ORNL/TM-5132

Crushing Strength of HTGR Fuel Particles

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OAK RIDGE NATIONAL LABORATORY
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JANUARY 1976

OAK RIDGE NATIONAL LABORATORY
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W. J. Lackey, D. P. Stinton, L. E. Davis,[1] and R. L. Beatty

ABSTRACT

The whole-particle crushing strengths of High-Temperature Gas-Cooled Reactor fertile and fissile coated particles were measured and correlated with fabrication procedures. The crushing strength of Biso-coated fertile particles was increased by the the following factors: (1) Increasing the outer coating thickness by 10 μm increased strengths by 0.3 lb (1.3 N) for annealed particles and by 0.5 lb (2.2 N) for unannealed particles. (2) An 1800°C postcoating anneal increased strengths by 1 lb (4.4 N) for particles with thick outer coatings and by 2 lb (8.9 N) for particles having thin coatings. (3) Increasing the inner coating density by 0.1 g/cm^3 increased strength by 0.6 lb (2.7 N). The crushing strength of Triso-coated fissile particles was proportional to the thickness of the SiC coatings, and strength decreased on annealing by about 0.2 lb (0.9 N) when a porous plate was used to distribute the coating gas and by about 1.5 lb (6.7 N) when a conical gas distributor was used. The strengths of fertile and fissile coated particles as well as uncoated kernels appear adequate to allow fuel fabrication without excessive particle damage.

INTRODUCTION

Coating strength is likely an important property relative to the irradiation performance of High-Temperature Gas-Cooled Reactor (HTGR)[2] coated particles since the coating acts as a pressure vessel[3] containing

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[1]Co-op student from the University of Cincinnati.

[2]H. B. Stewart, R. C. Dahlbert, W. V. Goeddel, D. B. Trauger, P. R. Kasten, and A. L. Lotts, "Utilization of the Thorium Cycle in the HTGR," *Peaceful Uses of Atomic Energy, Proc. 4th Int. Conf. Geneva, 1971*, United Nations, New York, and International Atomic Energy Agency, Vienna, 4: 433–47 (1972).

[3]T. D. Gulden, C. L. Smith, D. P. Harmon, and W. W. Hudritsch, "The Mechanical Design of Triso-Coated Particle Fuels for the Large HTGR," *Nucl. Technol.* 16: 150–9 (1972).

fission gases. Further, during the fabrication of fissile and fertile particles into a usable fuel form several operations mechanically stress the individual particles. These processing operations are pneumatic transfer of bare and coated particles, fuel rod molding, and fuel rod carbonization. Since less than one defective particle per thousand is allowed, it is important that the particles remain intact during processing, and thus particle strength is of concern. In this work particle strength was measured by use of a simple crushing test. The influence of coating conditions and particle type on crushing strength are reported here for particles prepared in 2 1/2- and 5-in.-diam (6.4- and 12.7-cm) coating furnaces.

The measurement of crushing strength is fast and does not require complicated equipment, yet it can be used to determine the effect of coating process variables and particle design on particle strength. The test consists of crushing a particle between two planar surfaces and noting the load to initiate fracture. Such crushing strength values for about 40 particles per batch can be statistically treated, usually as members of a normally distributed population. The major disadvantage of the test is the difficulty of relating the crushing load to failure stress in the coating. Nevertheless, crushing strength has been shown to be very closely related to the fraction of particles that suffer broken coatings during penumatic particle transfer[4] and the rod fabrication process,[5] and thus crushing strength is a valuable measure of particle quality.

EXPERIMENTAL PROCEDURE

Particles to be crushed were obtained by riffling the entire batch of coated particles to ensure that representative particles were tested. The particles were then placed into a holder for crushing. The holder was a stainless steel plate drilled with 64 flat-bottom holes. Each hole was sized for one particle. A pin made from hardened steel drill rod was then placed into the hole over the first particle to be crushed. The pin had flat ends, a weight much less than the crushing strength of the particles, and a diameter sized to be a slip fit in the hole. The pin was used to transmit the force from the crosshead of an Instron[6] testing machine onto the particle to be crushed. The loaded holder was then placed on a load cell, the output of which was connected to a strip chart recorder. The recorder output was then zeroed to cancel the holder's weight. The screw driven crosshead was then brought down onto the top of the pin at 0.020 in./min (8.5 μm/sec) until the particle broke. Then the crosshead

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[4]Personal communication with J. E. Mack, August 1975.

