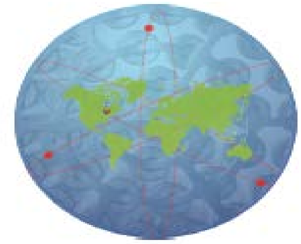

PELLET FABRICATION DEVELOPMENT OF ANNULAR PELLETS FOR INTERNALLY COOLED ANNULAR FUEL

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ABSTRACT – Annular pellets are a variant of the standard solid cylindrical pellet, the difference being the presence of a void along the cylindrical axis. Fabrication of such pellets is common and methods are well understood. Internally cooled annular fuel (ICAF) uses annular pellets clad both externally and internally so that coolant can flow through the centre as well as around the outside of the element. In order to maximize the heat transfer to the coolant flowing through the centre, the gap between the inside diameter of the pellet and the inner cladding must be minimized. This paper describes fabrication development efforts towards producing annular pellets with precise and accurate inside diameters in order to minimize the pellet-inner cladding gap for internally cooled annular fuel.

Introduction

Internally cooled annular fuel (ICAF) consists of annular fuel pellets clad on both the outside and inside circumferential pellet surfaces. Coolant can flow around the cladding on the outside of the pellets as well as through the channel formed by the cladding on the inside of the pellets. A schematic contrasting typical cylindrical solid fuel and ICAF is provided in Figure 1.

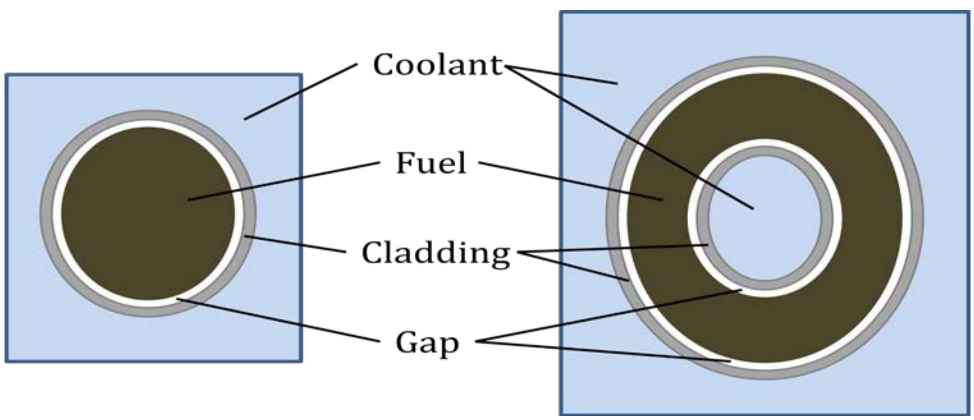

Figure 1: Cross-Sectional View of Cylindrical Solid Fuel and ICAF [1]

Based on promising studies of ICAF in fuel assemblies for pressurized water reactors (PWR) and boiling water reactors (BWR) [1][2], an ICAF bundle design meant for high-burnup applications in existing CANDU reactors is being assessed at Atomic Energy of Canada Ltd. (AECL). Part of the assessment includes development of pellet fabrication methods with tight control over the sintered pellet inner diameter so that the gap between the inside diameter of the pellet and the inner cladding can be minimized for better heat transfer without post-sintering processing.

Control of the outside diameter of typical power reactor fuel pellets is achieved by centerless grinding. This is a simple, rapid, and mature process used throughout the nuclear fuel fabrication industry. There is no equivalent process for the finishing of the inside diameter of a hard, dense annular ceramic pellet. Previous attempts to control the sintered pellet inner diameter have been reported in the open literature [3][4][5]. In one approach, a green annular pellet was pressed using conventional double-ended pressing. A precisely machined metal rod was then inserted into the void of the pellet and the green compact was pressed again in a cold isostatic press. The metal rod acted as a support for the green pellet during cold isostatic pressing and ensured that the inner diameter remained uniform across the length of the pellet. The cold isostatic pressing step served to improve the homogeneity of the green compact density so that pellet shrinkage during sintering was isotropic. The achieved tolerance on the sintered pellet inner diameter was ±0.012 mm [3]. In another approach, a green annular pellet was first compacted using conventional double-ended pressing. A precisely machined zirconia rod was then inserted into the void of the pellet prior to sintering. The rod acted as a stop for pellet shrinkage during sintering. The achieved tolerance for the sintered pellet inner diameter was ±0.003 mm [4]. In a similar study, a UO_2 rod was used instead of the zirconia rod as a stop for pellet shrinkage during sintering. The achieved tolerance for the sintered pellet inner diameter was ± 0.006 mm [5]. In this study, nearly fully dense, centerless ground, sintered UO_2 core pellets were used as a stop for annular pellet shrinkage during sintering.

1. Experimental

Natural uranium dioxide powder derived from the ammonium diuranate route (Cameco Corp.) was pre-pressed at 6700 psi in a 3.2 cm diameter die resulting in a compact density of 3.7 g/cm^3. The pre-pressed compacts were granulated by forcing them through a standard #12 stainless steel sieve using a porcelain pestle. Zinc stearate (0.2 wt. %) was mixed in with the granules before final pressing.

