

ESTIMATION OF MAXIMUM COATED PARTICLE FUEL COMPACT PACKING FRACTION

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Abstract

This work develops an analytic fuel fraction packing model for a high temperature gas cooled reactor fuel compact fabricated from overcoated particles of a single size. The model includes the effects of one dimensional compression and finite matrix grain size. One dimensional compression limits the maximum fuel packing fraction to about 48% for the pressed compact in this single sized particle system. This limit is due to two effects. The first is that the process of die loading limits the pre-compression packing configuration to one that is stable under gravity, which is not the most space efficient one. The second effect is due to the one dimensional compression which reduces only the axial dimension of the particle lattice rather than uniformly compressing the lattice. The die wall can also limit the maximum packing fraction by preventing the nearby particles from moving into a more space efficient configuration.

1. Introduction

High Temperature Gas Cooled Reactor Fuel is often made by first applying a thin layer of matrix material to coated particle fuel and then compressing this overcoated fuel into a cylindrical fuel compact [1-4, 9]. The matrix is a combination of powdered graphite and binder material which is deformable at the processing temperature and forms the basic structure holding the compact together. In practice, the overcoated particles are poured into a heated cylindrical die and compressed in the axial dimension by a plunger (single or double acting). The deformable overcoating material then flows into the interstices and the resulting structure is a cylinder composed of fuel particles surrounded by a void free matrix. It has been observed that number of particles broken in the final fuel form increases with particle packing fraction [1,4].

Experimental evidence has shown that particles are more likely to touch each other along the axis of compression than along either axis perpendicular to the pressing direction [2]. This implies that the particle mass is being selectively compressed in one direction and simple maximum packing fraction estimates based on uniform compression of the interstitial space between particles may not be correct.

This work examines the compacting process by first determining the initial packing fraction of the overcoated particles as they are poured into the die and settled by shaking. This uncompressed packing fraction is the ratio of the volume of the overcoated particles to the total volume of the die. The lattice structure of this initial bed of overcoated particles must be stable to gravity and thus one cannot assume that the starting point is a particularly space efficient one.

After compression, the fuel packing fraction is defined as the ratio of the volume of the fuel particles to the sum of the fuel particle volume and the matrix material volume. The final particle lattice structure no longer has to be stable to gravity, but the distance a particle can move is limited so a radical lattice restructuring is not likely.

First approximations of the compact particle packing have assumed a hexagonal close packed array [3]. However, compact like experiments on the compression of lead shot have shown that randomly packed spheres start with an average of 8.41 contacts per sphere at low compression force and end with an average of 14.16 contacts (sides) per sphere (now space filling polyhedra) when the interstices are filled at high compression force. Away from the wall of the die, the now compressed spheres have 10 to 18 sides. In contrast, when the spheres were carefully stacked one-by-one in cannon ball fashion, the compression resulted in nearly perfect dodecahedra; 12 points of contact at the start (hexagonal close packed) and 12 sides at the end. The unilateral pressure caused a minor asymmetry [5]. Thus, random packing led to a much different result than careful stacking did.

In addition, experiments on fuel compacting implied that particle breakage was occurring on the outer surface region of the compact, possibly due to friction with the die walls during compression [2]. The experiments with lead shot showed that near the die wall the spheres were compressed into polyhedra with 8 to 14 sides; the average was about 11 [5]. Thus, the forces driving the distortion of the spheres

(overcoated particles) were modified by the presence of the die walls. Compacting experiments performed at Oak Ridge National Laboratory on overcoated natural uranium oxycarbide (NUCO) TRISO particles also show that overcoated particles are pressed into polyhedra-type shapes and these shapes are influenced by the die wall. This series of compacts had a fuel particle packing fraction of 38%, the TRISO particles were 780 μm in diameter, and the overcoating thickness was 160 μm . Figure 1 is an image taken of the side of a compact that was not pressed to full density. The once spherical overcoated particles can now be seen to have formed, as a result of the axial forces during pressing and impinging on the die wall, into polyhedra shaped particles in the same manner as the lead shot experiments (the compact was not disassembled and the particle polyhedral sides counted).

