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THE EFFECT OF PROCESS PARAMETERS ON THE STRUCTURAL INTEGRITY OF THE COATED PARTICLES DURING FABRICATION

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1. INTRODUCTION

During fabrication of the nuclear compact, a packed bed of coated particles is subjected to an external molding pressure at the top of the bed. The pressure is required to inject the viscous matrix material into the packed bed. This pressure is primarily resisted by the packed bed. This leads to very high contact stress between the coated particles and the mold, potentially resulting in loss of structural integrity of the particles and, consequently, manufacturing defects.

The process parameters, such as composition and viscosity of the matrix material, time history of temperatures and molding pressures etc., have been nearly optimized for a full-size annular compact (3.507 in. o.d. x 1.395 in. i. d. x 1.45 in. length) in a separate development program. The particle failure rate has been found to be acceptable. A full-size compact is not advisable for irradiation experiments. Consequently, a reduced-size cylindrical compact of 0.49 in. diameter x 0.49 in. length has been proposed for some irradiation experiments in the technology qualification program.

The objective of this analysis is to find out how process parameters, in particular the molding pressure, affect the structural integrity of the coated particles for different compacts sizes.

The two parameter Weibull theory [1] is used to assess the initial failure of the coated particles under load. This theory accounts for the size effect of the compact. The Weibull modulus, m, is six based on the particle crush strength test data. The Weibull theory predicts that the molding pressure for a small compact would be about 2.35 times that for a full-size compact if the two compacts were assumed to have the same failure probability for the coated particles. The duration of this applied pressure can, however, be shorter.

2. OVERVIEW OF THE COATED PARTICLE DESIGN AND STRENGTH

The coated particle examined consists of a kernal of nuclear materials, surrounded by layers of coatings, which typically include a porous buffer layer, a dense inner pyrocarbon (IPyC) layer, a dense S_iC layer, and a dense outer pyrocarbon (OPyC) layer.

The mean diameter of the coated particle for a typical batch is about 0.0347 in. with a standard deviation of ~0.001 in. Average sphericity (defined as the ratio of maximum diameter to minimum diameter) of the particle is 1.03 with a standard deviation of 0.02.

Diametral compression tests were performed on the coated particles to determine their crush strength. A typical load deflection curve is given in Fig. 1. Almost all curves had a dip at about 3.0 lb load. The dip corresponded to a load reduction of 0.17 to 0.35 lb, and a deflection of 0.001 in. Few of the particles had two load dips. The initial cracking load, P_i, was taken as the lowest value in cases where there was more than one load drop in the load deflection curve. The test results are summarized in Table 1.

The dip in the load deflection curve has been interpreted in [2] as the Hertzian crack around the edge of the contact area between the spherical particle and the platen of the loading ram. P_i was associated with a Hertzian failure (cracking) of the OPyC layer, and the higher ultimate load, P_u, was associated with a failure of the S_iC layer and propagation of the crack(s) through the entire particle. Because of two different failure mechanisms, no correlation is expected between P_i and P_u.

During the diametral compression test, the particles were seated in a very rough circular socket. This reduced contact stress (by increasing contact area). This is the reason why in most cases one load dip was observed.

Every load deflection curve for a coated particle subjected to diametral compression test exhibited nonlinear behavior at a load below ~0.45 lb, as indicated in Fig. 1. The nonlinear initial portion of the curve was a manifestation of the deformation in the contact zone. The Hertzian theory [3] predicts that the applied load is proportional to $\delta^{3/2}$, where δ is the maximum local deformation in the contact zone. The observed load was consistent with that predicted within the accuracy of the recording curve.

Beyond the 0.45 lb load, the contribution to the total deflection from deformations of sources other than the contact zone was significant. Load deflection becomes linearly proportional to the applied load.

Reference 1 gave a handy formula to estimate the Weibull modulus, m, from the coefficient of variation (COV) for the two parameter Weibull theory. The formula is

$$m = (COV)^{-1/0.94}$$

Using this formula and the COV for P_i, one obtains m value ranging from 5.4 to 6.4. These values are consistent with the value of m for the strength of sectioned SiC ring [4]. For the current calculation, m is taken to be six for the coated particle compressed diametrally to initial cracking failure.

3. DESCRIPTION OF COMPACT FABRICATION PROCESS

The following is a brief description summarizing the compact fabrication process. First coated particles are poured into the mold. A plunger is used to pack the coated particle bed to achieve a void fraction of about 42%.