Annular fuel pellets were pressed from lubricated granules using a hydraulic press and the tooling pictured in Figure 2.

Figure 2: Annular Pellet Tooling

The core rod formed the void of the annular pellet during pressing. The core rod and die body were both "floating" during pressing to simulate double ended pressing.

Annular pellets were sintered in a refractory metal furnace at 1650°C for 4 hours in flowing 100% hydrogen.

Core pellets to act as stops for annular pellet shrinkage during sintering were cylindrical natural UO_2 pellets sintered, resintered, and centerless ground to an outside diameter of 8.507±0.002 mm.

2. Results and Discussion

The objective of this study was to control the inner diameter of annular fuel pellets during sintering using dense, precisely machined cylindrical UO_2 core pellets. The core pellets were meant to restrict shrinkage of the annular pellet during sintering to yield a uniform inner diameter. UO_2 core pellets were chosen to avoid any chemical interaction with the annular pellet at high temperature and to avoid complications due to differences in the linear thermal expansion coefficients of the materials. Each annular pellet contained two core pellets during sintering. Two core pellets were required so that their total length exceeded the length of the annular pellet.

One must consider the degree to which the annular pellet shrinks during sintering with core pellets inside. If the annular pellet shrinks beyond the dimensional constraint imposed by the core pellet, it is likely to crack. If the annular pellet does not shrink down to the core pellet, then the core pellet does not have any influence on the final inner diameter. The amount of shrinkage during sintering can be influenced by many variables but the most influential is the green density. Pellets with higher green densities exhibit less shrinkage upon sintering. In Test #1 of this study, annular pellets with green densities ranging from 4.88 to 5.45 g/cm^3 were pressed and sintered with core pellets to determine the green density that would result in optimal shrinkage. Pictures of the pellets before and after sintering are provided in Figures 3 and 4.

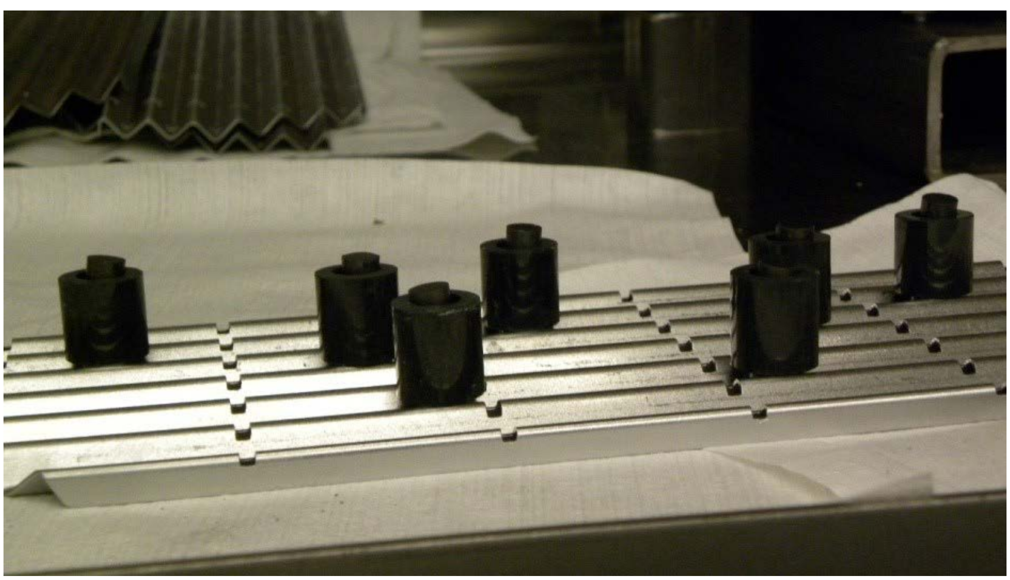

Figure 3: Test #1 Green Annular Pellets with Sintered Core Pellets Inside Prior to Sintering

