Journal of Nuclear Materials 406 (2010) 205–211

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

# Dry bag isostatic pressing for improved green strength of surrogate nuclear fuel pellets

G.W. Egeland *, L.D. Zuck, W.R. Cannon, P.A. Lessing, P.G. Medvedev

*Idaho National Laboratory, Idaho Falls, ID 83415, United States*

ARTICLE INFO

*Article history:*
Received 17 December 2009
Accepted 21 August 2010

ABSTRACT

Dry bag isostatic pressing is proposed for mass production of nuclear fuel pellets. Dry bag isostatically pressed rods of a fuel surrogate (95% $CeO_2$–5% $HfO_2$) 200 mm long by 8 mm diameter were cut into pellets using a wire saw. Four different binders and $CeO_2$ powder obtained from two different sources were investigated. The strength of the isostatically pressed pellets for all binder systems measured by diametral compression was about 50% higher than pellets produced by uniaxial dry pressing at the same pressure. It was proposed that the less uniform density of uniaxially pressed pellets accounted for the lower strength. The strength of pellets containing $CeO_2$ powder with significantly higher moisture content was five times higher than pellets containing $CeO_2$ powder with a low moisture content even though they were 25% less dense. Capillary pressure of the moisture was thought to supply the added binding strength.

Published by Elsevier B.V.

## 1. Introduction

Nuclear fuel pellets are typically fabricated using a rotary press with multiple tool stations by uniaxially pressing in double action dies at the rate of several hundred pellets per minute [1]. A number of problems, however, are associated with uniaxial die pressing that may lead to pellet rejection. These are: (1) cracking due to end capping and/or laminations, (2) density variation within the pellet, and (3) die wear leading to lack of dimensional control [2]. In addition, as-sintered pellets have a slightly hour-glass shape due to non-uniform packing of the powder compact and therefore require post-sinter machining. In this paper we consider as an alternate route for fabrication of green pellets using dry bag isostatic pressing of fuel rods and subsequent cutting of the above rods into pellets [3].

Dry bag isostatic pressing (DBIP or "isopressing") is used for mass production of ceramic parts, for instance, spark plugs, automobile oxygen sensors and ceramic rods and tubes. It is a method to mold powder into a green part by introducing granulated powder into a polyurethane mold and hydrostatically pressing it through a master bag within the high-pressure vessel. It distinguishes itself from "wet bag isostatic-pressing" by the fact that the mold does not contact the hydraulic fluid directly but through the master bag and so can be rapidly filled, inserted, pressed and taken out again. This method is suitable for mass production of simple shaped products with its labor-saving automatic operation. Pressing rates can be as high as one hundred parts per minute but for best results only a few parts per minute should be pressed. When pressing rates are too high, dimensional control suffers which is also true for uniaxial pressing. For the manufacturing of nuclear fuel pellet, a single rod may be cut into 20–30 pellets depending upon the pellet length requirement. DBIP can be automated all the way from powder filling the mold to product removal. The main advantage of using isostatic pressing over uniaxial pressing for nuclear fuel production is that much more uniform pellet densities are achievable [4]. Rods can be formed with a very uniform diameter except for the end which flares out, often termed "elephant foot" caused by the lack of contraction of the compact adjacent to the metal punch that seals the bag [5] but this can be reduced by special bag construction [6].

Although isopressing of single fuel pellets appears to be a very favorable manufacturing method, there may be an advantage to cutting pellets from a long rod. The pressing rate can be slowed down. The pressure release rate after pressing is most important in controlling uniform powder packing and dimensional control [7]. It may not be too difficult to chamfer and dish a pellet cut from a long rod. This would require post-press machining, but individual dry bag isopressed spark plugs and zirconia oxygen sensors are rapidly, robotically machined after pressing [2]. Independent of whether pellets are pressed one at a time or are cut from a longer rod, a further advantage is that the aspect ratio of the pellets is not limited to short lengths.

Importantly, the mechanical properties of the pellets are likely to be improved by DBIP, resulting in less rejection of green pellets due to chipping [8], end capping, and lamination cracking, and therefore, more robust resistance to handling damage. It is not known, however, whether pellets can be cut without degrading

* Corresponding author. Tel.: +1 208 355 7431; fax: +1 208 533 7863.
*E-mail address:* gerald.egeland@inl.gov (G.W. Egeland).

0022-3115/$ - see front matter Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2010.08.022

the strength of the rod. This paper compares the green strength of dry bag isopressed and cut pellets with uniaxial die pressed pellets.

In the present paper a wire saw is used to cut the green, unsintered rods into pellets for fracture testing because wire saws are known to cause minimal damage. Surface damage occurring from machining of sintered ceramics is almost always the source of the critical fracture flaw. Machining flaws, for instance, dominate the source of failure for flexure testing [9]. This is partly due to the fact that the maximum stress in a flexure test is at the surface of the specimen, but it is also due to the unavoidable damage from machining of the test specimens and the fact that the stress intensity factor of a surface flaw is higher than that of an interior flaw of the same size [10]. Improving the surface finish is not known to affect the strength of green parts and so this study also tests the affect of the polishing the cut surface of the green pellet.