The packed particle bed is then heated at its outer perimeter to a slightly elevated temperature. Viscous matrix material is immediately poured on top the bed. Constant gauge pressure is applied within a few seconds through a press (with a diameter of 2.75 in.) at the top of the bed. This pressure is maintained until the molten matrix material is forced into the void spaces within the bed. Excess matrix material is drained through the small holes at the bottom of the mold. The packed particle bed with the injected matrix material is cured at an elevated temperature for a few minutes to form a green compact. The green compact is removed from the mold, and is heat treated (carbonized) at high temperature in an inert environment to drive off volatile products.

In the fabrication of the reduced-size compact, all process parameters with the exception of the gauge pressure, are at or near the optimized values used for the full-size compact.

Prior to the application of the pressure load, the only load on the bed is the dead weight of the particles. Immediately after the pressure load is applied, the bed is subjected to the full pressure load. In parts of the compact, this pressure load is lower due to mold wall friction and particle-to-particle friction. The majority of the applied pressure is resisted by the particle bed and small fraction of it forces the matrix material to flow through the porous bed. Particles at this time have the highest contact stress. A few seconds later, the bed with the matrix-filled void is in a quasi-static equilibrium under a nearly hydrostatic pressure state. As the curing occurs, the compact is solidified.

The fabrication process is a transient phenomenon. The particle stress is time dependent, and highest immediately after the pressure load application. Consequently, it is assumed that the most particle damage occurs at this time.

4. EFFECT OF PRESSURE ON PARTICLE BREAKAGE IN ANNULAR COMPACT

Data on coating breakage in resin-based full-size annular compacts from burn-back examinations of over 300,000 particles for each compact are summarized below:

Compact	Nominal Gauge Pressure, psi	Breakage %
A	200	0.03488
B	350	0.06534
C	500	0.08935

Regression analysis is preformed to correlate the percent of breakage (Br) to the gauge pressure (P_g in psi), one obtains

$$Br\ (\%) = 2.833 \times 10^{-18}\ P_g^{\ 6} + 4.666 \times 10^{-2} \qquad (1)$$

The correlation coefficient is 0.884 indicating a good fit to the data. The positive slope indicates that the gauge pressure increases the particle coating breakage.

In the following evaluation, P_i is taken to be 3.0 lb with a COV of 18.6 %. The standard normal distribution is also assumed. Then for a given breakage rate and the mean failure load of a particle, the estimated lower tail initial cracking load, F_i, on the particle is 1.107, 1.206, and 1.25 lb for the compacts A, B, and C, respectively. A regression analysis performed to correlate F_i and the molding pressure at the top of the head, P_0, yields the following relationship

$$F_i = 0.5447\ P_0^{\ 0.1344} \qquad (2)$$

with a correlation coefficient of 0.994.

As mentioned before, the whole fabrication process is in transient state. However, the Blake-Kozeny equation [5] for laminar flow through a packed wetted particle bed can be used to estimate the pressure force needed to press the matrix material into the void. A bounding analysis shows that a small fraction of the molding pressure (less than 17 psi) is consumed by viscous friction, and the bulk of it is resisted by the particle bed. In the following analysis the viscous friction will not be considered.

5. EQUILIBRIUM OF PACKED BED UNDER MECHANICAL LOAD

Now the equilibrium of a packed annular bed subjected to an external molding pressure of P_0 at the top through a metal piston is examined. Summing all of the forces in the vertical direction, leads to

$$AP_0 + \rho AL = AP_L + \int_0^L 2\pi\, F(\ell)\,(r_0 + r_i)\, d\ell \tag{3}$$

where A = area of the annular region $= \pi(r_0^2 - r_i^2)$
ρ = apparent density of the bed,
L = height of the bed,
P_L = pressure at the bottom of the bed,
F = frictional pressure between the bed and the sidewall,
r_0 = outer radius,
r_i = inner radius.

Let α be the friction coefficient between the particle and the sidewall,

$$F = \alpha\, P_H \tag{4}$$

where P_H is the radial (horizontal) pressure. The radial pressure is linearly related to the vertical pressure, P_V, by

$$P_H = KP_V \tag{5}$$

where values of α and K can be estimated from the relevant studies in the field of soil mechanics, to be 0.225 and 0.64, respectively, for the coated particle bed.
Substituting (5) and (4) into (3), dividing both sides by A, and rearranging to obtain

$$\frac{dp}{dL} = \rho - \frac{2\alpha K}{r_0 - r_i} P$$

The solution to this equation is

$$P = \frac{\rho(r_0 - r_i)}{2\alpha K}[1 - e^{-2\alpha K/(r_o - r_i)L]} + P_0 e^{-2\alpha K/(r_0 - r_i)L} \tag{6}$$

For the application to the particle bed, the first term on the right hand side is much smaller than the second term, hence it can be dropped.
For the full-size compact, for example, assuming a gauge pressure of 200 psi (compact A in Section 4) , or equivalently P_0 of 146.2 psi at the top of the bed. The pressure at the bottom of the mold is found to be 98.4 psi using equation (6).