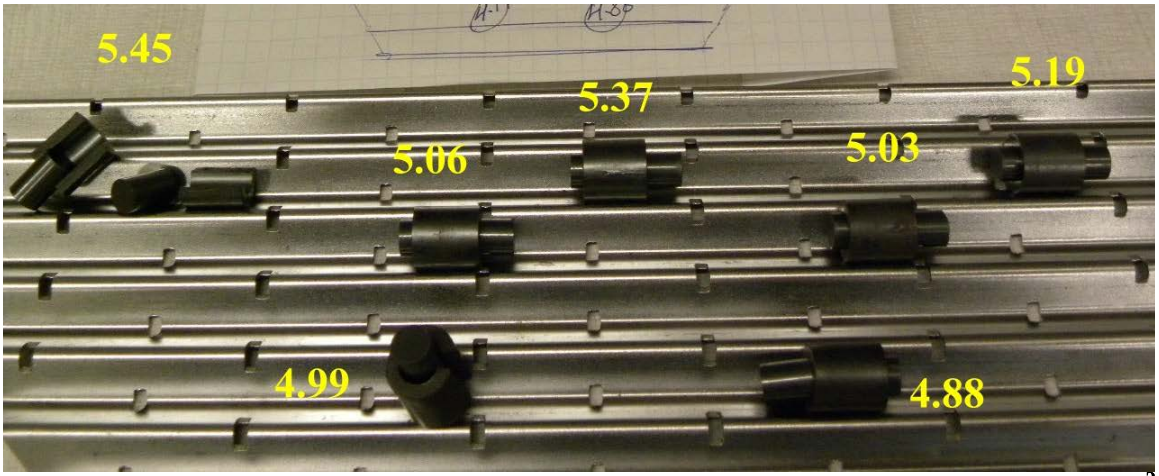

Figure 4:Test#1 Sintered Pellets- green densities for the pellets are indicated in g/cm^3

Green pellets were arranged vertically on corrugated molybdenum trays for sintering in Test #1. The shrinkage during sintering caused all but one of the pellets to fall over in the trays as shown in Figure 4. Pellets with densities below 5.19 g/cm^3 cracked as they shrank beyond the core pellets. The pellet with a green density of 5.37 g/cm^3 was intact but the pellet with a green density of 5.45 g/cm^3 was cracked. Since the pellet with a higher density should shrink less, it was surmised that the 5.45 g/cm^3 green density pellet broke as a result of falling over in the tray rather than shrinking beyond the core pellet. From Test #1, a green density range of 5.3-5.4 g/cm^3 was selected for the next pressing test.

For Test #2, annular pellets were pressed to a green density of 5.3-5.4 g.cm^3 and sintered in a horizontal orientation so that the pellets would not fall over during sintering. The green pellet inner diameters ranged from 10.68 to 10.73 mm. Eight pellets contained core pellets and two did not. The sintered pellets are pictured in Figure 5.

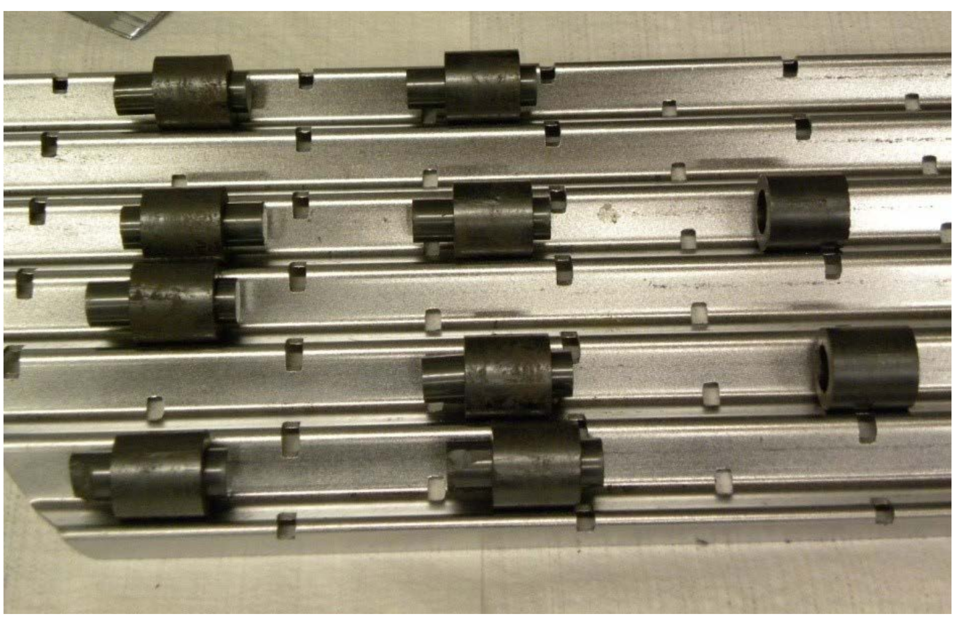

Figure 5: Test#2 Sintered Pellets

Assuming a uniform green density, the sintered annular pellet inner diameter can be estimated using the following equation:

$$Sintered\ Pellet\ ID = Green\ Pellet\ ID * \left[\frac{Green\ Density}{0.98 * Sintered\ Density}\right]^{1/3} \quad (1)$$

The 0.98 factor in the denominator accounts for pellet mass loss due to UO_{2+x} reduction during sintering and was determined from previous sintering tests with the UO_2 powder used. Assuming a green pellet inner diameter in the middle of the measured range (10.705 mm), Figure 6 plots Equation 1 as a function of green density for a range of sintered pellet densities.