Diametral compression (sometimes called the indirect tensile test or Brazilian test) is used to measure green pellet strength. The calculated elastic stress state is biaxial with compressive stresses along the loading axis (vertical) and tensile stresses along the horizontal axis as shown in Fig. 1. Failure occurs by a crack propagating down the vertical axis (between loading points). The tensile stress at failure is determined from the maximum elastic stress

$$\sigma_f = \frac{2P_{\max}}{\pi D t} \tag{1}$$

where $P_{\max}$ is the load at failure, $D$ and $t$ are the disk diameter and disk thickness, respectively. Unlike the horizontal tensile stress, the elastic compressive stress state down the vertical diameter varies with the distance from the center and becomes very large near the contact point. It is given by

$$\sigma_y = -\frac{2P}{\pi t}\left(\frac{1}{D} + \frac{1}{(\frac{D}{2} - y)}\right) \tag{2}$$

In contrast to the bend tests the maximum tensile stress is not at the surface but is the same through the thickness of the pellet.

It has been widely shown in the literature that the diametral compression test of both green ceramics and sintered ceramics yields lower mean fracture strength values by almost a factor of two than bend tests. However, in comparing different types of samples the same pattern of relative values of strengths are seen as in the bending tests [11,12] so that it is a valid method of comparing mechanical strength of pellets prepared under different conditions and avoids the complexity of preparing bend bars.

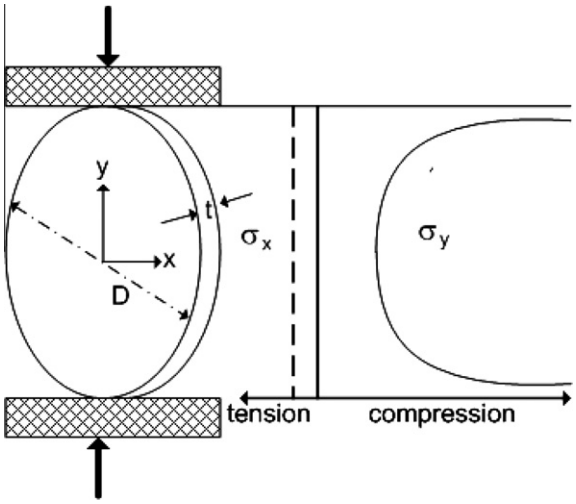


**Fig. 1.** Elastic solution for stress distribution in diametral compression.

In this paper $CeO_2$ and $HfO_2$ are used as non-radioactive surrogates to represent MOX powder mixtures. The purpose of the paper is to determine whether the strength of pellets are comparable or better when pellets are cut from rods fabricated by isopressing than by uniaxial pressing of individual pellets.

## 2. Experimental procedure

### 2.1. Powder preparation and processing

Cerium oxide powder from two different sources, namely TJTM and IAM, were used in this study. Details of the powder origin and properties are shown in Table 1. Cerium oxide was mixed with hafnium oxide powder to have a composition of $CeO_2$–5 wt.% $HfO_2$. (See Table 1 for hafnium oxide properties.) Table 2 lists four binders used in the study. The additives and processing techniques to prepare samples sets for die pressing and isostatic pressing were as follows: (A) 0.2 wt.%[1] Carbowax 8000 and 0.2 wt.% zinc stearate were mixed in a Turbula™ mixer with six large alumina balls for 1 h, (B) Same as A except the binder was 0.5 wt.% POLYOX instead of Carbowax, (C) 0.2 wt.% oleic acid was mixed in a Turbula™ for 10 min and then mixed in a Waring Blender for 90 s at the highest RPM setting while an aqueous solution of Mobilcer Q was added through RAZEL syringe pump. The total Mobilcer Q was 0.5 wt.% based on ceramic powder. (D) 0.2 wt.% zinc stearate was mixed with the ceramic powder in a Turbula™ mixer for 10 min. The powder was then removed from the Turbula™ and mixed in a Waring Blender for 90 s at the highest RPM setting while 1.0 wt.% QPAC (90 parts QPAC - 10 parts propylene carbonate) was added through RAZEL syringe pump. (E) was the same as sample set A except that IAM powder replaced the TJTM powder.

The moisture content of TJTM and IAM $CeO_2$ powder after storage in the laboratory for many months was measured with a Sartorius M100 moisture analyzer. The moisture content of TJTM and IAM powder were 0.22 and 2.34 wt.%, respectively. The difference was presumably due to the much higher specific surface area of the IAM powder since their respective moisture contents are proportional to the specific surface area.

### 2.2. Granule evaluation

Powder was granulated to improve the flowability by a roll compactor (Vector TFC Lab Micro, Marion, IA) and then passed through a 60 mesh screen. Sample set E powder containing IAM $CeO_2$ powder tended to stick to the auger and roller and, therefore, did not feed well into the roller. Hence, this powder was precompacted uniaxially at 70 MPa into discs and then granulated by passing through a 60 mesh screen. The granulated powders were evaluated for fill density, tap density and compaction ratio. Fill density is the density of the powder as it flows into the mold and is measured by pouring a measured weight of powder into a graduated cylinder. Tap density was measured accordance with ASTM standard B527-93 (Autotap, Quantachrome Instruments, Boynton Beach, FL) using 3000 taps in a 10 ml graduated cylinder. The compaction ratio is the pressed density divided by the fill density. The pressed density was measured geometrically on 12.7 mm diameter pellets pressed at 207 MPa.

### 2.3. Sample preparation

For uniaxial pressing granulated powder was pressed in a single action 12.7 cm diameter die (Carver, Wabash, IN) at 207 MPa to yield an aspect ratio of approximately 0.5 (diameter divided by

[1] All wt.% values for organic additives are relative to total ceramic powder.

