Journal of the European Ceramic Society **18** (1998) 1539–1543

PII: S0955-2219(98)00033-8
0955-2219/98/$—see front matter

Minimising Density Distribution in Die-pressed, Cylindrical Compacts*

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(Received 14 July 1997; accepted 2 February 1998)

Abstract

There is significant interest in potential new 'net shape' or 'near-net shape' forming processes for ceramic materials, to cut production costs through eliminating finishing operations. However, the new processes presented generally require a complete change from conventional industrial processing, which may well lead to problems with broad acceptance. Die-pressing is a widely used, cost effective industrial process. However, green compacts are produced with a density distribution which leads to shape distortions in fired components, precluding 'net shape' forming. This paper shows that, through optimising process variables, to minimise density distribution, the 'net shape' forming of simple, cylindrical components is achievable, by die-pressing, i.e. without large departures from the standard processing technique. For more complex shapes further factors become significant, however, this work should be an important step towards also achieving the 'net shape' forming of more complex shapes, by die-pressing. © *1998 Elsevier Science Limited. All rights reserved*

1 Introduction

'Net shape' forming is the production of components with the required tolerances directly from the forming process. However, it is normal in industry that ceramic components need a grinding or polishing operation as a finishing process. The inherent hardness of ceramics means that such operations are a relatively slow and expensive element of the ceramic production route. Therefore, their elimination would present considerable cost savings to processing and this has been the driver for significant research in the search for appropriate new processes.

The target for this work was to develop a 'net shape' forming process to produce a right cylindrical, pressurised water reactor (PWR) uranium dioxide fuel pellet with an allowable tolerance of only $\pm 12{\cdot}5\,\mu$m, on a diameter of the order 8 mm (the actual diameter value varies from customer to customer) without grinding. The pellet height is usually around 10 mm but is not as critical in terms of tolerance. An additional requirement was finding a process that caused the minimum disruption to the standard production route.

There are a number of new and existing fabrication processes under development for utilisation as 'net shape' forming processes.[1–8] However, when assessed against the above criteria all current processes were found to have some aspect that made them unacceptable, particularly in keeping to the requirement for minimal disruption to current practice. Therefore, the development of new processes based upon simple modifications of the standard die-pressing technique were investigated. This led to the development of two new techniques. A 'layering' technique, reported elsewhere,[9,10] and a technique based upon milling and lubrication of powder which is now reported here.

2 Background

Oxide nuclear fuel pellets are prepared using a standard powder compaction and firing route. Generally, granulates of uranium dioxide powder are mixed with a die lubricant (zinc stearate), fed into a cylindrical die cavity and uniaxially die compacted at pressures of around 4–6 tonnes cm^{-2} by top and bottom punches to produce compacts which are fired to provide the final component.

On compaction of the powder friction between powder-powder contacts and powder-die wall

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contacts prevents load being transmitted evenly throughout the compact. As a result powder compacts are produced with an uneven density.[11,12] In the case of a cylindrical shape, with an equal compaction load applied to both pellet ends, the central section has a lower average green density than the pellet ends. On firing the same final density is achieved, therefore, the centre of the pellet shrinks more than the ends. An 'hour glass' profile results. For a typical PWR pellet, end diameter can be up to 100 μm larger than the pellet centre diameter. Fired fuel pellets must therefore, be centreless ground to achieve tolerance.

Many factors, such as powder particle size, size distribution, shape, etc., will affect the compaction behaviour of a powder. The main factors in powders that hinder compaction are the presence of hard powder agglomerates and friction between powder-powder particle contacts and powder die-wall contacts.[10–12] Both these points were addressed in this work. The effect, with respect to pellet profile, of the quantity and distribution of lubrication was assessed and also the effect of milling of powders. The aim was to achieve the highest density possible in a green compact by applying the minimum load possible, in the belief that this would present the minimum density distribution and hence the minimum distortion on firing.