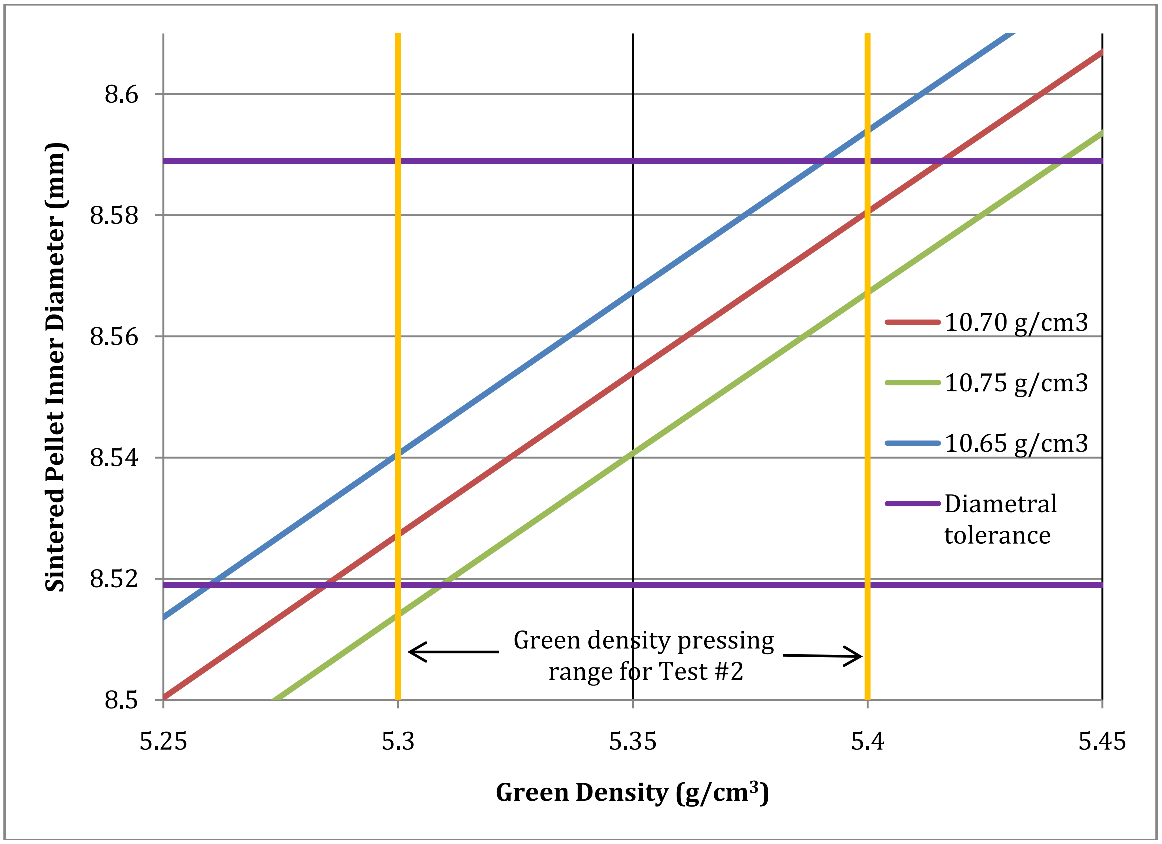

Figure 6: Sintered Pellet Inner Diameter as a Function of Green Density

Also plotted on Figure 6 are lines indicating diametral tolerance. These lines are centered around 8.554 mm, the sintered pellet inner diameter corresponding to the middle of the green pellet density range for Test #2 (5.35 g/cm^3) and a sintered pellet density of 10.70 g/cm^3. The diametral tolerance lines represent a range of 70 microns which matches the diametral clearance range for CANDU fuel. As can be seen from Figure 6, some of the predicted sintered pellet inner diameters for the high and low sintered density cases fall outside the diametral tolerance range near the limits of the 5.3-5.4 g/cm^3 green density range. For all of the sintered pellet inner diameters to fall within the diametral tolerance range, the range of sintered pellet densities must be narrower, in this case, between 10.67 and 10.73 g/cm^3. The sintered annular pellets in Test #2 had sintered densities between 10.69-10.73 g/cm^3. Therefore, all the sintered pellet inner diameters were expected to fall within the diametral tolerance range. Although the core pellet outside diameter (8.507±0.002mm) is outside the predicted sintered pellet inner diameter range, the observation from Test #1 was that the annular pellet pressed to a green density of 5.37 g/cm^3 had shrunk down to the core pellets. This will be discussed later in the context of green density variation within the pressed pellets.

After sintering, the core pellets were removed from the annular pellets from Test #2. Some of the core pellets were removed easily by hand while others required greater force to remove, specifically those from pellets 7 and 8. The inner diameters of the pellets were measured at the pellet ends and in the middle with a three anvil bore micrometer. At each axial position the inner diameter was measured four times. For the second, third, and fourth measurements, the pellet was rotated about its axis 30°, 60°, and 90°, respectively, with respect to the position of the first measurement. Figure 7 shows the complete set of measured inner diameter data for the ten annular pellets from Test #2. The inner diameter profiles for the pellets are presented in Figure 8 along with lines indicating the average of the measurements and the standard deviation.

