Ceramics International 29 (2003) 99–105

CERAMICS INTERNATIONAL

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Short communication

Light attrition of uranium dioxide powder

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Received 13 June 2001; received in revised form 11 February 2002; accepted 13 March 2002

Abstract

Sinterable uranium dioxide powders prepared through the ammonium diuranate route are prone to agglomeration causing difficulty in compaction and sintering. UO_2 powder after light (short duration and low ball to charge ratio) attrition has been found to require lower compaction pressures and resulted in higher yields after sintering and finish grinding. Details of an in-house-built attritor are given. Experimental results concerning the effects of attrition on powder characteristics, green density, green strength, sintered density and recovery are presented and explained on the basis of green strength theory. The process is especially suited for low or room temperature precipitated (and hence fine or loosely agglomerated) powders. Even though only uranium dioxide powders have been handled, the principles are applicable to other ceramic powders.

Keywords: A. Powders, Attrition; Uranium dioxide

1. Introduction

The word ‘attrition’ is used in two ways. Undesirable dust formation and size reduction when powdered materials are being handled or transported is called ‘attrition’ [1]. On the other hand, it may mean intentional comminution to bring about a reduction in particle size of a powder. It is in the latter sense ‘attrition’ is used in this paper. The attritor, patented in 1956 [2] is increasingly being used for comminution [3,4] and in mechanical alloying [5–9] by virtue of its several advantages, such as operational flexibility, high productivity, moderate energy consumption, small space requirements, ease of operation etc. Grinding rates more than 10 times higher than those typical of conventional mills [10], submicron sizes [11] and narrow particle size distribution [12] can be achieved. A two-stage attritor was developed to overcome powder sticking onto the inner wall [13]. The attritor has been particularly attractive in nuclear fuel fabrication being less cumbersome than ball/hammer or jet mill and amenable to containment, dust free operation and minimal waste [14]. The packing efficiency of ceramic UO_2 powders in the compaction die is limited by the presence of agglomerates leading to low density and spring-back defects [15,16]. Several steps are necessary in the preparation of fine powders with low agglomerate content [17,18]. In addition to these, light attrition of the powder, sufficient to achieve further deagglomeration is proposed in this work. Vucak et al. [19] found that crystal aggregation is the mechanism, which determines the overall particle size at lower temperature (30 °C), while crystal growth dominates at higher temperatures. Light attrition is sufficient to separate the loosely held particles constituting agglomerates formed from low temperature precipitation. On the other hand, crystal breakage requires a very much larger force than that required to separate loosely held particles in an agglomerate. Hence powders made from high temperature precipitated powders require long duration energy intensive heavy attrition along with precautions against powder contamination and mill wear. Heavy attrition is used elsewhere for particle size reduction (for example, graphite used for nuclear fuel sheath inner coating) [20].

In this paper, details of an attritor (for the purpose of deagglomeration of UO_2 powder) and a compressive strength test rig (for axial compression testing of compacts), both built in-house are given. Experimental results on light attrition and frequency distribution of compressive failure stresses on green pellets made from both the attrited and unattirted powders are presented. Weibull modulii are evaluated.