**Table 1**
Ceramic powders used in this study.

| | $CeO_2$ | | $HfO_2$ |
|---|---|---|---|
| | Tianjiao international (TJTM) | Inframat advanced materials (IAM) | Wah Chang |
| Specific surface area, $m^2/g$ | 3.6 | 43.6 | – |
| Particle size, μm | 1.3 | $d_{50} < 1$ | 4 |
| Purity, % | 99.95 | 99.95 | 99.5+ |
| Type | 1017 | 58-R-0803 | S grade |

**Table 2**
Binders used in study.

| Trade name | Chemical/molecular wt. | Density ($g/cm^3$) |
|---|---|---|
| 1. Carbowax 8000 | Polyethylene glycol M.W. 8000 | 1.12 |
| 2. POLYOX WRS301 | Polyethylene oxide M.W. 4000,000 contains 3% fumed silica | 1.15 |
| 3. Mobilcer Q | Microcrystalline polyethylene parafin wax M.W. est. 1000–5000 | 0.93 |
| 4. QPAC | Poly(propylene) carbonate M.W. 100,000–150,000 | ~1.26 |

thickness). The diametral compression test standard, ASTM D3967-08 calls for an aspect ratio not to exceed 0.75. Pellets properties with an aspect ratio of 0.5 in a single action press are approximately equivalent to the top half of a pellet pressed in a double action press [13]. Thus the 0.5 aspect ratio for single action pressed pellet approximates the nuclear fuel pellets with an aspect ratio of 1.0 pressed in a double action die.

Isopressed samples were pressed from granulated powders at 207 MPa into a rod of about 200 mm in length and 8 mm in diameter. These were cut using a diamond wire saw (Wells, Norcross, GA) to also yield an aspect ratio of approximately 0.5. The cutting load was adjusted to cut through the sample in approximately one minute.

### 2.4. Dry strength

Fracture strength of green pellets was measured by diametral compression for both the uniaxial and isostatic pressed pellets. The tests followed ASTM standard D 3967-08. Eight to 10 pellets were tested from all sample sets. The load was released in a few seconds after failure and the samples were examined to determine whether the crack was along the vertical axis. None of the uniaxially pressed samples in sample set D fractured along the vertical axis but instead crushed at a low stress and so were excluded from the results.

### 2.5. Sintering

Sintering was performed in a Linderburg Blue M tube furnace with SiC elements rated to 1700 °C (1973 K). A computer using LabView software with a custom VI was used to interface and control the furnace temperature. The sintering schedule was set as a standard based on PEG burnout, and used a 1400 °C pre-sinter rest.

Ramp rates were 100 °C/h to 275 °C (548 K), then 275–325 °C (548–598 K) at 50 °C/h, then 325–1400 °C (598–1673 K) again at 100 °C/h. There was a 3 h soak at 1400 °C (1673 K), after which the temperature was raised to 1600 °C at 100 °C/h (1873 K). This sintering rest was held for 3 h, after which the furnace was cooled to ambient no faster than 200 °C/h.

**Table 3**
Granule properties.

| Sample set | Fill density | | Tap density | |
|---|---|---|---|---|
| | g/cc | % theo.* | g/cc | % theo.* |
| A | 1.7 | 23 | 2.8 | 38 |
| B | 1.5 | 20 | 2.6 | 35 |
| C | 1.6 | 23 | 2.7 | 37 |
| D | 1.6 | 22 | 2.6 | 36 |

* Assumes 7.36 $g/cm^3$ theoretical density.

## 3. Results

### 3.1. Processing

Rapid and uniform flow of granules into the dry bag cavity is important to insure uniform density of the isopressed rod. Table 3 contains results of the properties of the granule of the four sample sets described above. Fill density and tap density values are higher than typical literature values for $UO_2$ granules according to Wilson [1]. For instance, typical fill densities for $UO_2$ made by powders of Integrated Dry Route (IDR), ammonium diuranate (ADU) and ammonium uranyl carbonate (AUC) are 6%, 14% and 18–21% T.D., respectively. Their tap densities are 16%, 22–25% and 18–27% T.D., respectively. However, according to Reed [14] they are still lower than the desirable fill densities for dry pressing in a manufacturing environment and should be in the range 25–35% T.D. Nevertheless, the average compaction ratio (pressed density/pour density) is 2.6 which is within the limits for successfully isopressing.

Table 4 lists the green densities for both uniaxial and isostatic pressed pellets. In spite of the equivalent pressing pressures, the densities of the isostatic pressed pellets are found to be slightly higher. The difference is presumably due to die wall friction causing lower transmitted stresses to the bottom half of the compact and lower densities near the center of the pellet. The difference in densities is only approximately 1% but the non-uniformity of density inside the uniaxially pressed pellets is probably more significant, as will be explained below [7]. Most striking is the low green densities of sample set E containing IAM $CeO_2$. Low green densities are almost always observed for high specific surface area powders. The reason for this is that intragranular and intergranular pores are retained in high surface area powders even under pressures as high as 200 MPa [15].

Compaction curves such as the one shown in Fig. 2 were generated by pressing granulated powder in a 12.7 cm diameter die with a universal testing machine (Instron, Canton, MA). At very low stresses, granules rearrange to improve their packing. The break in the curve has generally been interpreted as the crushing or yield strength of the granules. The yield strength of all sample sets was on the order of 0.1 MPa, which is a very low value and much below the pressing pressure of 207 MPa. Thus granules are sufficiently soft that no remnants of the granules are expected to be left after pressing [16,17].