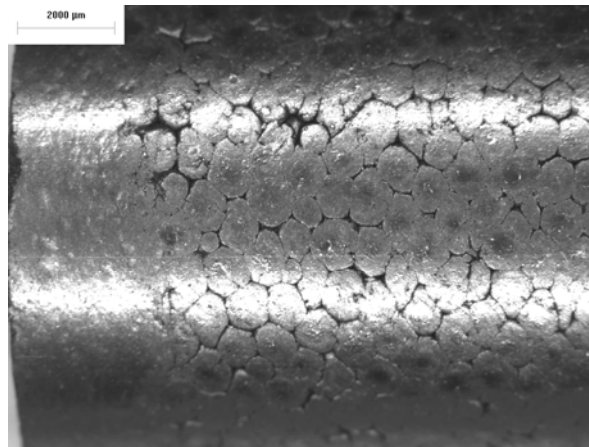


Figure 1. Photograph of the side of a compact that was not pressed to full density. The photo shows how spherical overcoated particles changed into space filling polygons as a result of the compression and interaction with the die wall.

Experiments on mechanical sphere packing have shown that the initial (as poured and vibrated) configuration is a combination of orthorhombic (mostly) and double nested structures [6]. These structures have 8 and 10 contact points, respectively, and thus one would expect just over 8 contact points per sphere (on average) as was seen in the lead shot experiments. However, the final (compressed) state of the lead shot with just over 14 contacts per sphere indicates that the spheres were not only compressed, but may have changed packing structure as well (the carefully stacked spheres started with 12 contacts and ended with 12 contacts). The fact that differences were seen between the spheres near the wall and those in the interior of the bed implies that the die wall may be modifying the structure in two ways. First, the presence of the wall limits the possible movement of the spheres as they are compressed; they are limited to moving along the wall or forcing their way back into the bed. Formation (by pressing) of NUCO compacts suggests that the overcoated particles cannot move back into the bed once they encounter the die wall. As a result, the overcoated particles begin to line up along the die wall, the boundary that will become the surface of the compact after it is ejected from the die.

X-ray tomography was performed on one compact and Figure 2 shows that the particles lined up vertically atop one another along the compact side due to the die wall effect rather than moving inward toward interstitial regions. This type of alignment presents a risk to the layers of the TRISO fuel particles located at the center of the overcoated spheres. As the particles begin to align on top of and next to one another, the overcoat between them can be squeezed out leaving TRISO particles touching one another. As pressure is continually applied during compaction, the TRISO particles can continue to press against one another, now in the absence of a cushioning overcoat layer, and crack.

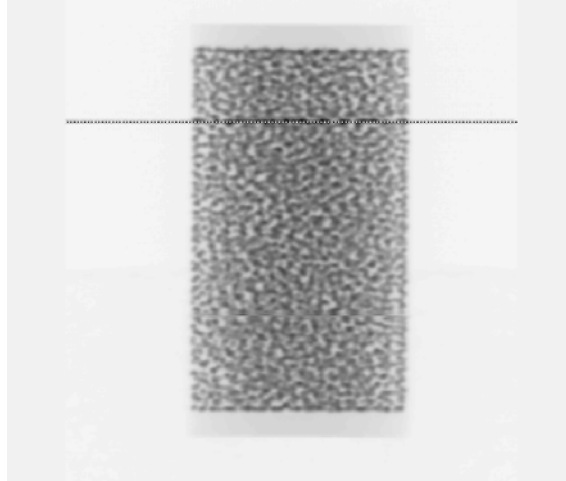


Figure 2. Radiograph of a compact showing particles lining up along the edge of the compact. In this rendering, particles are seen as dark spheres and porosity is seen in white. The dotted line is a machine reference.

Figure 3 is a cross-section of a compact that illustrates such cracking. The TRISO particles were aligned directly next to each other with little or no overcoat matrix remaining between them, leading to high local forces which resulted in a cracked particle.