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PII: S0272-8842(02)00079-2

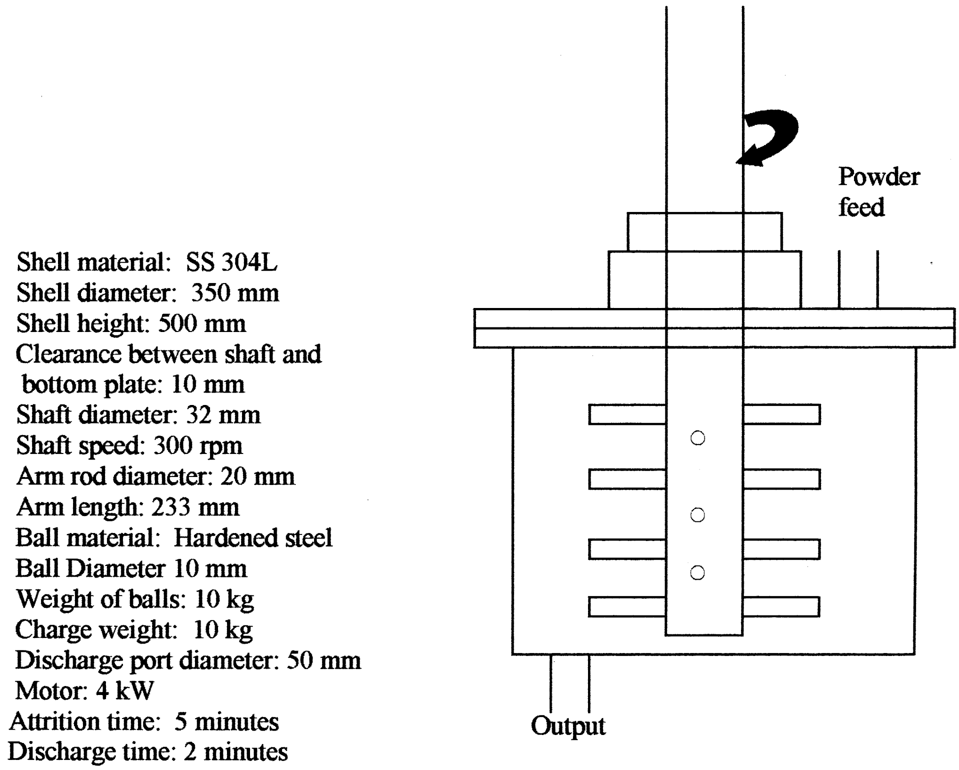

Fig. 1. Attritor (Schematic).

2. Experimental

Details of the attritor used in this work are given in Fig. 1. There is no water-cooling. The outlet of the attritor had rods welded with 6 mm wide gaps between them to allow free passage of powder and retention of the media. About 20 kg of UO_2 powder was homogenised in a mixing machine and called a "lot". Four such lots were prepared. Each lot was divided into two equal parts. One part, in as received state was precompacted (at 70 MPa), granulated (to −14 mesh) and final compacted (at 212 MPa) into 18 mm diameter cylindrical pellets. The other part was attrited for 5 min and then processed as before. Before pelletizing, attrited and unattrited powder samples were taken for determination of Specific Surface Area by BET method and Average Particle Size by Fisher Sub-sieve Sizer (Permeability) method. A known mass of the powder was poured into a measuring cylinder and the volume occupied by the powder was noted to get "pour density". The measuring cylinder was tapped gently to allow packing of the powder by particle rearrangement and the volume of the powder was noted again to get the "tap density". Particle size distributions were obtained using Horiba LA 500 Diffraction Particle Size Analyser. Forty five green (unfired) pellets each from attrited and unattrited powders were tested using the hydraulic compression test rig until failure and the load at the commencement of failure was noted. Some attrited and unattrited type green pellets were also sintered in cracked ammonia at 1700 °C in a pusher type continuous sintering furnace. The sintered pellets were centreless ground to achieve uniformity of diameter. Densities of the green pellets and sintered pellets from the attrited and unattrited categories were measured. The finished sintered pellets were inspected as per density, dimensional, integrity and microstructural in-house specifications. Recovery, as given by the number of pellets accepted and divided by the number of pellets inspected, was noted.

3. Results

Table 1 gives the BET specific surface area and Fisher's average particle size of attrited and unattrited UO_2 powders from four different lots of powder. Both the

Table 1
Powder characteristics before and after light attrition (5 min) of 10 kg lots of UO_2 powder

Specific surface area, $m^2 g^{-1}$		Average particle size, μm	
Before	After	Before	After
3.11	3.52	0.65	1.90
3.25	3.39	1.00	1.46
3.44	3.76	1.02	1.65
3.39	3.36	1.20	1.38