3 Experimental

Uranium dioxide powder produced by the BNFL integrated dry route (IDR) process was used. In the IDR process gas streams of uranium hexafluoride and steam are reacted in a rotary kiln, in a hydrogen atmosphere, to produce uranium dioxide and hydrogen fluoride.

Powder pour density was quantified by pouring approximately 50 g of powder into a measuring cylinder and taking readings of mass and volume. Tap density was measured by tapping the powder mass repeatedly until no further volume decrease occurred. Particle size analysis was measured by a standard sieve analysis technique to assess agglomerate size and by coulter counter.

The 'standard' process for uranium dioxide fuel pellet production was simulated in the laboratory as follows. Granules were produced using a precompaction method. Approximately 30 g of powder was poured into a 25 mm diameter steel precompaction die. The powder was then uniaxially compacted at 0·75 tonnes cm^{-2} using top and bottom punches to form a 'slug' compact which was broken through a 1·18 mm sieve to form 'rough' granulates of around 1 mm diameter. The granulates were then placed in a glass jar on rollers and 'conditioned' with 0·2 wt% zinc stearate die lubricant by turning for 10 min. The conditioning process mixes in the die lubricant and improves the flowability of the granules. The resultant granules were then pelleted, using an Apex press, in a 1·1 cm diameter steel die with a spring loaded mechanism to ensure pressing with equal load by the punches from both ends of the pellet. Green pellets were fired in a hydrogen atmosphere for 5 h at 1750°C. Pellet green density was determined by mensuration and fired density by a standard water immersion technique.

Milled powders were produced by dry ball milling 400 g of powder in a rubber lined ball mill with steel milling media. Time periods of 2–6 h were used. Powders produced by ball milling with lubricant were pelleted directly without using the precompaction stage.

Fired pellet profile was measured using a stylus profilometer in which pellets are laid on their side and a fine stylus dragged over the surface from one end of the pellet to the other.

4 Results and Discussion

Table 1 shows general characterisation data for the as-received uranium dioxide powder. Figure 1 shows the particle size distribution. Previous work[13] has shown that the powder has a plate like dendritic structure, common to a powder formed via a gaseous reduction process, it has also shown that this structure can be broken down by extended milling to a more spherical form. Figure 2 shows the compaction behaviour of 8 g samples of powder processed by the standard route. The maximum green density achieved was 6·00 Mg m^{-3} for a pelleting load of 4 tonnes cm^{-2}. The fired density achieved ranged from 10·62–10·82 Mg m^{-3} (Fig. 3).

The lubricant level added at the conditioning stage was increased from the standard 0·2 to 0·5 wt %. Comparing the green densities achieved in both cases for a pellet pressed at 1 tonne cm^{-2} a corresponding slight increase in green density from 4·73 to 4·89 Mg m^{-3} was observed. The fired density values remained the same.

Powder was ball milled then pelleted using the standard route. Figure 4, shows the increase in

Table 1. General characterisation data for the as-recieved powder

Pour density (Mg m^{-3})	0·75
Tap density (Mg m^{-3})	1·78
Surface area (m^2 g^{-1})	2·7
Sieve ret on 212 μm g/100 g	0·040
Sieve ret on 250 μm g/100 g	< 0·01
Sieve ret on 355 μm g/100 g	0·000
Sieve ret on 710 μm g/100 g	0·000

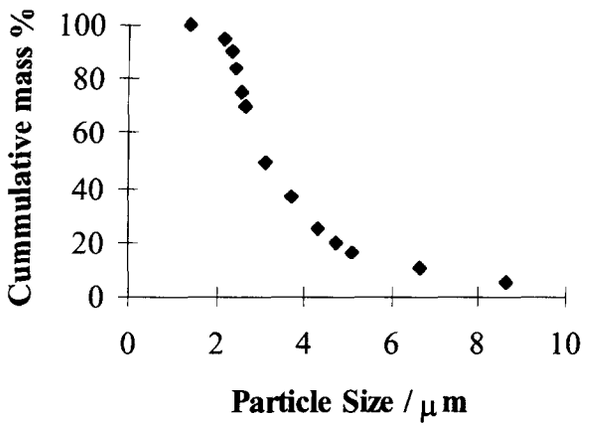


Fig. 1. Particle size distribution of the as-received powder measured by Coulter counter.