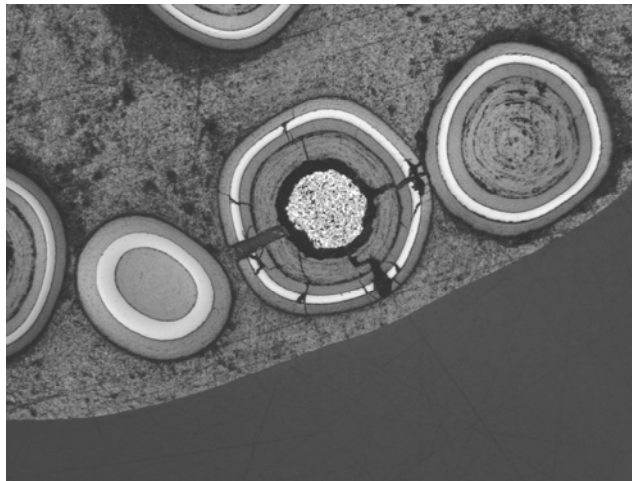


Figure 3. Cross-section of compact showing a cracked TRISO particle due to edge-to-edge alignment with another particle during pressing. Edge-on alignment was caused by the particle's inability to move while in contact with the die wall.

The second factor is that the friction between the wall and the spheres also restricts their movement and changes the force distribution at the wall, supplying tangential as well as normal forces on the compact surface. The result of these additional forces appears to aid the die wall stabilizing effect and further prevents the needed local lattice restructuring.

Since the fuel particles are located within the formed polyhedra and the flow of the overcoating material near and around the fuel particles can affect the near neighbor location and local forces, knowledge of the compressing may provide insight into the compact fabrication process. The above experimental evidence implies that the formation of fuel compacts by unilateral compression of a (settled) random sphere packing may influence the maximum practical particle fraction and this fraction may be different at the compact surface than in the compact interior.

2. Compact Compression Model

Random sphere packing has a packing fraction of about 0.625 when settled and an average of 8.41 contact points per sphere [5, 6]. The packing fraction for the orthorhombic packing with 8 contact points per sphere is 0.605 and that for the double nested with 10 contact points per sphere is 0.698. If one computes the fraction of the bed that is orthorhombic from the reported number of contact points (8.41), one gets 0.795; if one uses the packing fraction (0.625), one gets 0.785. Thus, one is reasonably confident that the data is consistent and that the initial bed can be approximated by a structure that is 0.79 orthorhombic and 0.21 double nested. The reader should note that this is not the most volume efficient sphere packing, but is a configuration that is stable under the shaking and settling process (gravity stable) used to fill the die.

When the spheres (overcoated particles) are poured into a circular die and settled by axial vibration, the spheres arrange themselves into concentric circles as viewed from the top and a staggered arrangement when viewed from the side [6].

The packing fraction, P_{orth} , as a function of angle is:

$$P_{orth} = \frac{\pi}{3\sqrt{3} \sin \theta} \quad (1)$$

The configuration is orthorhombic when $\theta=90^\circ$ and double nested when $\theta=60^\circ$. To achieve the 0.79 orthorhombic fraction, $\theta=75.3^\circ$ on average. Note that Equation (1) is the uncompressed overcoated particle packing fraction which is different from the fuel packing fraction. Its relevance is due to the fact that it defines the starting point for the process.

It should be noted that another approximate model for a randomly packed bed of spheres is a linear combination of a hexagonal close packed array and a cubic array [7]. However, these are unstable structures when shaken under gravity. The gravity stable starting lattice is an important constraint for this problem because, as noted above, the pre-compression starting condition is not the most space efficient packing.

Now suppose that the structure is compressed only vertically and the spheres transformed to space filling polyhedra so that the horizontal dimensions remain approximately the same, but the vertical dimension is reduced to 0.625 of its original value to allow the compressed spheres (now polyhedra) to completely fill the space. This is an approximation of the compact compression process based on the observation that touching or nearly touching particles are more likely to be seen in the axial cross section and the fuel particles tend to line up in columns near the die wall [2]. Since the TRISO particle must be protected from excessive force, only the matrix material (overcoating) can be distorted to fill the empty space. The goal of this process is maintain a roughly uniform thickness of matrix material around each TRISO particle during compression so no particles are touching.