### 3.2. Strength measurements

Fracture strength results for uniaxial pressed and isopressed pellets are compared in Fig. 3. The mean fracture strength for the isopressed pellets is approximately 50% higher than that of the uniaxially pressed samples. The binder systems acted similarly for uniaxial pressed and isostatically pressed pellet strengths, however, no binder system greatly enhanced or decreased the strength except for uniaxially pellets of sample set D which failed by crushing. In all other cases a vertical crack appeared suddenly at the onset of the specimen's failure.

**Table 4**
Green and sintered pellet properties.

| Sample set | Avg. green density uniaxial pressed | | Avg. green density isostatic pressed | | Avg. sintered density uniaxial pressed | | Avg. sintered density isostatic pressed | |
|---|---|---|---|---|---|---|---|---|
| | g/cc | % theo.* | g/cc | % theo.* | g/cc | % theo.* | g/cc | % theo.* |
| A | 4.00 | 54 | 4.05 | 55 | 7.08 | 96 | 7.06 | 96 |
| B | 4.05 | 55 | 4.11 | 56 | 7.00 | 95 | 7.02 | 95 |
| C | 3.91 | 53 | 4.14 | 56 | 7.01 | 95 | 7.00 | 95 |
| D | 4.03 | 55 | 4.13 | 56 | – | – | 7.09 | 96 |
| E | 3.00 | 41 | 3.01 | 41 | 6.58 | 90 | 6.58 | 90 |

* Assumes 7.36 g/cm$^3$ theoretical density.

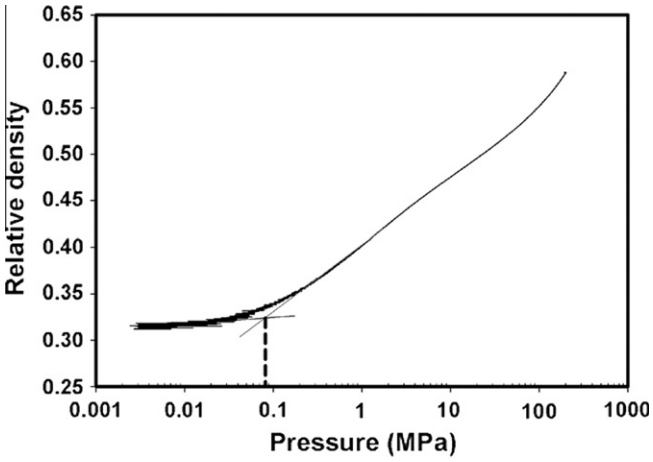


**Fig. 2.** Compaction curve for sample set A granules pressed in a 12.7 cm die by a universal testing machine. Density was calculated from the crosshead displacement and powder mass.

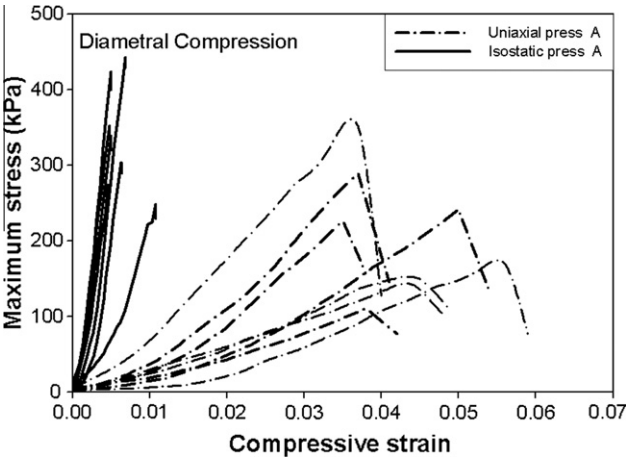


**Fig. 4.** Maximum stress vs. strain curves for sample set A. Maximum stress was calculated from Eq. (1) and compressive strain was determined by dividing the cross head deflection by the pellet diameter.

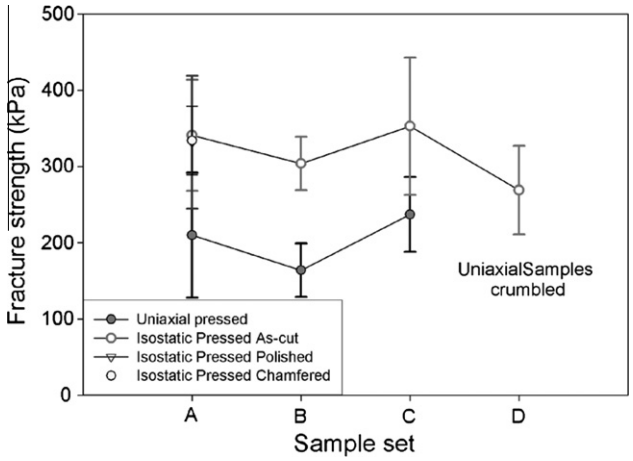


**Fig. 3.** Comparison between fracture strength of uniaxially pressed pellets and dry bag isostatically pressed pellets for four sample sets.

Fig. 4 compares the stress strain curves of uniaxial and isostatic pellets. The stress is the maximum tensile stress given by Eq. (1) and the strain is the total compressive strain of the specimen. The strain axis was plotted this way to compare compressive strains of pellets with different diameters. It is clear that the compressive strains of the uniaxially pressed pellets are much larger although they reach a smaller maximum tensile stress. It was also observed that weaker specimens did not exhibit sudden brittle failure but a slight amount of ductile failure.