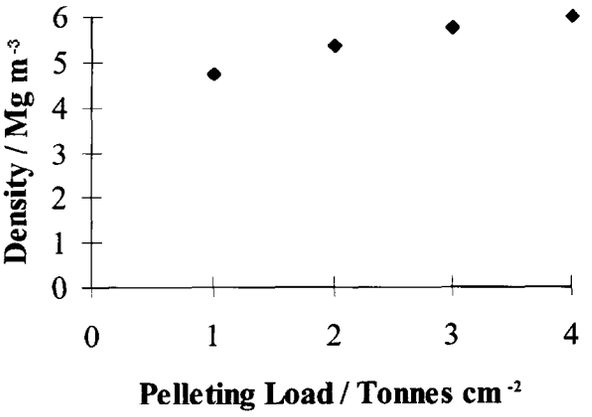


Fig. 2. Compaction behaviour of as-received powder prepared by the standard route.

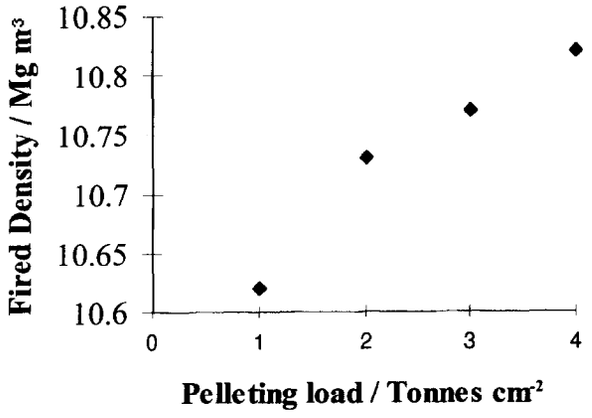


Fig. 3. Fired density as a function of pelleting load for powder processed by the standard route.

green density achieved for 8 g samples of powder pelleted at 1 tonne cm^{-2} after milling for increased periods of time. Materials milled for 6 h achieved a green density of 5·77 Mg m^{-3} a further improvement in comparison to the previous examples above in which only further lubricant was added. The maximum density achieved, for a pellet pressed at 3 tonnes cm^{-2} from powder milled for 6 h, was 6·32 Mg m^{-3}.

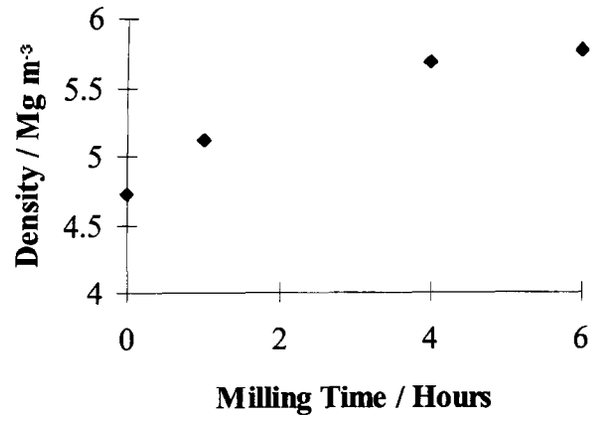


Fig. 4. Green compact density as a function of milling time for powders pressed at a load of 1 tonnes cm^{-2}.