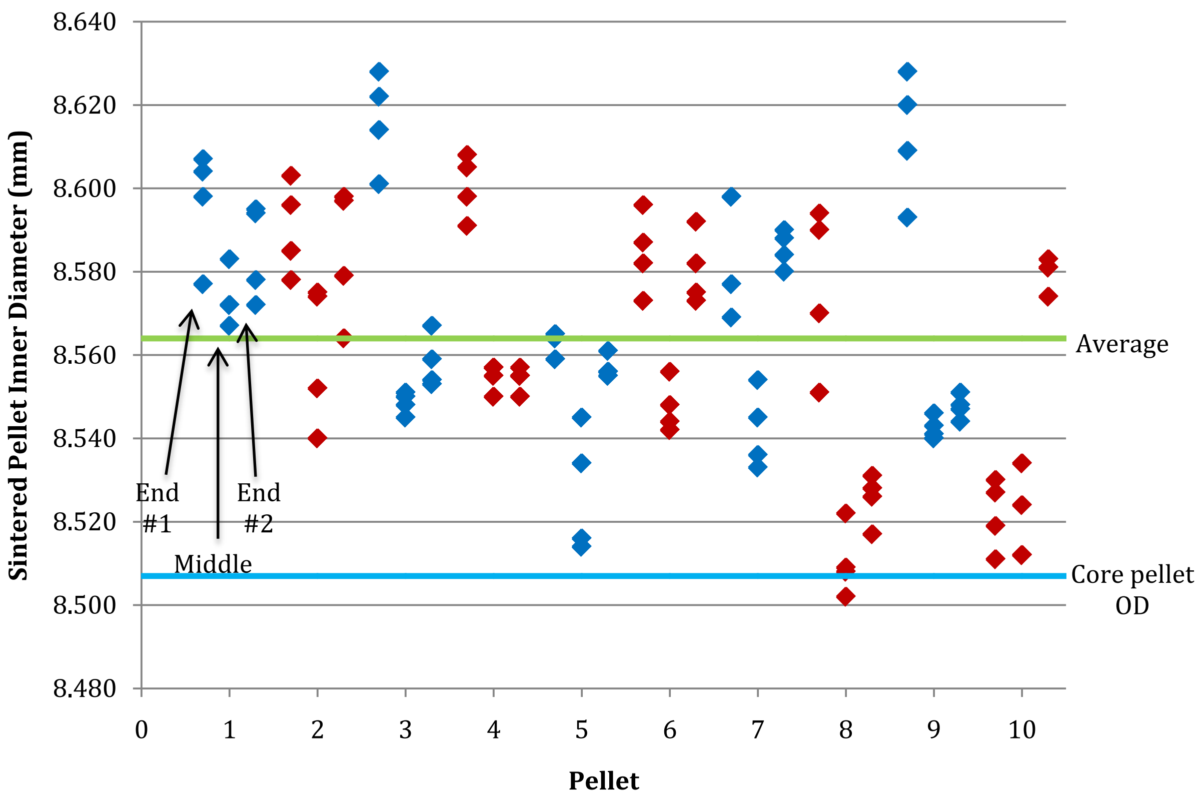

Figure 7: Sintered Pellet Inner Diameters for Test #2

As can be seen in Figure 7, the average sintered pellet inner diameter from Test #2 is within 10 microns of that predicted from Figure 6. However, the spread of the data was 126 microns, larger than the predicted 70 micron range and the diametral tolerance range. For this to have occurred, the green density range of the pellets must have been wider than that indicated on Figure 6. This is plausible given that the green densities measured were for the bulk pellet. In actuality, there were regions of higher and lower density in the green pellet. The green density inhomogeneity in the pellet results from pressing pressure variations due to friction between powder particles and friction between the powder and the die wall during pressing.

Another observation is that the data for the sintered annular pellets with core pellets are not distinguishable from those without core pellets. The annular pellets that were sintered without core pellets have inner diameters within the range of the remainder of the data. The spreads of the inner diameter data for the individual pellets is also similar for pellets sintered with core pellets as those sintered without. From Figure 7, it would appear that the core pellets did not influence the inner diameter of the sintered annular pellets. The measured inner diameters for the pellets that contained core pellets (8.502-8.628 mm) were all larger (with the exception of one data point for pellet #8, 8.502 mm) than the outside diameters of the core pellets (8.507±0.002 mm). However, some of the core pellets were difficult to remove after sintering, suggesting that the annular pellets shrank down to the core pellets. Perhaps the locations of the inner diameter measurement did not correspond to the minimum inner diameter of the pellet. To determine the minimum inner diameter range of the pellets, steel gauge plugs of known diameter (8.46, 8.50, 8.51 and 8.54 mm) were tested for passage through the void of the pellets. The minimum inner diameter range is defined as being between the outside diameters of the gauge plug that could pass and the one that could not. Table 1 shows the minimum inner diameter range for the pellets.