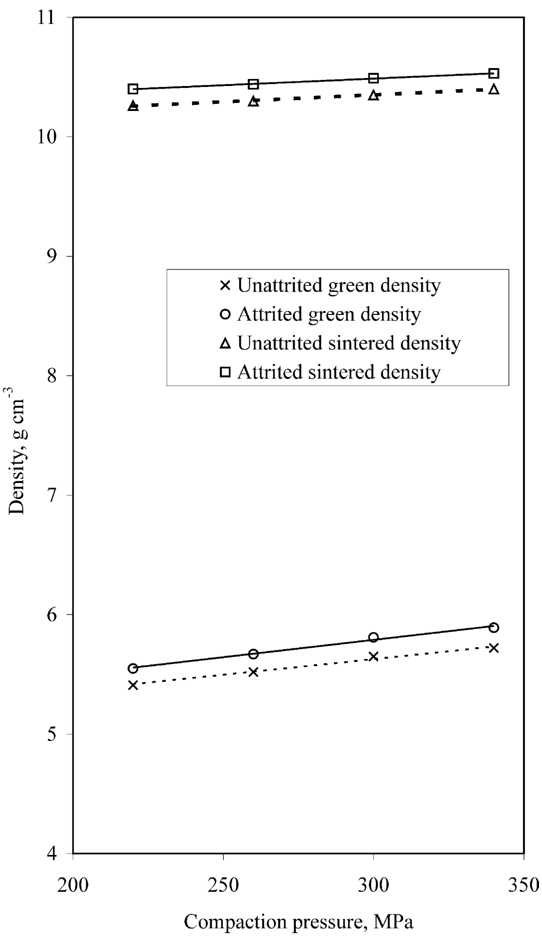

Fig. 2. Variation of green and sintered densities of pellets made from unattrited and attrited powders with varying compaction pressures.

specific surface area and the average particle size have increased on attrition. The increase in specific surface area indicates that new surface has been generated by the separation or fracture of bigger particle-clusters into smaller clusters. The increase in average particle size indicates loose re-agglomeration without loss of the newly generated surface. Pour density and tap density were 1.85 and 2.43 g cm^{-3} respectively before attrition and 1.77 and 2.68 g cm^{-3} respectively after attrtition. The increase in tap density indicates an improvement in packing efficiency of the powder on attrition. Fig. 2 gives green density and sintered density versus compaction pressure for compacts from attrited and unattrited powders. To achieve the same green density, the compaction pressure for attrited powders is about 80% of that required for unattrited powders. It is also noteworthy that the scatter in the density of compacts made from light attrited uranium dioxide powder is significantly less when compared to the corresponding unattrited category. For example, the standard deviation obtained for 10 samples from compacts made from 10 kg powder of unattrited powder is 0.106 g cm^{-3} whereas the corresponding attrited category showed

Table 2
Axial compressive stress for failure of green compacts made from unattrited and lightly attrited uranium dioxide powders and the number of compacts that failed

Unattrited		Attrited	
Failure stress MPa	No. of compacts failed	Failure stress MPa	No. of compacts failed
3.2664	3	4.083	2
3.5386	2	4.6274	3
4.083	4	4.8996	4
4.6274	2	5.444	3
4.8996	4	5.9884	1
5.444	7	6.2606	1
5.9884	5	6.5328	2
6.5328	3	6.805	4
6.805	3	7.6216	6
7.3494	2	8.166	5
7.6216	3	8.7104	4
8.166	4	8.9826	1
8.9826	1	9.527	3
10.0714	2	10.0714	1
–	–	10.3436	3
–	–	11.4324	1
–	–	11.7046	1

standard deviation of only 0.045 g cm^{-3}. Similarly, the standard deviation in the sintered density of these compacts are 0.051 g cm^{-3} and 0.036 g cm^{-3} respectively. Table 2 gives the failure axial compressive stress for green pellets prepared using attrited and unattrited uranium dioxide powders.

The mean compressive stress of failure for the lightly attrited category is significantly higher than that for the unattrited category. Fig. 3 gives typical recoveries of finished pellets from lightly attrited and unattrited powders of the same lot. Each point in the graph corresponds to one powder lot. The X coordinate of the point represents recovery from the powder in unattrited condition. The Y coordinate of the point represents recovery in attrited condition. If the recoveries from both categories are the same, all the points would lie on the 45° straight line from the origin. In all the cases, the recoveries from attrited powders are higher than those from unattrited powders.

4. Discussion

Green strength is important not only for handling the compacts but also to minimize defects, such as end-capping and lamination in the compacts [15]. Admixed lubricants and binder addition is one way to enhance green strength [21]. Attrition of the powder is another method. The coordination number, n_c among particles in a loose powder is 2–4 [22–24]. A powder compact with green density 59%, on the other hand, has the n_c value of 7–9 [25]. By virtue of higher green density in