2.1 Compacting Without Restructuring

In general, one would expect at least some lattice restructuring of the packing bed as the compression takes place. However, in one important case this may not be true. As described in the paragraphs above, near the die wall the particles (or lead shot) seem to have a different packing structure than in the center of the compact. In fact, Williams et al. investigated this phenomenon by performing a series of experiments on the region near the die wall. Williams et al. first formed the compact using their normal method and measured the broken particle fraction (overcoating was one of several methods investigated). These particular compacts were made in a two step process where a particle bed was lightly compressed under moderate temperatures in an undersized die (premolded) and then transferred to a compacting die and compressed under higher temperatures and pressures. Two cases of interest to this work were investigated. In the first case the premolded form was simply transferred to the compression die and compressed. In the second case, the premolded shape was coated with extra matrix material so that the compact outer particles would not touch the die wall. The authors were investigating the effects of die wall friction, but they also observed a restructuring effect (which they did not further speculate or comment on).

With the unfueled matrix region between the particle bed and the die wall, two key observations were made. First, fewer broken particles were noted; second, particles were seen to move from their premolded position into the applied fuel free matrix zone on the outer surface of the compact. This movement implies that the die wall was now having less effect on the bed structure near the outer edge of the compact. In other words, particle contact with the die wall appeared to stabilize the initial structure in a way that was not favorable to compact formation, above and beyond the effects of die friction. Evidence of particle breakage due to an inability to restructure near the die wall was shown in Figure 3. As the overcoated particles reach the die wall they become essentially frozen in place and are forced to line up edge-to-edge.

A contributing factor to this edge-on alignment is the malleability of the overcoat. If the overcoat is somewhat rigid and reluctant to flow in between the particles, the die wall effect will be magnified and further help to force particles into an edge-to-edge alignment. If the overcoat is more malleable it can more easily move into interstitial spaces between particles and therefore allow the internal TRISO particle to shift and avoid an edge-to-edge alignment with another TRISO particle. Processing techniques were developed to increase the malleability of the overcoat layer and prevent TRISO particle cracking at the compact edge as a result of edge-on alignment and retarded restructuring. Figure 4 is an image of a compact side after steps were taken to increase the malleability of the overcoat layer and more easily allow TRISO particles to lattice restructure.

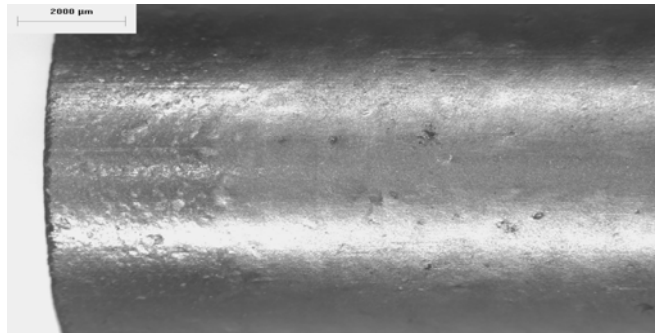


Figure 4. Photo of compact side after processing steps were performed to increase the malleability of the overcoat layer and aid in TRISO particle lattice restructuring at the die wall. The smooth surface indicates that fewer particles were pressed against the die wall (although the general effect is still present).

Basically, a more malleable overcoat allows the particle and the overcoat to move somewhat independently of one another in order to develop a more favorable (packing efficient) alignment during pressing. However, too malleable an overcoat will not cushion the TRISO particles sufficiently during compression; it will rapidly “squeeze out” from between particles and allow undesired particle contact. The fabricator is thus faced with a tradeoff situation.

We can investigate this effect by simply compressing the model without changing the angle θ or the horizontal spacing. Figure 5 shows the geometry of this configuration.