Table 1: Minimum Inner Diameter Range for Pellets from Test #2

Pellet #	Minimum ID Range (mm)
1	8.50-8.51
2	8.50-8.51
3	8.51-8.54
4	8.51-8.54
5	8.50-8.51
6	8.50-8.51
7	8.46-8.50*
8	8.50-8.51**
9	8.50-8.51
10	8.50-8.51

*The 8.50 mm gauge plug could be inserted so that the end of the gauge plug was flush with the opposite end of the pellet, but the entire length of the gauge plug would not pass through the pellet
**The 8.51 mm gauge plug could be inserted so that the end of the gauge plug was flush with the opposite end of the pellet, but the entire length of the gauge plug would not pass through the pellet

Table 1 shows that six out of the eight pellets had minimum inner diameter ranges that included the outside diameter range of the core pellets or were smaller. Pellets 3 and 4 allowed the 8.54 mm gauge plug to pass through. This data suggests that these pellets did not shrink down to the core pellets. This is supported by the data in Figure 7 which shows that neither of these pellets had measured inner diameters less than 8.54 mm. The minimum inner diameter range for pellet 5 included the outside diameter range of the core pellets. It also had a measured inner diameter within 7 microns of the outside diameter of the core pellets. This data indicates that pellet 5 likely had some contact with the core pellets. Pellet 7 appears to have shrunk beyond the core pellet outer diameter from the gauge plug data. Perhaps one or both of the core pellets shrank during sintering. Remeasurement of the core pellet outer diameters confirmed that some of the outer diameters were smaller by a few microns. The gauge plug results would seem to contradict the measured inner diameter data for pellet 7, however, it was difficult to remove the core pellets after sintering, implying that the gauge plug result is accurate. Pellet 8 had a minimum inner diameter range that included the outside diameter range of the core pellets. It also had an inner diameter data point below the outside diameter of the core pellets as well as other measured data only slightly greater than the outside diameter of the core pellets. The measured inner diameter less than the outside diameter of the core pellets may be attributable to measurement error or core pellet shrinkage. In any case, the core pellets were difficult to remove after sintering for pellet 8 implying that there was contact. Finally, pellets 1, 2, and 6 all had minimum inner diameter ranges that included the outside diameter range of the core pellets. None of the measured inner diameter for these pellets suggests contact with the core pellets. For pellet 1, one of the core pellets required moderate force for removal. Perhaps there was contact in this case. For pellets 2 and 6, however, the core pellets were removed easily. It is difficult to determine if there was any significant contact with the core pellets in these cases.

One interesting observation noted below Table 1 is that for pellets 7 and 8, the gauge plugs representing the upper limit of their respective minimum inner diameter ranges could be inserted so that the end of the gauge plug was flush with the opposite end of the pellet. However, the gauge plugs could not pass though the pellets. Therefore, the pellet inner diameters must be at least the same size as the gauge plug outer diameters or perhaps there was a kink in the inner channels of the pellets preventing the gauge plugs from passing through. A possible cause for this is that the axes of the core pellets were not aligned during sintering. This could be rectified in subsequent experiments if only a single, long core pellet is inserted in the annular pellets during sintering.

In Figure 8, the axial inner diameter profiles for each pellet in Test #2 are plotted. The average inner diameters vary from 8.537 to 8.585 mm. The standard deviations for the data are also shown and range from 14 to 34 microns. The average and standard deviation data indicate that there was significant dimensional variation between pellets as well as within individual pellets. As can be seen, there are no distinguishing features between the profiles of those pellets sintered with core pellets and those that were sintered without. The profiles indicate an hour glass shape for many of the pellets. This is typical of pellets that have been double-ended pressed and is caused by the green density variations in the pellet. As can be seen from Figure 6, pellets pressed to lower green densities shrink more than those pressed to higher green densities. Similarly, within the pellet, the middle of the pellet tends to shrink more because it has a lower green density than the ends. It is clear from the data presented in Figure 8 that the non-uniformity in the green pellet densities results in an appreciable spread in the measured sintered pellet inner diameters. The data also suggests that the pellet ends are less likely to be in contact with the core pellets than the middle. The green density variation within the annular pellet must be minimized if it is to shrink uniformly and contact the core pellets across its entire length.

To summarize the results from Test #2, the gauge plug experiments and other supporting evidence suggested that some of the annular pellets had shrunk down to the core pellets. However, the measured inner diameters from Figures 7 and 8 indicated that the annular pellets did not contact the core pellets along their entire length during sintering. Therefore, in this test, while some of the annular pellets appear to have contacted the core pellets during sintering the influence of the core pellets in controlling the inner diameters of the annular pellets was limited.