Although the aspect ratios of the pellets averaged around 0.5, they varied from sample to sample and were generally larger for isostatically pressed samples. Fig. 5 compares the aspect ratio vs. strength for all specimens. There is no significant affect of strength on aspect ratio.

Diametral compression strength of uniaxially pressed green pellets are often higher than the current results, usually on the order of 300–500 kPa depending on the amount of binder used. Here slightly lower values are seen probably because of the lower binder content. However, strengths of uniaxially pressed pellets of set A containing 0.2 wt.% (1.3 vol.%) carbowax are higher than literature diametral strengths for green alumina pellets uniaxially pressed (90 MPa) containing 0.7 wt.% (2.4 vol.%) carbowax (40 kPa) [18] .

The mean fracture strength of isostatically pressed specimens (set E), containing the IAM $CeO_2$, were almost five times stronger than those containing the TJTM $CeO_2$. For isostatically pressed pellets of sample set E the mean fracture strength was 1533 ± 337 kPa. This is a particularly high strength considering the small binder content and very low green density. The most plausible explanation is the increased strength resulted from high capillary pressure due to the higher specific surface area in the IAM $CeO_2$ [19]. This subject will be investigated further in a future publication.

After cutting pellets from isostatically pressed rods, periodic surface striations from successive wire-saw strokes were apparent (see Fig. 6a). To determine the affect of surface damage from the wire saw cutting, ten samples of sample set A were polished with 1200 grit SiC paper by hand (see Fig. 6b). As can be seen in Fig. 3 the mean strength and error bar width of the ten polished specimens of sample set A was essentially unchanged (see Table 5). Chipping along the pellet edge was widely observed in sample sets

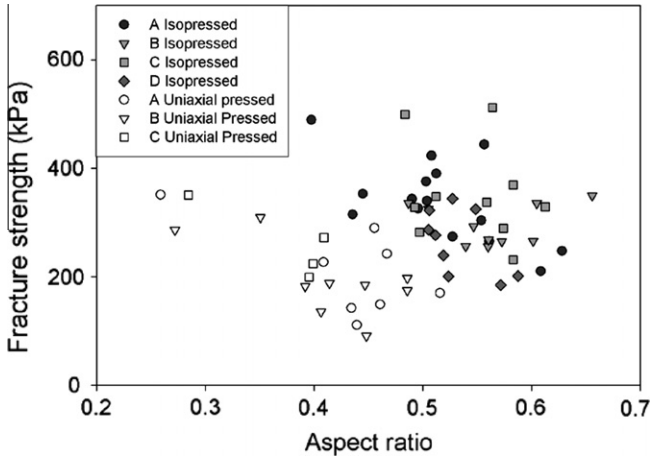


**Fig. 5.** Fracture strength of all specimens vs. their aspect ratio.

A, B, C, and D but not E (see Fig. 6c). To determine whether the chipping affected the fracture strength, edges near the vertical axis of the specimen were chamfered by lightly polishing the edge with 1200 mesh SiC paper. The mean strength also was not affected though several of the load vs. deflection curves contained a jog which probably indicated that the contact point collapsed prematurely. Nevertheless, the specimen did not fail until the higher stress was reached.

Fracture strength values were shown to follow a Weibull distribution. Other authors [20,21] have observed similar results when measuring fracture strength of green ceramics. The Weibull modulus was determined for all specimens by taking the least squares slope of $\ln\left(\ln\left(\frac{1}{(1-P_f)}\right)\right)$ vs. $\ln\sigma_f$ (Table 5), where $P_f$ is the probability of failure. The Weibull modulus was higher for isopressed pellets, indicating a narrower strength distribution. Although there was no improvement in strength by chamfering, the Weibull modulus did improve. It is also interesting that sample set B had a higher Weibull modulus. No explanation is given.

Sintered densities are listed in Table 3. As was the case with green densities, there was little difference between the sintered densities of the pellets containing different binders. Although

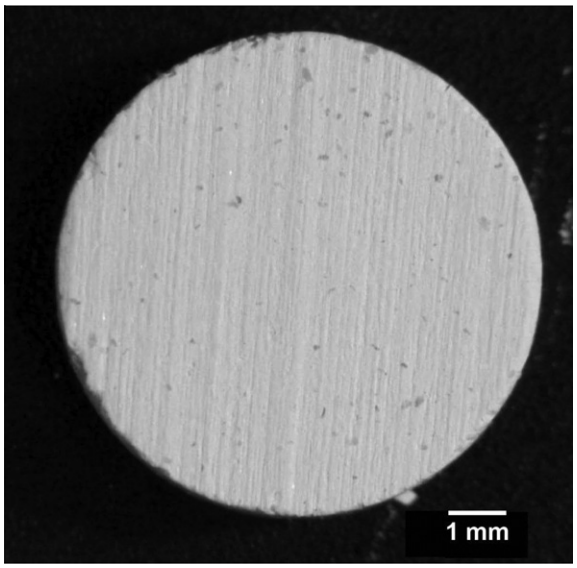


**Fig. 6.** Surface of sample set A pressed by dry bag isostatic pressing. (a) As-cut showing striations from successive strokes.