[5]A. J. Caputo, "Fuel Element Assembly," *Gas-Cooled Reactor Programs Annu. Progr. Rep. Dec. 31, 1973,* ORNL-4975, pp. 45–53 (in press).

[6]Model TM-S-L, Instron Corporation, 2500 Washington St., Canton, Mass. 02021.

was returned to its original position, and the particle crushing strength was read from the strip chart. The recorder was recalibrated and rezeroed frequently, but drift was not a problem.

RESULTS AND DISCUSSION

Biso-Coated Particles

The Biso coating design consists of a porous inner carbon layer referred to as the buffer coating and a dense carbon layer referred to as the Low-Temperature Isotropic or LTI layer. Biso-coated ThO_2 is the reference fertile particle for the HTGR.

The thickness of the outer coating is one particle characteristic that was determined to strongly influence the strength of Biso-coated particles. Figure 1 shows the relationship between strength and LTI thickness for Biso-coated ThO_2 particles. Each point in the figure represents the average strength of 50 particles. A definite increase in average particle strength with an increase in average LTI thickness is observed. The coating conditions for this set of particle batches were deliberately varied over large ranges in an attempt to determine the influence of coating conditions on crushing strength. The ranges were as follows: coating temperature of 1225 to 1525°C, propylene (C_3H_6) flow rates of 1.44 to 5.76 ft^3/min (0.68–2.72 liters/sec), and charges of 0.5 to 2.5 kg. In some of the coating runs, the propylene was undiluted, while for other runs equal flows of propylene and helium were used. In each case coating was performed in a 5-in.-diam (12.7-cm) furnace using a 30°-included-angle cone as the gas distributor. A common large batch of buffer-coated ThO_2 having an average kernel diameter of 383 μm and a buffer coating thickness of 89 μm was used as substrate. Additional details of the experimental procedure have been reported.[7] Note in Fig. 1 that most of the batch-to-batch variation in crushing strength can be accounted for by differences in LTI thickness (i.e., the other variables, including whether or not a diluent was used, had a relatively small effect on crushing strength).

The influence of LTI thickness on crushing strength can also be seen in Fig. 2. Here as before the propylene coating gas flow rate was not always the same from one coating run to the next, and in some runs the gas was not diluted, while in others a helium diluent was used. While these variables have some influence on particle strength, as will be shown below, their effect was not large enough to mask the strong effect of LTI thickness. The curves in Fig. 2 are based on data for samples having buffer-coated ThO_2 as the substrate, unless otherwise noted. For some runs, the kernel diameter was 380 μm, while for others the diameter was 500 μm. Kernel diameter did not have a discernible influence on strength.

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[7]W. J. Lackey, W. H. Pechin, and J. D. Sease, "Measurement and Control of Shape of Fuel Particles for High Temperature Gas-Cooled Reactors," *Am. Ceram. Soc. Bull.* 54: 718–24 (1975).

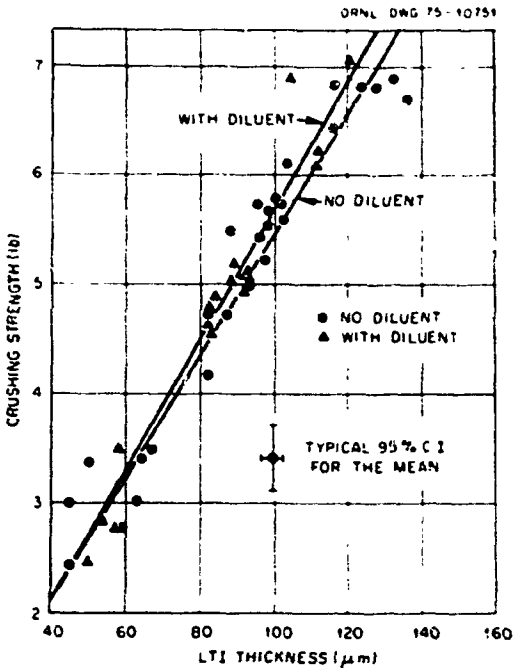

Fig. 1. Relationship Between Crushing Strength of Unannealed Biso-Coated Thoria and the Average LTI Thickness of a Batch of Particles. 1 lb force = 4.45 N.