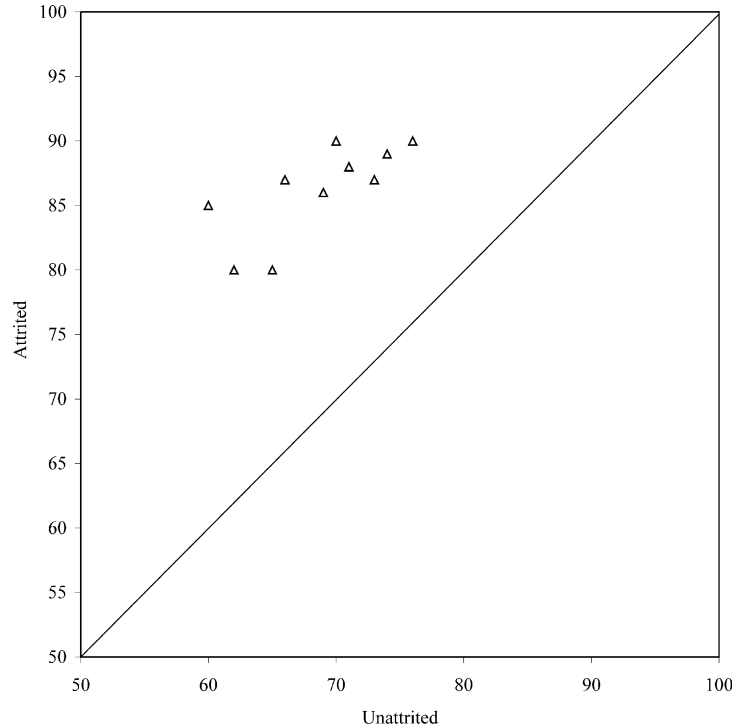

Fig. 3. Comparison of recoveries obtained with pellets made from unattrited an attrited powders.

attrited powder compacts, a higher coordination number is expected. Green strength is expected to increase as the coordination number increases. Factors affecting green strength were studied by Thomson [26] and Rumpf [27–30]. The Rumpf equation was modified by Kendall et al. [31–33] with the application of Griffith theory of fracture. Taking into account flaw length, elasticity of the particle assembly and other factors, green strength is given by

$$\sigma = 0.85\varphi^2 E^{1/6}\gamma_{gb}^{5/6}/\left(c^{1/2}r^{2/3}\right)$$

where c is the flaw length, E is Young's Modulus and $\gamma_{\mathbf{gb}}$ is the grain boundary energy, r is the particle radius and φ is the solid volume fraction. The higher green density of the compacts from attrited powders means a higher value of φ and therefore greater green strength. In an attritor, the agglomerates get separated into individual particles or into smaller particle clusters, thus decreasing the value of r. The flaws (whose length is represented by c) in a green compact could be the voids present or cracks generated on compaction pressure withdrawal (endcapping) or on ejection (lamination). Voids between clusters are larger than those between particles. Voids in compacts from attrited powder are therefore much smaller, leading to higher green strength, enabling the compact to withstand stress relief and ejection without flaw generation.

The experimentally obtained failure load frequency distribution (shown in Fig. 4) indicates that the largest

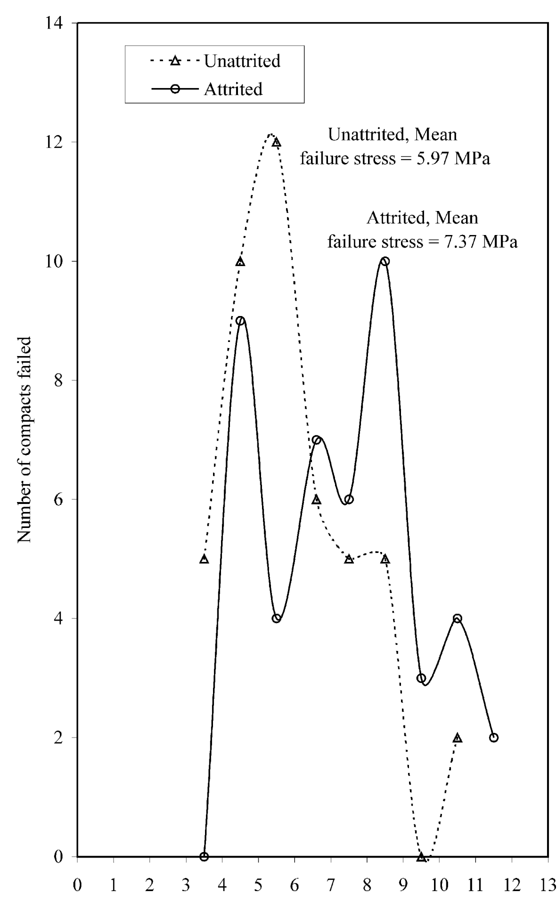

Fig. 4. Failure frequency distribution of compacts made from unattrited and attrited powders.