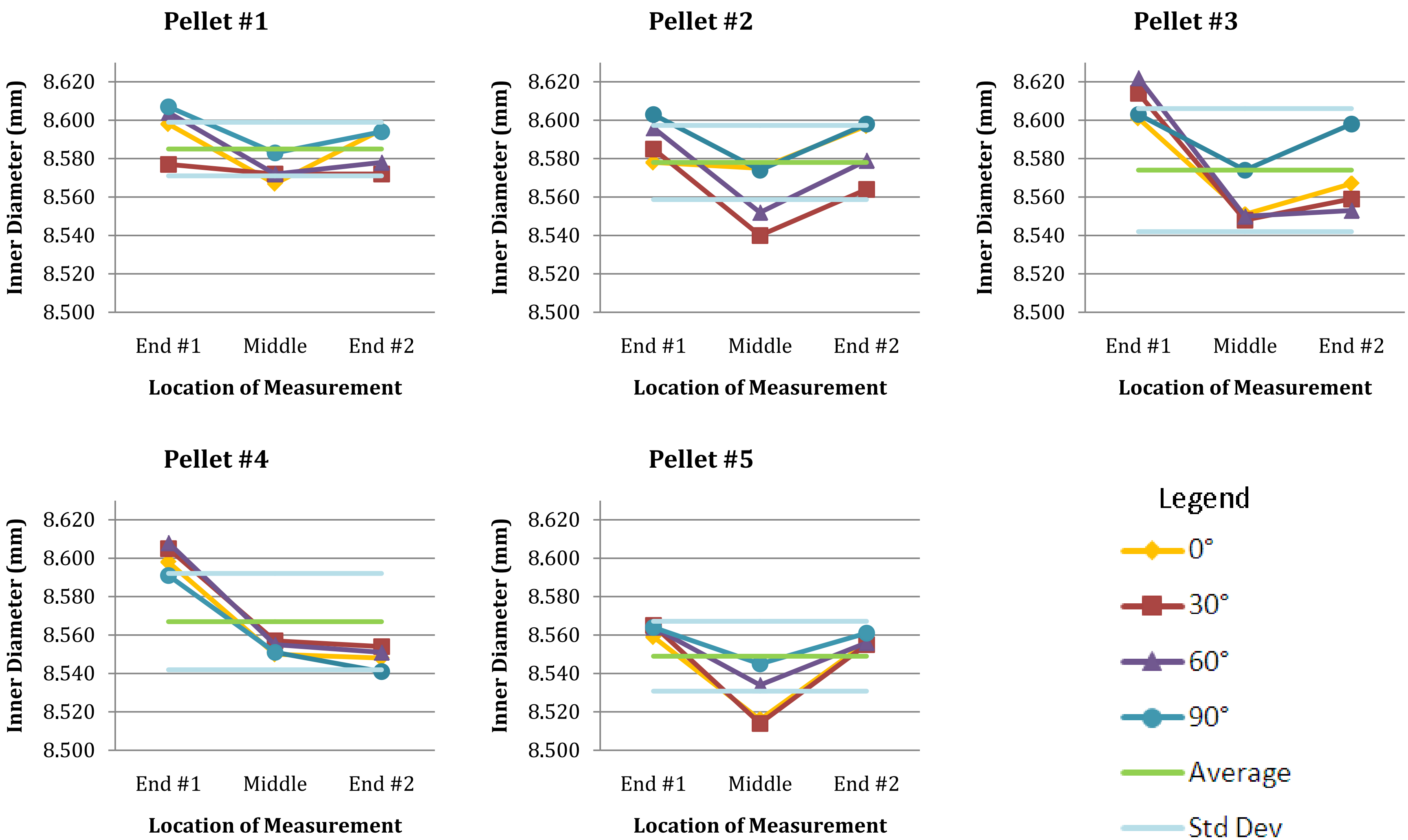

Figure 8: Test#2 Measured Inner Diameters

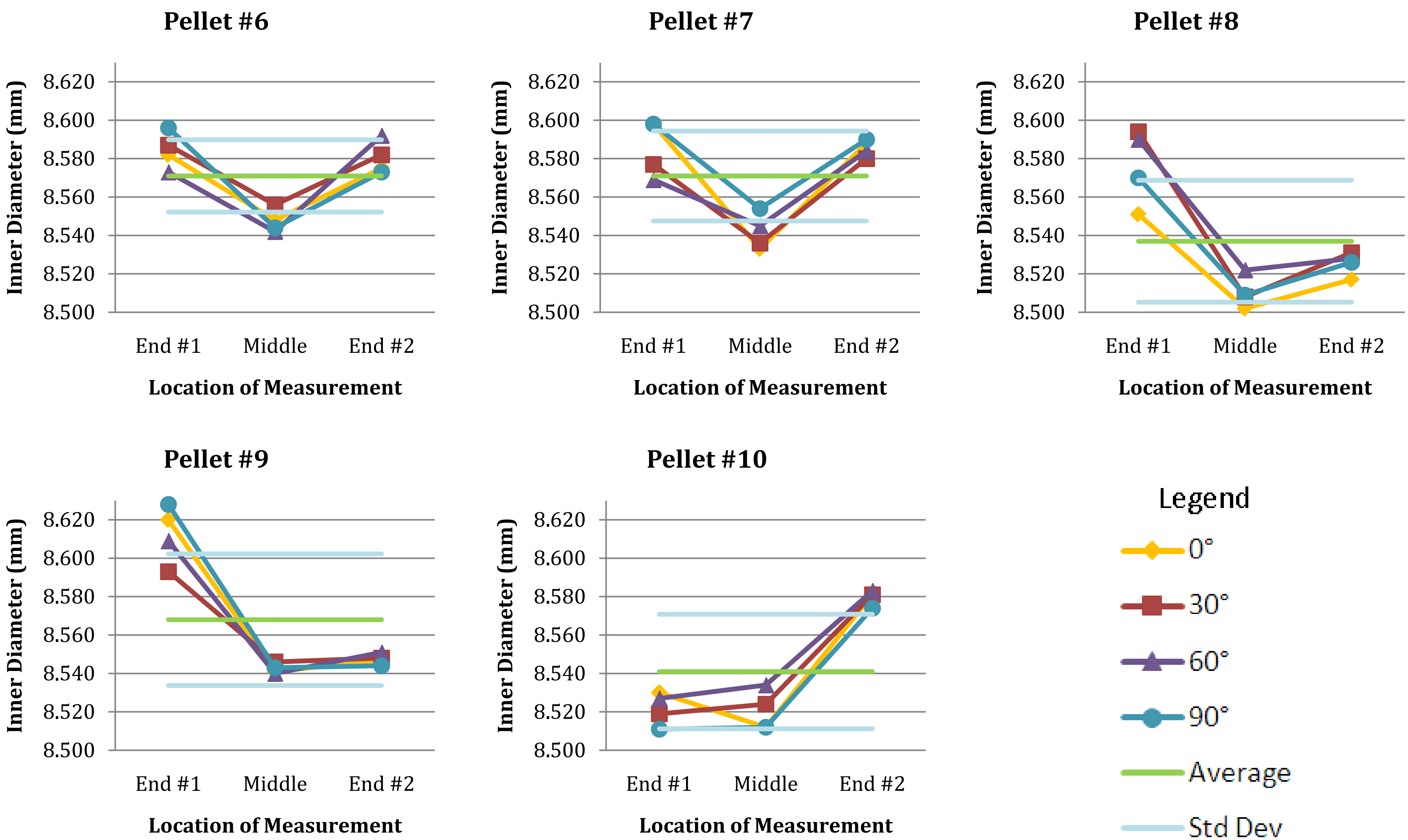

Figure 8 (cont.): Test#2 Measured Inner Diameters

The method of using precisely machined UO_2 core pellets to control annular pellet inner diameter is only effective if the annular pellet shrinks uniformly down to the core pellet during sintering. The uniformity of shrinkage is affected by the green density gradient in the annular pellet. Therefore, an understanding of the green density variation in the annular pellets is important. In Test #3, an annular pellet was pressed using the same pressing parameters as in Test #2. The pellet was then mounted in resin and sectioned into four pieces. The pellet slices were released from the mount material using acetone and their green density was measured. Inspection data from the as-pressed pellet and the pellet slices is provided in Table 2.

Table 2: Inspection Data for Sliced Green Pellet in Test #3

Sample	Weight (g)	Outer diameter (mm)	Inner diameter (mm)	Length (mm)	Green Density (g/cm^3)
Whole Pellet	17.923	18.64	10.64	18.38	5.3
Slice #1 (top)	5.520	18.78	10.65	5.78	5.1
Slice #2	3.873	18.78	10.68	3.96	5.2
Slice #3	3.369	18.7	10.73	3.81	4.8
Slice #4 (bottom)	3.600	18.7	10.71	3.83	5.1

As can be seen in Table 2, the green densities measured for the slices were less than that measured for the whole pellet. This is believed to be due to volumetric swelling of the pellet slices while they were being released from the mounting resin. The outer and inner diameters of the slices are larger than those of the whole pellet which supports this theory. Therefore, the inspection data cannot provide direct insight into the actual green density gradient in the as-pressed pellet; however, it does provide an indication of the relative density differences. Using the information in Table 2 and Equation 1, and assuming a sintered density of 10.70 g/cm^3, the sintered pellet inner diameter range should be on the order of 0.18 mm (8.45 mm-8.27 mm= 0.18 mm). The actual measured inner diameter range for the sintered pellet slices was 0.11 mm (8.53 mm -8.42 mm= 0.11mm). While the actual spread in the data was smaller than that predicted, it is clear that the green density variation within the annular pellets must be narrowed to achieve more uniform shrinkage in the pellet during sintering.