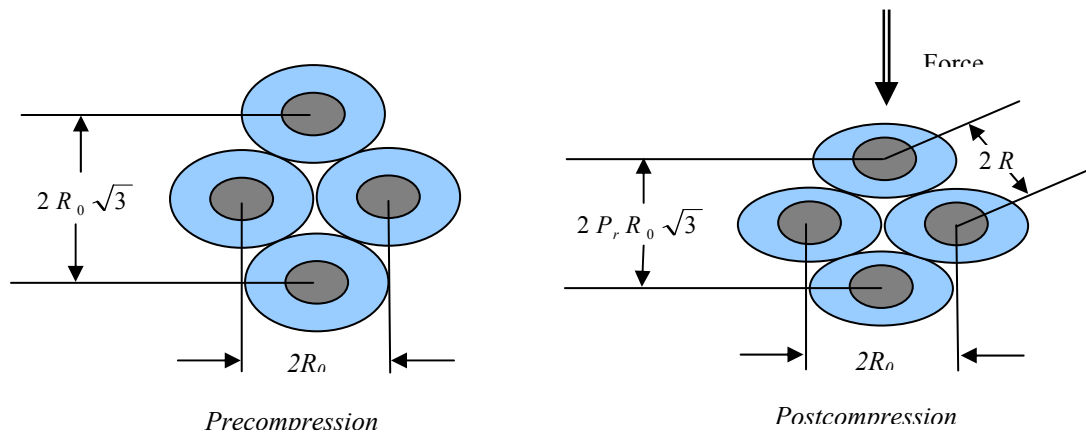


Figure 5. Compression without restructuring model

The relation between the spheres of interest (along the diagonal) is now:

$$2R = R_0 \sqrt{3P_r^2 + 1} \quad (2)$$

where R_0 is the uncompressed overcoated particle radius which is the sum of the particle radius, r_p , and the initial overcoating thickness, δ_0 ; R is the compressed overcoated particle dimension along the diagonal which is the sum of r_p and the compressed (no longer uniform) overcoating thickness, δ ; P_r is the random uncompressed sphere (overcoated particle) packing fraction. Equation (2) then allows us to form a relationship between the pre- and post-compression overcoat thickness along the diagonal (closest spacing):

$$\delta_0 = \frac{2}{\sqrt{3P_r^2 + 1}} (\delta + r_p) - r_p \quad (3)$$

Touching particles have been shown to be factor in fuel damage during compact formation and it is believed that some minimum distance between particles must be maintained during the compact forming process [2-4]. Equation (3) allows us to estimate the initial overcoat thickness as a function of the allowable compressed compact particle spacing and fuel particle radius. Throughout this work it will be assumed that the overcoating thickness is sufficient to fill all the interstitial space and that the matrix density does not significantly change during the compression process.

The fuel packing fraction (in the fueled zone), P_f , is:

$$P_f = \left[\frac{r_p}{r_p + \delta_0} \right]^3 \quad (4)$$

To just avoid crushing particles ($\delta=0$), Equation (3) leads to the following inequality:

$$\delta_0 \geq \left(\frac{2}{\sqrt{3P_r^2 + 1}} - 1 \right) r_p \quad (5)$$

Equation (4) then shows that a one dimensional vertical compression with no bed lattice restructuring leads to a maximum fuel packing fraction near the die wall of

$$P_{f \max}(wall) = \frac{(3P_r^2 + 1)^{3/2}}{8} = 0.40 \quad (6)$$

This implies that packing fractions of less than 40% would be safe targets since one would not have to depend on the particles near the die wall moving over or around each other to avoid hard contact.

Reference 4 suggests a particle center to center spacing of no less than 1100 microns for a pebble type fuel form; for their particle radius (460 microns), this corresponds to an average δ of 90 microns or a maximum P_f near the wall of 23.4% for this restrictive condition in an equivalent compact (this result appears to be quite conservative). It also implies that the approximately 12% pebble fuel loading fraction is quite a safe value regardless of the uniformity of the compression process.