number of compacts failed at 5.5 MPa in unattrited category. In the attrited category, the largest number of compacts failed at considerably higher load of of 8.5 MPa. This must be due to the fact that the number of large flaws in powder compact from unattrited powder is greater than that in the compact made from an attrited powder. The flaw size in a fine powder is expected to be small in relation to that of a coarse one. The Particle Size Distribution (PSD) of an unattrited and attrited powder are shown in Figs. 5 and 6. Before attrition, the powder was coarse and unimodal (10 μm). After light attrition for 5 min, some fines were generated while some coarse particles remained, making the distribution bimodal (0.4 and 10 μm). It is expected that prolonged attrition would finally lead to fine unimodal PSD (0.4 μm). It has been reported elsewhere that a powder of bimodal PSD became unimodal after attrition for one hour by the destruction of aggregates into primary particles [34]. However, the increase in attrition times for uranium dioxide powder is limited by concerns of contamination by grinding media and the possibility of reoxidation of UO_2 powder.

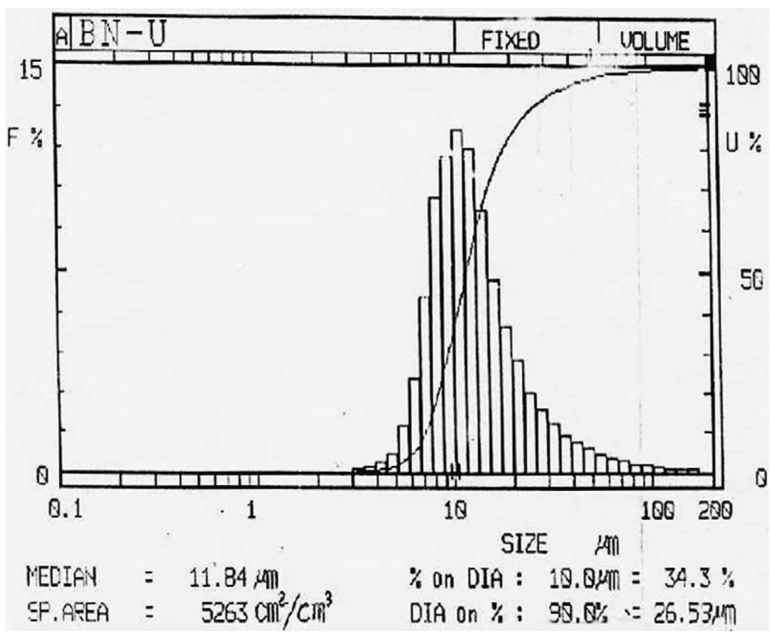

Fig. 5. PSD of unattrited powder.

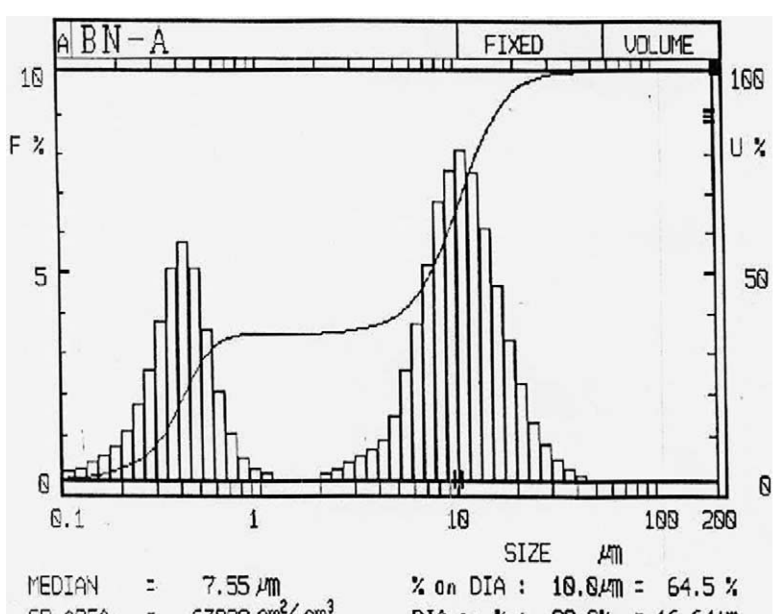

Fig. 6. PSD of lightly attrited powder.