One method of reducing the green density variation of a pellet is to reduce the length to diameter ratio [6]. All the pellets that have been described thus far have had a length to diameter ratio of approximately 1. In subsequent pressing trials, pressing pellets with smaller length to diameter ratios will be considered. Also, the green density gradients in a pellet are a result of powder to powder, and powder to die wall friction during pressing. If additional lubricant is used during pressing, the frictional forces can be reduced. A combination of pressing annular pellets with smaller length to diameter ratios and pressing with more lubricant could yield more uniform shrinkage during sintering. If these additional steps are taken during pressing and a precisely machined UO_2 core rod is inserted in the annular pellet during sintering, better control over the inner diameter of the annular pellet may be achieved.

3. Conclusions

In this study, precisely machined UO_2 core pellets were used to restrict annular pellet shrinkage during sintering in an attempt to yield a precise and accurate inner diameter. However, it was determined that during sintering, the annular pellets did not shrink down to the core pellets along their entire lengths. The measured inner diameters for the sintered annular pellets had an appreciable spread and the core pellets did not have their intended effect. The non-uniform shrinkage observed after sintering was attributed to green pellet density gradients present from pressing. To minimize the green density variation in the annular pellets in future trials, it is recommended that pellets with smaller length to diameter ratios be pressed and that the amount of lubricant used during pressing be increased.

4. Acknowledgements

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5. References

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