For a fuel compact 12mm in diameter, roughly the outer 1mm would be expected to be in the die wall affected region. This corresponds to about 17% of the compact volume, a modest, but significant amount. One would expect that most particles that come into contact would slide by each other without undo problems, but clearly the potential for damage exists. Thus, focusing research effort on wall

interactions beyond friction may be of benefit. This situation is, of course, a worse case condition and applies only to the surface region of the compact, but it serves to illustrate that the as poured overcoated particle bed must undergo a lattice restructuring from its initial packing configuration to some other more space efficient configuration if high fuel packing fractions are to be obtained with overcoated particles, unidirectional compression and random die filling.

Since workers in this field have spent a considerable amount of effort in controlling the compacting conditions to avoid particle breakage, it is clear that reliably obtaining this change in structure, especially near the die wall, is not easy. The genesis of this problem is that the initial filling of the die must have a lattice that is stable under gravity and this lattice is not the most space efficient one. The one dimensional compression exaggerates this non-optimal situation.

2.2 Compacting With Restructuring

During the compacting process, the particle lattice planes can shift. Since the matrix material now holds the particles in place, the configuration does not have to be gravity stable. It is not exactly clear how this final state can be determined, but it seems likely that a radical change would not happen since the average compression distance per particle is less than half of an overcoated particle diameter and the compression is a smooth process. Thus, it reasonable to assume that the structure can still be globally approximated by an orthorhombic and double nested mix.

The reader may wonder why the assumption of a hexagonal close packed lattice is not made as the final lattice. There are two reasons for this. The first is that the experiments sited in the previous sections showed that compression of a random bed of spheres (lead shot) lead to polyhedra with not only a rather wide distribution of (uneven) sides, but with an average number of sides that was different than what one would expect for a hexagonal closed packed lattice (about 14 versus 12). In fact, the spheres had to be hand packed one-by-one to get the hexagonal close packed lattice. In addition, we have seen that local effects such as a die wall can hamper lattice changes.

Second, there are only limited mechanisms for the movement of the particles. As was noted above, the compression process offers little hope as a mechanism of radical restructuring because of the limited range of movement and smooth compression; the pre-compression lattice is not shaken or stretched to the extent that particles can replace one another or move the length of a lattice side or more. The TRISO fuel particle is essentially trapped near the center of the overcoated particle and only moves a fraction of a lattice dimension as the matrix material flows around it. The strong forces that could effect a major change in global lattice structure, hard particle-to-particle contact, are the same forces we wish to avoid. Thus, we come to the conclusion that as a first approximation the lattice structure change must be no more than a modest shift in lattice planes since this structure change does not require more than a fractional shift in fuel particle position and meets our constraint of minimal TRISO fuel particle contact.

Figure 6 shows the compression of the restructured mix (mostly double nested shown). In this case the compression is slightly different because the restructuring makes the lattice more volumetrically efficient so that the compression distance (after the lattice changes) is now P_{orth} .

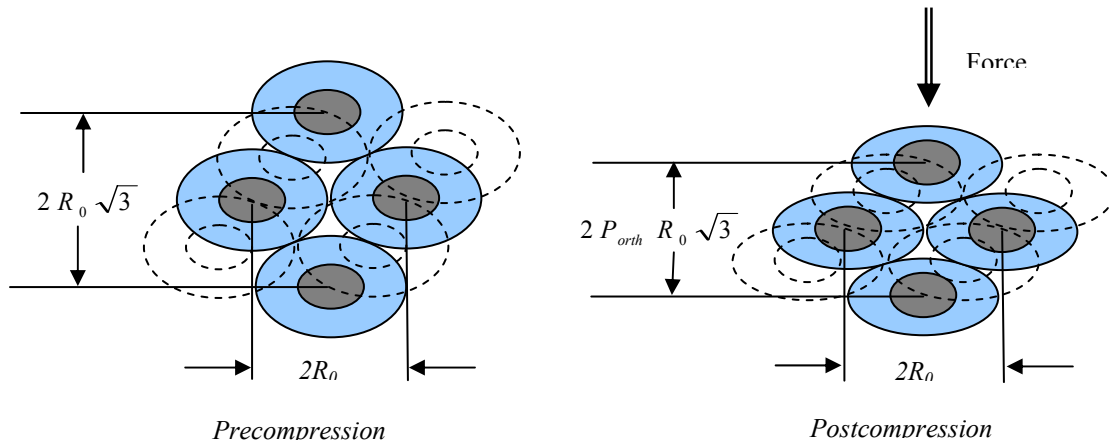


Figure 6. Compression with restructuring model

Since the restructuring only involves shifting a side plane of atoms, the vertical geometry remains the same as before and Equation (3) can be generalized:

$$\delta_0 = \frac{2}{\sqrt{3P_{orth}^2 + 1}} (\delta + r_p) - r_p \quad (7)$$

where $60^\circ \leq \theta \leq 90^\circ$ ($\sim 60^\circ$ for the highest packing fraction). Equation (5) then becomes:

$$\delta_0 \geq \left(\frac{2}{\sqrt{3P_{orth}^2 + 1}} - 1 \right) r_p \quad (8)$$

Finally, the maximum fuel packing fraction ($\delta=0$) can also be generalized:

$$P_{f \max} = \frac{(3P_{orth}^2 + 1)^{3/2}}{8} \quad (9)$$

With a complete lattice change to double nested, $P_{f \max}=0.483$; this is higher than the previous unstructured value of 0.40, but still far less than the 0.625 (or more for hexagonal closed packed array) value one might get from the isometric compression of a randomly packed bed. In actual practice this model may be somewhat more restrictive than necessary since it does not account for horizontal movement, but the compression is clearly not isometric and the starting structure is not well approximated by a hexagonal close packed array. The literature limit on one dimensional compacting appears to be about 50%, close to the above value [9].

3. Estimation of Minimum δ

Fuel particles can be damaged with a force of roughly 25N so control of the applied plunger force during compacting is important as well as the force required to deform the overcoating [1-4]. Particle damage can occur by pressing particles against a die or plunger with excessive force, by pressing two particles together with excessive force (as seen in Figure 3), or by pressing two particles together with unyielding overcoating material between them. If the overcoating is too soft, the fuel particle may push through the overcoat, thus leaving it uncushioned and susceptible to damage. If the overcoat is too difficult to deform, it may transmit high forces to the particle. In practice, the process is a trade off because one would like to use as great a compressing force as possible to insure a high density matrix [2, 3]. The compacting research on NUCO TRISO particles has found that the optimal piston pressing force for a 12mm diameter by 25mm long compact is less than 1780N with a matrix having the ideal viscoelastic texture. If steps are not taken to increase the malleability of the overcoat a pressing force of >4900N is needed to form the compact.

The minimum acceptable spacing between particles is not simple to determine. However, an order of magnitude estimate can be made by focusing on the underlying granular nature of the matrix material (overcoating). When the dimensions of the flow region are of the same order as the material grain size, material flow can be inhibited or blocked by the creation of granular arches. Several grains of material can stick to each other and one end of this structure can contact the flow region defining surface (in our case, the surfaces of the fuel particles). Flow shear (higher velocity in the center of a flow channel than near the wall) can then rotate this structure perpendicular to the flow direction and wedge it into place across the flow path, thus blocking the flow and forming a rigid force transmitting bridge held in place by friction and hydraulic pressure.

While the flow channels in a particle bed are complex, one can begin to make some rough estimates. The most basic limitation on the particle spacing is that the spacing must be at least that of a matrix grain diameter, g_m , to prevent a grain from being trapped between fuel particles and forming a rigid contact region. In this case we have:

$$g_m < 2\delta \quad (10)$$

This is certainly a lower limit; however, it does not address the bridging problem. Studies on the flow of particle laden fluids in micro tubes have show that tube plugging is most likely to occur when the diameter of the grain is between approximately 0.33 and 0.46 the diameter of the tube [7]. While the flow through the compact interstices is more complex than simple tube flow, we can use it as a rough bounds. Using the fuel particle spacing, 2δ , as an effective tube diameter and the lower limit on the plugging grain ratio, 0.33, one can see that the particle spacing should be at least 3 times the matrix grain size:

$$1.5g_m < \delta \quad (11)$$

Equation (11) should be viewed in the light of an order of magnitude estimate as other factors such as the stickiness of the binder and grain shape also come into play; nonetheless, the results of this simple analysis show that the grain size of the matrix material must be taken into consideration and could become important as the upper packing limit is approached, if large grained matrix material is used, or if the matrix has a tendency to clump.