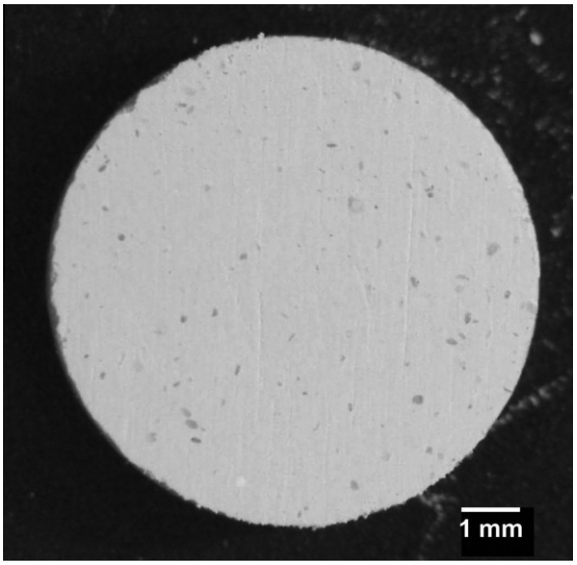


**Fig. 6b.** Polished with 1200 grit SiC paper.

green densities of isopressed pellets was approximately 1% higher than those of uniaxially pressed pellets, the sintered densities did not differ between isopressed and uniaxially pressed pellets. It is commonly observed that sintered densities of specimens with higher green densities are higher but the difference narrows during sintering. Since the difference was only one percent in the green state, it is not unreasonable that the differences in their sintered densities should be insignificant. It is also noted that difference between green and sintered densities in samples set E narrowed. The higher specific surface area of sample set E leads to a greater driving force but this is not always translated into a higher final density [15]. Sample set E did undergo a greater percentage shrinkage but it started at a much lower density. The change in relative density of sample set E was 49%, whereas the change in relative density of other sample sets was approximately 40%. Although there appears to be only a slight improvement in sintered density by isostatic pressing, isostatic pressed pellets contained no observable cracks whereas the uniaxial pressed specimens had many cracks after sintering. These cracks likely arose from the density differentials within the specimen which upon sintering translated into cracking due to differential shrinkage.

## 4. Discussion of results

One possible cause of lower failure strengths in uniaxially pressed pellets might be the presence of end capped cracks. End capped cracks develop as the pressure is released on the punch or as the pellet begins to be ejected from the die and as the compressed powder compact rebounds [2]. These cracks are thought to initiate at the edges of the pellet at approximately 80° to the die wall [22]. Since these cracks run almost parallel to the face of the pellet, the maximum tensile stress in a diametral compression test does not act across them, therefore they are a less likely source of a critical flaw. Several pellets failed from end cap cracks prior to testing. One specimen failed from a crack around the periphery of the pellet and may have initiated from an end cap crack but otherwise pellet fracture was not believed to be initiated from end caps. Thus although it is an important source of failure in practice, diametral fracture tests did not capture this source of failures.

Another source of cracks is ring cracks formed around incompletely crushed granules, however, the high pressures used in this study were thought to eliminate these defects. Internal lubricants

**Table 5**
Strength/Weibull modulus for diametral compression strengths.

| Forming method | Mean fracture strength (kPa)/Weibull modulus | | | | | | |
|---|---|---|---|---|---|---|---|
| | A | | | B | C | D | E |
| Uniaxially pressed | 210/3.0 | | | 164/4.5 | 261/4.5 | – | Not measured |
| | As-cut | Polished | Chamfered | | | | |
| Isostatically pressed | 341/5.5 | 332/4.7 | 334/8.3 | 304/9.2 | 353/4.7 | 269/5.1 | 1553/5.0 |

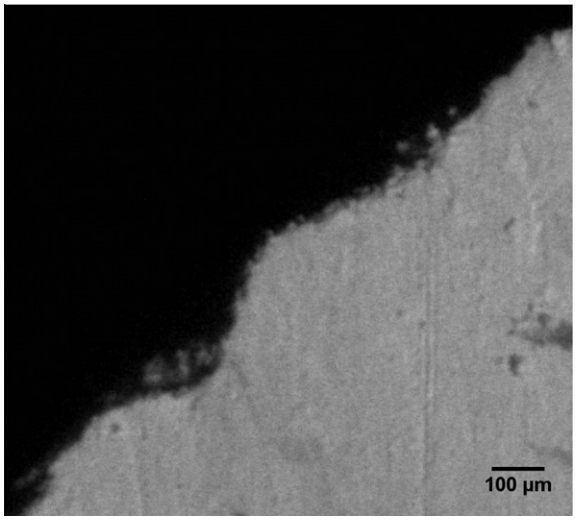


**Fig. 6c.** Higher magnification of typical edge chip.

(zinc stearate) also reduce the incidence of these cracks [17]. Shinohara et al. [16] used special optical techniques to examine green compact fabricated from spray dried powder and found intergranular pores and cracks as a source of flaws. This type of flaw may be important, but edge chipping flaws are much larger and so it is unlikely that intergranular pores or cracks are the source of failure flaws.

Edge chipping did not account for the lower strength of uniaxial pressed specimens since edge chips were similar size in the isostatically pressed pellets. It is somewhat surprising that edge chips, which are of the order of a hundred microns in diameter (see Fig. 5c), did not affect the strength. The flaws leading to fracture in this study have not been identified, but because these specimens were tested using the diametrical compression, the fracture origin may not be in a pre-existing flaw but may have their origin in shear flow in the powder compact prior to failure.