Finally, powder was ball milled with increased lubricant. The lubricant in this case being added to the powder prior to milling instead of at the granule conditioning stage as in the previous examples. This means the powder and lubricant are milled together and hence a more intimate mix should result. Figure 5 shows the compaction behaviour of 8 g samples of powder milled for 4 h with a 0·5 wt % lubricant addition. Pelleting at 1 tonnes cm^{-2} produced a compact of green density 6·34 Mg m^{-3} and the highest green density achieved, for a pelleting load of 4 tonnes cm^{-2}, was 6·74 Mg m^{-3}. This is a significant increase of 6·75% in theoretical density in comparison to the as-received material prepared by the standard route. Pour and tap densities also showed a significant increase to 3·15 and 3·53 Mg m^{-3}, respectively, compare with Table 1. A narrow range was found in fired density from 10·83–10·88 Mg m^{-3}. However, a fired density over 10·8 Mg m^{-3} was only found in the as-received material prepared by the standard route when a pelleting load of 4 tonnes cm^{-2} was used, it could be achieved in this final example for each pelleting load down to 1 tonnes cm^{-2}.

Using the above results conditions were chosen for pellets to be prepared for fired profile measure-

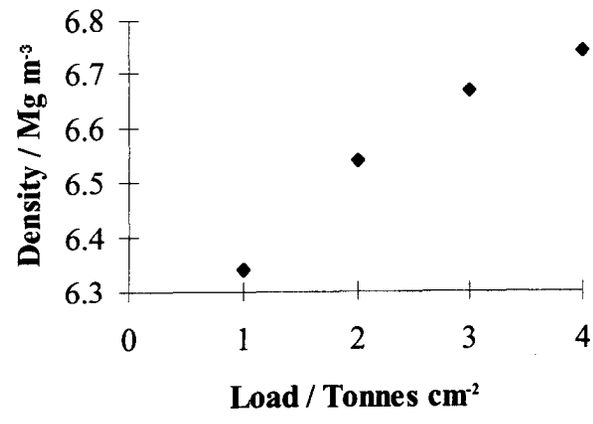
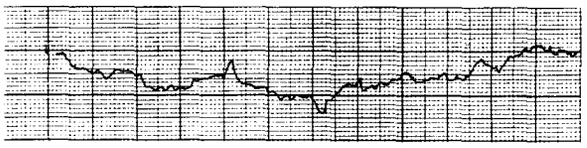


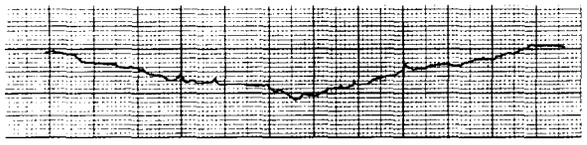
Fig. 5. Compaction behaviour of powder ball milled with 0·5 wt% lubricant and directly pelleted.

ment as an assessment of 'net shape' forming potential. 8 g pellets prepared via the standard route exhibited the expected 'hour-glass' fired pellet profile, with a peak to trough value of around 25 μm. The profiles showed no load dependence (Fig. 6).

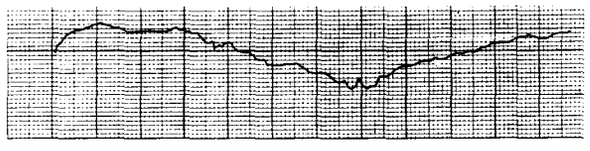
A vast improvement was found in pellet profile for 7 g pellets made from powders ball milled for 4 h with 0·5 wt% lubricant, with the 'hour-glass' effect reduced to a range of around 6–8 μm (Fig. 7). However, 8 g pellets showed increased 'hour-glass' profiles of the order 10–12 μm (Fig. 8). The fired pellet profile was improved further by preparing pellets from powders ball milled with lubricant for 4 h with a 1 wt% lubricant addition. For 8 g pellets minimal evidence of an 'hour-glass' profile was observed with a peak to trough difference along the pellet profile of the order of only 4 μm being achieved (Fig. 9). Such pellets could be considered to have a 'straight' profile. The straight profile was maintained for the 1 wt% lubricant example when the pellet mass was increased to 9 g (Fig. 9). However, pellets prepared with 1 wt% lubricant were fragile as a result of this high level of lubricant addition and green density measurements were not attempted. A further increase in powder pour and tap density was noted for the 1 wt% lubricant example in comparison to the 0·5 wt% example with values of pour density of 3·34 and tap density 3·67 Mg m^{-3} being measured.