In Section 2, a δ of 90 microns was computed based on past literature. Using this in Equation (11) would imply an effective matrix grain size of 60 microns, which appears to be about a factor of 6 too high (the actual grain size is unknown to these authors). Thus, the factor of 1.5 may be on the low side or other factors during the fabrication or even irradiation may dominate. Given the limited information available in this area, Equation (11) is probably the best that can be done at the present time.

4. Discussion

An equation relating the maximum acceptable packing fraction, P_{acc} to particle and matrix grain size can be generated by combining Equations (4), (7), and (11):

$$P_{acc} = \frac{1}{8} \left[\frac{\sqrt{3P_{orth}^2 + 1}}{(1 + 1.5g_m/r_p)} \right]^3 \quad (12)$$

Typical values for r_p and g_m might be 460 microns and 10 microns, respectively, leading to a P_{acc} of 44% for a fully doubled nested lattice. This estimate, of course, assumes only ideal vertical compression and does not include the random variations that one can expect to occur in a process of this nature, but it does show that the non-ideal effects of one dimensional compression and non-zero matrix grain size can have a significant effect on the maximum fuel packing fraction, greatly reducing it from the 62.5% value for (settled) randomly packed spheres with isometric compression and modestly reducing it from the zero grain size value of 48%.

A worst case die wall packing fraction with the above parameters except with $\theta = 90^\circ$, leads to a P_{acc} of 34%, which can certainly be assumed to be the lower limit on P_{acc} .

If we compute P_f for a δ of 90 microns with the other parameters unchanged [Equations (4) and (7)], one gets $P_f=0.28$. Since high quality compacts have been made in the 35~40% packing fraction range, this value for δ may be somewhat conservative.

One question that arises is whether this analysis can be applied to other fuel form fabrication methods. It certainly does not apply to the injection method for compact fabrication, since overcoating is not applied to the fuel particles in this case and the matrix material is injected into a static particle bed. The fabrication of fuel pebbles (spheres) is more isometric than compacts, so the compression effects should be much less pronounced. The matrix grain limits would apply, however. In addition, the target packing fractions for pebbles are only about one half or less than that of compacts, which would mitigate these problems.

5. Conclusion

The inclusion of the non-ideal effects of gravity stable die filling, one dimensional fabrication compression, and non-zero matrix grain size was shown to have a significant effect on the maximum compact fuel packing fraction for a single size particle system. The model maximum was shown to be 48% with no matrix grain effects and in the neighborhood of 44% when including some possible matrix grain effects for typical parameters. Unimpeded die wall effects could lower this value to 40% (or even less if grain size effects are important). Thus, the maximum practical compact packing fraction appears to be in the neighborhood of 40-50% with this model, assuming particles can slide over one another to some extent, especially near the die wall.

When compared to past data, this model is somewhat conservative as literature packing values of up to 50% have been reported [9], although recent production level targets have been closer to 40%. A model limitation is that the particles are not allowed to slide by one another by more than about a half an overcoated particle diameter during compacting and no horizontal movement is included.

The initial source of these problems is the unidirectional bed compression combined with the fact that the initial overcoated particle loading of the die must have a lattice that is stable to gravity under shaking. The resulting starting structure is not the most packing efficient one and its lattice structure must change during compacting if high packing fractions are to be achieved. The particle movement or lack of movement during compacting can then lead to touching particles which greatly increases their likelihood of damage. In addition, the die wall can have a stabilizing effect on the nearby particle structure which inhibits the restructuring necessary to generate high particle packing fraction compacts. Future compacting work might focus on this compact/wall interaction region with the goal of providing room for the edge particles to move prior to contacting the wall.

6. References

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