One may visualize a process of localized shear bands opening-up of extra large pores or flaws when the shear flow from opposite directions meet or differential shear within the pellet open up flaws. The concept is similar to ductile failure of metals. There is evidence in Fig. 4 that shear is important because pellets exhibited strain behavior which does not appear to be elastic. For instance, we observed occasional jogs and bumps in the curves. Furthermore, strain to failure greatly exceeds that typical for a brittle material. In considering the shear, the maximum shear strain at 45° to the principal strain which is in the vertical direction is given by

$$\varepsilon_{xy} = \frac{1}{2}(\varepsilon_y - \varepsilon_x) \tag{3}$$

where $\varepsilon_y$ is the principal strain (axial compressive strain) and $\varepsilon_x$ is the principal tensile strain (horizontal strain in the plane of the pellet face). Along the compressive stress axis powder rearranges by shear to a higher packing density and in the tensile direction to a lower packing density. Since total axial compressive strains in isostatically pressed pellets range from $\varepsilon_y = -0.005$ to $-0.01$ (Fig. 4), then $\varepsilon_{xy} < -0.005$ to $< -0.01$ which are very large shear strains. Even much larger is the shear strain for uniaxially pressed pellet. It is as large as 0.06. It is proposed that the bonding strength of uniaxially pressed pellets in sample set D were insufficient to hold the pellet together long enough for a brittle crack to propagate along the path of maximum tensile stress.

Propagation of cracks in mode II (by shear) is very unfavorable in brittle ceramics. Observation of vertical cracks tell us that failure in all cases is mode I (tensile) failure. However, in uniaxially pressed pellets shear cracks did propagate near the contact where the shear stresses are highest as shown in Fig. 7 but did not become the critical flaw.

Since isopressed specimens have a slightly higher density and if fracture has its origin in shear, there might be a correlation between green density and strength. However, in comparing, green density and strength (Tables 4 and 5) between uniaxially pressed pellets and isopressed pellets there is, if anything, an inverse relation between density and strength. Considering the above results, the lower fracture strengths of uniaxially pressed pellets are concluded to be related to their non-uniform density. It is die wall friction during uniaxial die pressing that leads to non-uniform density of pellets, particularly when the pressing pressure is >100 MPa. Yanai et al. [7] used EDX to measure the local green densities distribution of pellets pressed in a double action die at 230 MPa from three $UO_2$ powder sources, IDR, ADU and AUC. They found between one to five percent higher densities at the edge of the $UO_2$ green discs than the average density. Isostatically pressed pellets from the same powder were very homogeneous. Briscoe et al. [23] studied uniaxial die pressing of alumina powder and found an even greater differences in densities across the pellets by using a colored

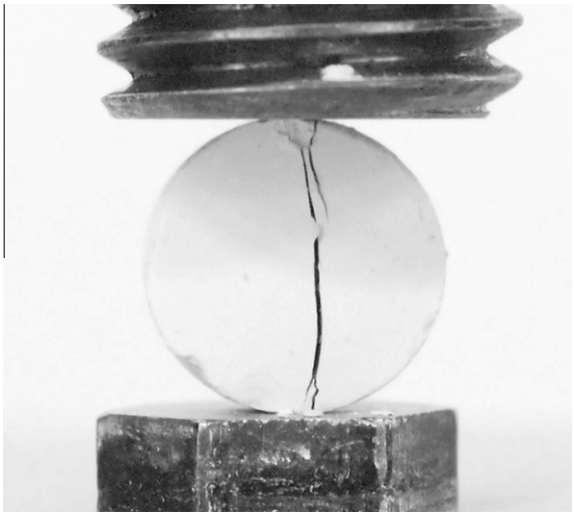

**Fig. 7.** Fracture pattern for pellet of sample set A uniaxially pressed showing both a nonpropagating contact shear crack and the mode I failure crack.

layer technique to measure density differences. Using a double action die at a forming pressure of 148 MPa with and without externally lubrication of zinc stearate, the difference from edge to center was 11.9% and 20.6%, respectively. They proposed that the degree of density non-uniformity depended on how easily powders rearrange under pressure [23]. So it may not be the average density of the pellet that is important but the density at the center of the pellet where fracture initiates.

It is proposed that the lower strength of uniaxial pressed pellets resulted from lower density regions of the pellet which ultimately initiate failure along the vertical plane down the center of the specimen which is the plane of maximum tensile stress. The resistance to failure depends strongly on the binding force between particles. For instance, sample set E contained strong binding forces presumably from capillary forces of moisture and therefore, had higher fracture strength. In addition to the source of failure differential densities are necessarily accompanied by residual stresses. These stresses have not been measured in green ceramic compacts but they can be very large in pressed metal compacts [24].

Besides exhibiting lower fracture strengths under diametral compression, several uniaxially pressed pellets from each sample set failed in handling. A few of these failed from laminating cracks or end cracks and others were merely more fragile. These fractures may have had their origin from differential densities and residual stresses. Many of these problems may be ameliorated by adjusting the binder, external lubrication, dwell time under pressure, etc., but DBIP appears to offer a more robust manufacturing method.

Although this study did not indicate that diametral compression strength was affected by edge chipping, this defect could still be a problem because of loss of material [8]. Reed [14] states that diametral fracture strengths must exceed 500 kPa to avoid edge chipping. Results of this paper supported the fact that chipping did not occur in sample set E pellets which were the only pellet having strength in excess of 500 kPa. Both binder type and increased binder content are known to yield pellets much stronger than 500 kPa [19], however, in this study powder characteristics which affected the moisture content rather than the use of various binder showed the greatest affect. This approach to strengthening green pellets may be very beneficial in nuclear fuel fabrication since it may lead to avoidance of binder additions altogether.