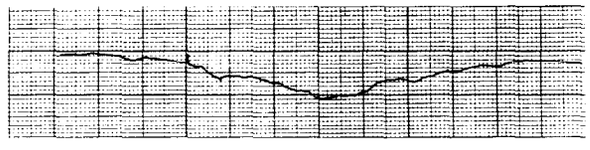
a) Compaction load 1 tonnes cm $^{-2}$



b) Compaction load 2 tonnes cm $^{-2}$



c) Compaction load 3 tonnes cm $^{-2}$



d) Compaction load 4 tonnes cm $^{-2}$

Scale : x-axis 1 mm = 0.1 mm, y-axis 1 mm = 2 μm.

Fig. 6. Fired pellet profile for 8 g pellets prepared by the standard route.

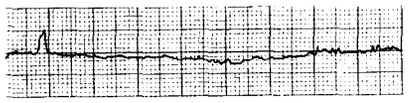


Fig. 7. Fired pellet profile for a 7 g pellet of powder ball milled for 4 h with 0·5 wt% lubricant and pressed at 1 tonnes cm^{-2} (Scale: x-axis 1 mm = 0·1 mm, y-axis 1 mm = 2 μm).

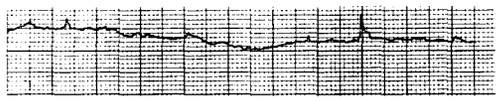
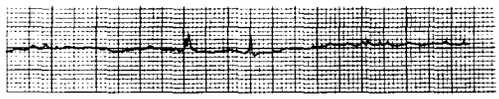
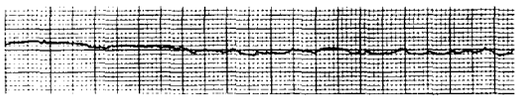


Fig. 8. Fired pellet profile for 8 g pellet of powder ball milled for 4 h with 0·5 wt% lubricant and pressed at 1 tonnes cm^{-2} (Scale: x-axis 1 mm = 0·1 mm, y-axis 1 mm = 2 μm).



a) 8g pellet



b) 9 g pellet

Fig. 9. Fired pellet profile for powder ball milled for 4 h with 1·0 wt% lubricant and pressed at 1 tonnes cm^{-2} (Scale: x-axis 1 mm = 0·1 mm, y-axis 1 mm = 2 μm).

5 Conclusions

Milling of powder with lubricant led to powders which compacted to higher green density at lower loads than that required for powders prepared from as-received material by the standard route. A maximum green density of 6·74 Mg m^{-3} was measured for powder milled with 0·5 wt% lubricant for a pelleting load of 4 tonnes cm^{-2}. This is a significant increase of 6·75% in theoretical density in comparison to the as-received material prepared by the standard route and pelleted at the same load. Fired pellet density was also increased.

The substantial increases achieved in pour and tap density for powders milled with lubricant in comparison to as-received materials is an important feature to note for the milled powder system. The packing behaviour of powder milled with

lubricant also indicates that internal friction between powder/powder contacts may play a more significant role in the compaction process than previously thought; the previous emphasis being on powder particle/die-wall friction.

Compaction of powder milled with 1·0 wt% lubricant produced fired pellets showing minimal evidence of density distribution in the green state via their fired pellet profile. 'Straight' pellets up to 9 g in weight were produced comparing very favourably with the 'hour-glass' profile produced by the standard route. This, therefore, presents a potential 'net shape' forming route for PWR pellets.

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