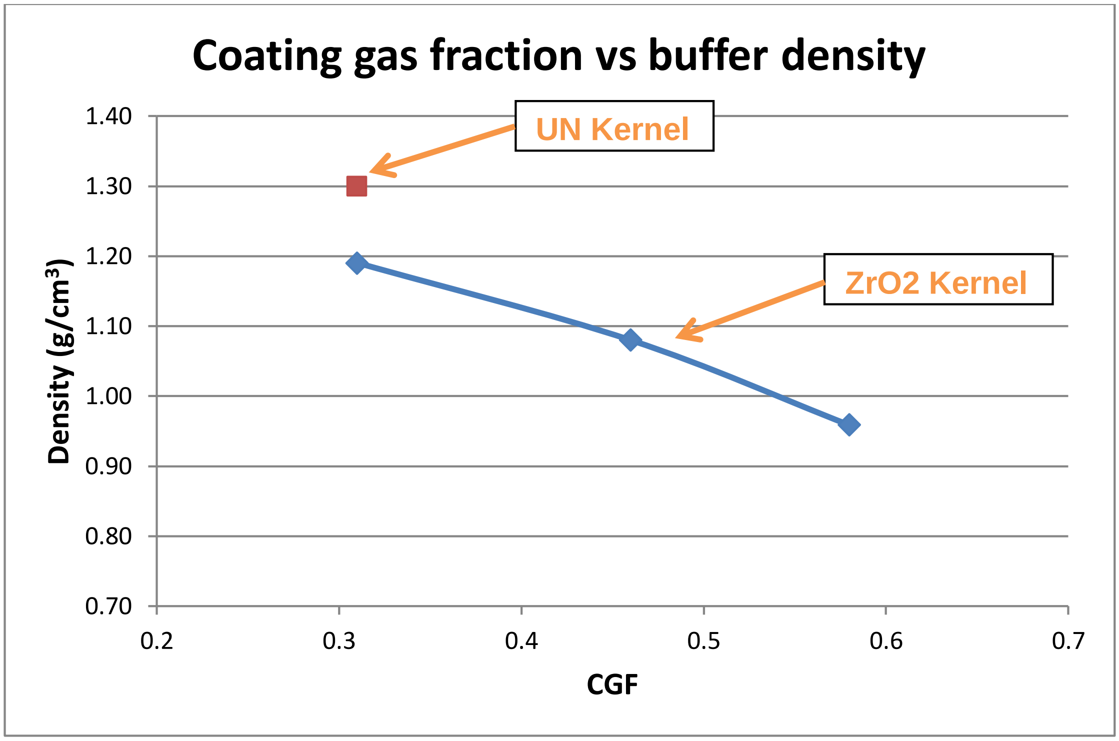

Figure 5: Buffer density as function of coating gas fraction at 1500°C

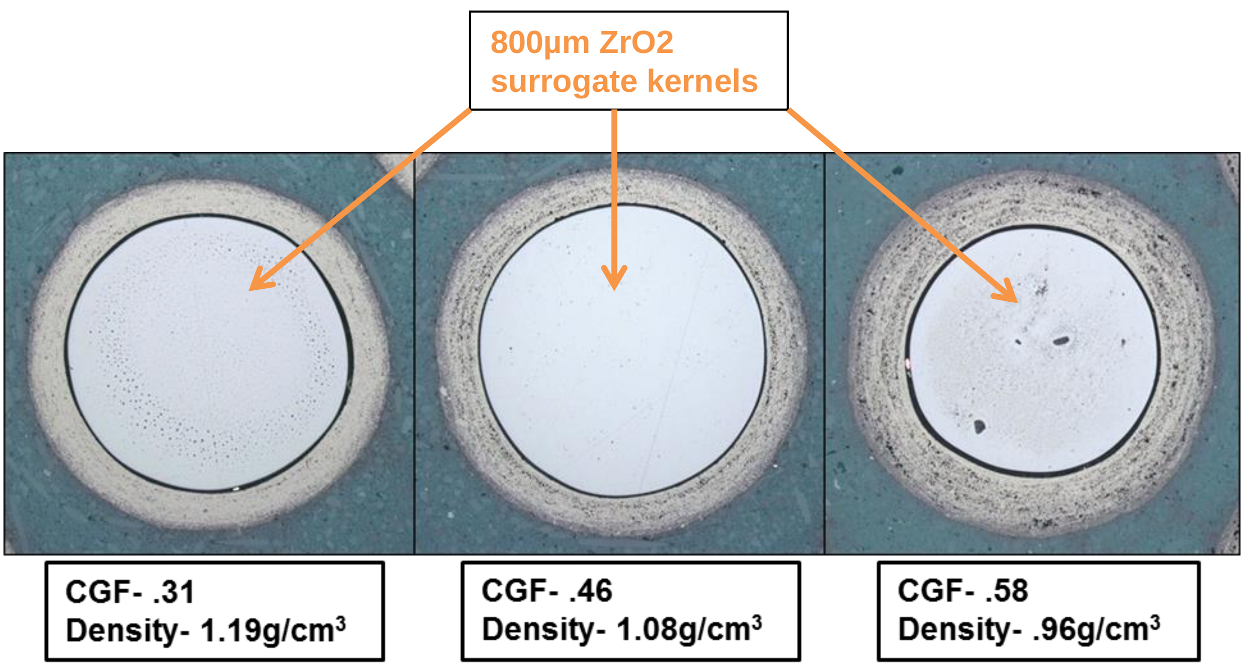

Figure 6: Cross sectioned buffer coated ZrO2 from surrogate buffer study

SUMMARY

A coordinated effort is underway to produce TRISO coated fuel particles from recently processed uranium mononitride kernels at ORNL. These particles will be used in subsequent irradiation tests of the FCM fuel concept under prototypic LWR conditions.

Now that the FBCVD coating furnace has been successfully refurbished and consistent higher density UN kernels are available, coating development is well underway. Initial UN buffer coating experiments produced coatings with unacceptably high densities. However, parameters to correct this problem have been identified using surrogate material and will now be applied to the UN kernels.

ACKNOWLEDGMENTS

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