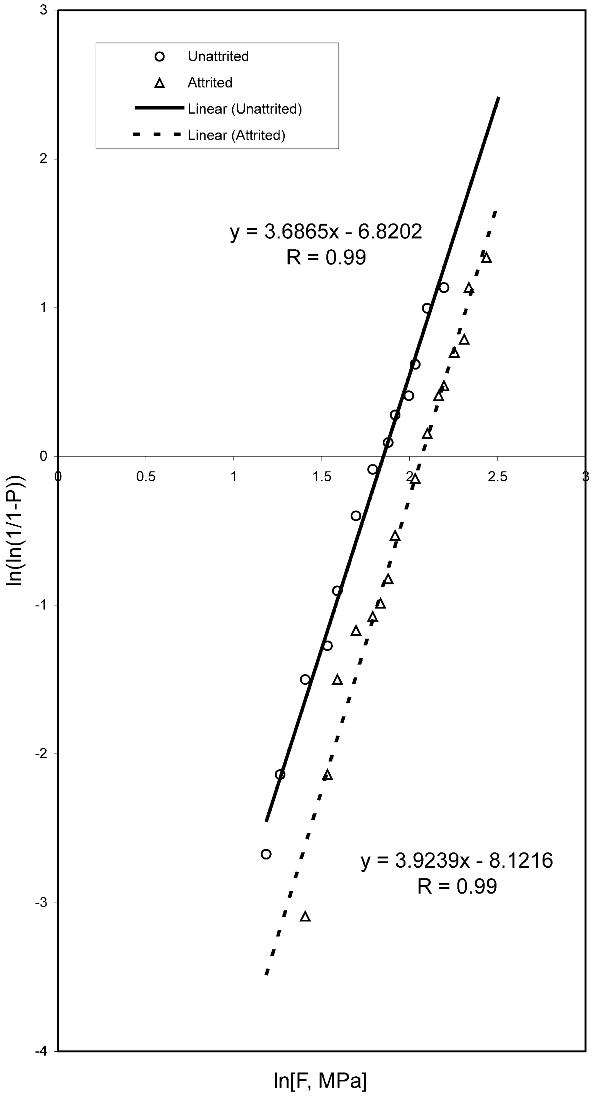

Fig. 7. Weibull plot for green compacts made from unattrited and lightly attrited UO_2 powders.

The presence of flaws and defects of varying sizes and orientations in a green compact leads to the scatter in the value of axial compressive stress of failure even in the pellets derived from the same lot of powder. It is possible to estimate a measure of failure stress under this kind of variation with adequate confidence with the use of the Weibull statistic [35,36]. Using the experimentally obtained data of axial compressive stress of failure for the green compacts of unattrited and attrited varieties, the Weibull plots for unattrited and attrited categories are shown in Fig. 7. The stresses corresponding to 0.5 probability of failure are 7.24 and 5.75 MPa for lightly attrited and unattrited categories respectively. The Weibull modulii are 3.92 and 3.69 respectively. These are indicative of the greater strength and lesser spread in the attrited category. The above values are applicable for a light attrition period of only five minutes.

5. Conclusion

A simple attritor and a hydraulic compressive strength testing rig have been built in-house and used for UO_2 powder. The attrition process is especially suited for powders prepared from low or room temperature precipitated precursors, being loosely agglomerated. Significant de-agglomeration could be achieved by attriting 10 kg of powder per batch in only 5 min, without the risk of contamination from the grinding media. A minor improvement in green density and sintered density, and a significant improvement in green strength (by virtue of higher packing efficeincy and smaller flaw sizes resulting from finer powders), density variation and recovery are the benefits of processing attrited powders. The results have been discussed in the light of attrition effects, green strength theory and Weibull analysis.

Acknowledgements

We thank Mr. R.M. Kartha for carrying out UO_2 powder attrition, Mr. R.R.K. Raju for compaction, Mr. R.N. Jayaraj for high temperature sintering, Mr. Vasant Rao and Mr. R. Ashok Kumar for the fabrication of pellet testing rig and to Mr. S.C. Jain, Mr. S. Syamsundar, Mr. B. Gopalan, Mr. N.P.S. Katiyar and Mr. R.B. Yadav for their useful discussions.

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