6. APPLICATION OF THE WEIBULL THEORY

The two parameter Weibull theory is customarily used to describe the failure of the coated particles under a diametrally applied compressive load [2]. It is now used to assess the particle failure probability in both the full- and reduced-size compacts. According to the theory, the survival probability, S, is given by

$$\ln S = -\int_V a^m P^m \, dV = -a^m A \int_0^L P(\ell)^m d\ell \qquad (7)$$

where the average stress in an volume element is assumed to be proportional to the applied pressure, P, with a proportionality constant, a, which is essentially spatially independent. The pressure P is only a function of the axial distance ℓ.

The Weibull integrals (right hand side of equation 7) of the two compacts' stress distribution must be equal in order for two types of compacts to have the same survival probability. For a failure rate of 0.035%,

ln S	=	4.4×10^{13}	$(\text{psi})^6 \bullet \text{in}^3$	for the full-size compact
	=	$0.0266(P_0)^6$		for the reduced-size compact

which means that P_0 =343.8 psi, which is 2.35 times that for the annular compact. The pressure at the bottom of the bed for the cylindrical compact is 142.4 psi.

7. CONCLUSIONS

The various loads experienced by coated particles during compact manufacture have been defined. The magnitude of these loads and their impact on the probability of coating failure have been compared for the manufacture of full-size, annular compacts and reduced-size, cylindrical compacts. The following can be concluded from the results of these analyses:

(1) The principal loads on the particles during compact manufacture arise from the molding pressure.
(2) The pressure needed to inject matrix into the reduced-size cylindrical compacts is larger than that needed for the full-size annular compacts.
(3) The force acting on the particles within the bed decreases along the length of the bed from a maximum at the top to a lower value at the bottom. This load decreases more steeply with length for the reduced-size compacts than for the full-size compacts.
(4) For a given applied molding pressure, the failure probability in the reduced-size compact is lower than that in the full-size compact. The reduced-size compact and the full-size compact have equal failure probability when the molding pressure applied to the reduced-size compact is more than twice that applied to the full-size compact.

7. REFERENCES

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[3] Timoshenko, S., and J. N. Goodier, "Theory of Elasticity," McGraw-Hill, New York, New York, 1951.

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[5] Bird, R. B., et al., "Tranport Phenomena," John Wiley & Son, 1960.

Table 1. Crush strength of coated particles

	Batch A	Batch B	Batch C
No. of Measurements	23	50	50
Initial Cracking Load P_i, lb	3.10 (0.49)	3.03 (0.62)	3.00 (0.52)
Ultimate Load P_u, lb	5.24 (0.74)	5.45 (1.11)	5.88 (1.02)
Slope of P vs δ Curve (below P_i) lb/in	964 (168)	___	___

Note: Standard deviation is given in parenthesis.

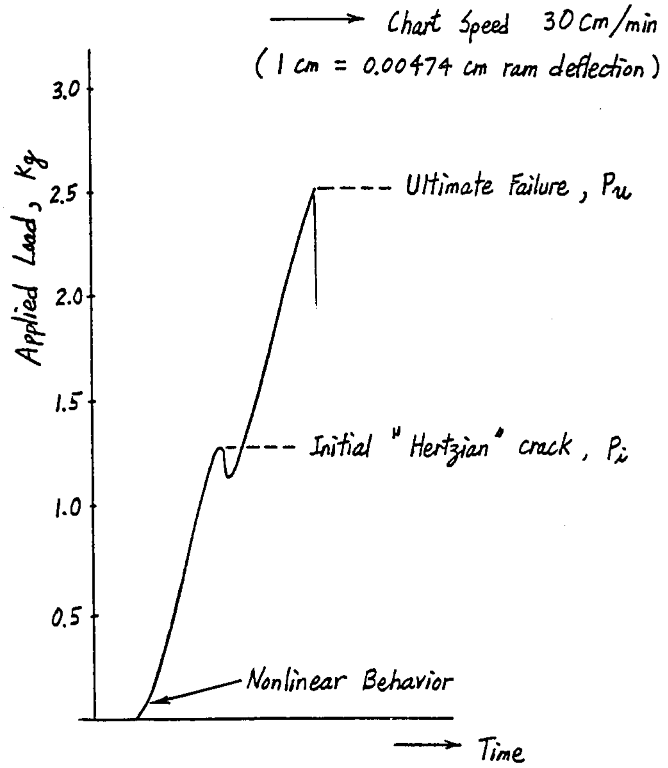

Fig. 1. A typical load deflection curve for a coated particle subjected to diametral compression