## 5. Conclusions

The results of this work show that pellets can be fabricated from isopressed rods by cutting them with a wire saw. The fracture strength as measured by the diametral compression test is much better than that for uniaxially pressed pellets. Although the wire saw may produce flaws on the surface and chips on the edges, our efforts to remove these flaws did not affect the fracture strength. The origin of failure is difficult to determine in a green ceramic but it is concluded that a large amount of shear strain occurred prior to failure. It is proposed that the critical flaw developed during this deformation since the fracture strength was not sensitive to large visible flaws. It is further proposed that variability of density within the uniaxially pressed samples lead to the lower fracture strength.

In this study four different binder systems were used. All binder contents were lower than typically used in ceramic manufacturing but there was little difference in strength between the binder systems. A much greater affect was the source of powder. It was believed that the large difference in capillary pressure due to moisture on the two powders used here accounted for the difference in the fracture strength. This difference is likely due to a higher capillary pressure in the pellet with a high moisture content.

## Acknowledgements

We would like to thank DOE for funding this project under LDRD NU101. We also would like to thank Randy Lloyd of Idaho National Lab for invaluable assistance with mechanical testing. The authors also wish to thank Kenneth J. Morris of Isoform Ltd., Shropshire, TF9 1QS, United Kingdom for dry bag isostatic pressing of rods. STI Number: INL/JOU-09-17331.

## References

[1] P.D. Wilson, The Nuclear Fuel Cycle: From Ore to Waste, Oxford University Press, 1996.
[2] D.W. Richerson, Modern Ceramic Engineering: Properties, Processing and Use in Design, third ed., Marcel Dekker, New York, 2006.
[3] P.A. Lessing, Method for forming nuclear fuel pellets and nuclear fuel pellet structures formed by such methods, Patent Application, USA, 2009.
[4] R.M. Gill, J. Byrne, Application of isostatic pressing techniques to the production of dense ceramic bodies, in: Proceedings of the Fourth International Conference on Science of Ceramics, 23–27 April 1967, British Ceramic Society, Stoke-on-Trent, UK, 1968, pp. 91–104.
[5] M. Koizumi, M. Nishihara, Isostatic Pressing, Technology and Applications, Elsevier Sciences Publishers Ltd., 1991.
[6] B.P. Bewlay, D.J. Dalpe, Mold for isostatic pressing, in: General Electric Company (Ed.), Patent No. 5631,824, 1996.
[7] K. Yanai, S. Ishimoto, T. Kubo, K. Ito, T. Ishikawa, H. Hayashi, J. Nucl. Mater. 224 (1995) 79–84.
[8] S. Borell, H. Widegren, H. Shah, Improved Fuel Performance by Enhanced UO2 Fuel Fabrication and Inspection Methods, in: Europ. Nucl. Soc., 2006, pp. 100–105.
[9] R.W. Rice, Mechanical Properties of Ceramics and Composites: Grain and Particle Effects, Marcel Dekker, New York, 2000.
[10] J.B. Wachtman, W.R. Cannon, M.J. Matthewson, Mechanical Properties of Ceramics, second ed., Wiley & Sons, Inc., Hoboken, N.J., 2008.
[11] Y. Nayak, R.P. Rana, S.K. Pratihar, S. Bhattacharyya, J. Mater. Sci.: Mater. Med. 19 (2008) 2437–2444.
[12] J.L. Amoros, V. Cantavella, J.C. Jarque, C. Feliu, J. Eur. Ceram. Soc. 28 (2008) 701–710.
[13] R.A. Thompson, Am. Ceram. Soc. Bull. 60 (1981) 237–243.
[14] J.S. Reed, Introduction to Principles of Ceramic Processing, Wiley, New York, NY, 1988.
[15] Y. Jorand, M. Taha, J.M. Missiaen, L. Montanaro, J. Eur. Ceram. Soc. 15 (1995) 469–477.
[16] N. Shinohara, M. Okumiya, T. Hotta, K. Nakahira, M. Naito, K. Uematsu, J. Mater. Sci. 34 (1999) 4271–4277.
[17] S. Balasubramanian, D.J. Shanefield, D.E. Niesz, J. Am. Ceram. Soc 85 (2002) 134–138.
[18] S.K. Verma, M. Roubin, Polyethylene Glycol in Ceramic Powders Processing, in: Fourth International Conference on Ceramic Powder Processing Science, Nagoyua, Japan, 1991.
[19] S.A. Uhland, R.K. Holman, S. Morissette, M.J. Cima, E.M. Sachs, J. Am. Ceram. Soc. 84 (2001) 2809–2818.
[20] J.L. Amoros, V. Cantavella, J.C. Jarque, C. Feliu, J. Eur. Ceram. Soc. 28 (2008) 701–710.
[21] S. Baklouti, T. Chartier, J.F. Baumard, J. Am. Ceram. Soc. 80 (1997) 1992–1996.
[22] R.A. Thompson, Am. Ceram. Soc. Bull. 60 (1981) 244–247.
[23] B.J. Briscoe, S.L. Rough, Colloid Surf. A: Physicochem. Eng. Aspects 137 (1998) 103–116.
[24] S.T. Hong, Y. Hovanski, C.A. Lavender, K.S. Weil, J. Mater. Eng. Perform. 17 (2008) 382–386.