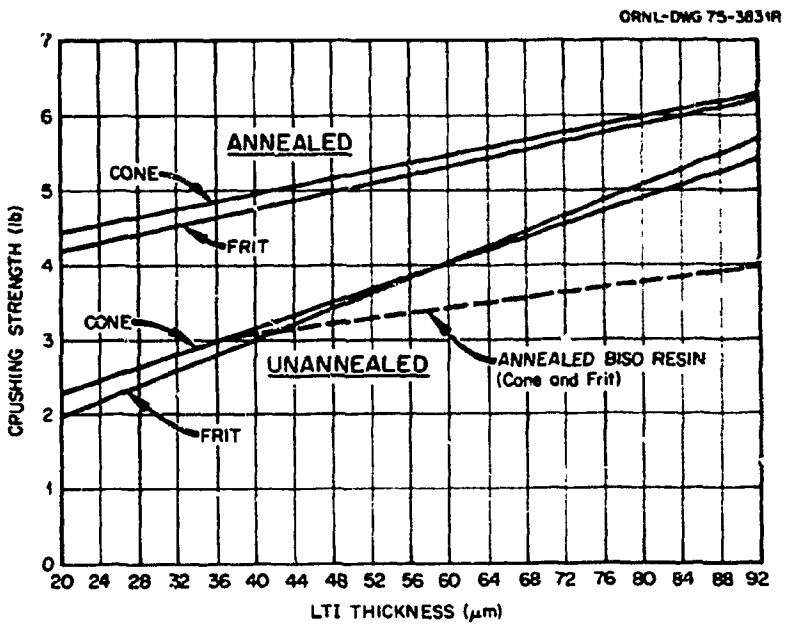

Fig. 2. Relationship Between Average Crushing Strength and LTI Thickness for Bisc-Coated ThO_2. Each line was obtained by least squares analysis using data from ten or more coating runs. 1 lb force ~ 4.45 N.

Figure 2 shows strength values for particles coated with a 30° conical distributor or with a specially contoured porous plate[7] gas distributor referred to as a frit. The scatter in the data is such that no statistically significant effect could be attributed to gas distributor type. The data do indicate that little if any real difference exists.

The data of Fig. 2 clearly show that annealing appreciably increases the strength of Biso-coated ThO_2. The annealing treatment used here was always 30 min at 1800°C in argon. The beneficial effect of annealing could be the result of removal of stresses built into the coating during the deposition process or could be the result of densification and the associated change in pore volume. The pore volume for the coatings described here typically decreases on annealing by about 20%. Note in Fig. 2 that for fertile particles having the reference LTI thickness of 75 μm the strength increases from about 4.8 to 5.8 lb (21.4–25.8 N) on annealing. Work by others[5] has repeatedly shown such an increase in strength to be a large improvement in the ability of particles to survive forces present during fuel rod fabrication. The fact that annealing increases particle crushing strength and the ability of particles to withstand the mechanical forces present during fuel rod fabrication is an important finding. Previous work[8] in which the strength of pyrocarbon deposited on disks was measured by three-point bending shows annealing to result in a decrease in strength. The earlier work was thus misleading as regards the influence of annealing on the ability of particles to survive the rod fabrication process.

The difference between Biso-coated uranium-bearing particles prepared from weak acid resin (process described in following section) and Biso-coated ThO_2 particles can also be seen in Fig. 2. Note that coated thoria particles are stronger than coated resin particles. This is evidence that particle crushing strength depends not solely on the nature of the coatings, but also on kernel properties.

A coating characteristic that appears to be significantly related to particle strength for a limited set of deposition conditions is LTI deposition rate. Figure 3 shows the relationship between deposition rate and strength for a series of batches of Biso-coated 500-μm-diam thoria. For each curve shown the deposition rate was varied by varying the coating gas flow rate. For low flow rates the thoria particles were diluted with low-density inert (carbon) particles to permit fluidization. The furnace diameter was 2 1/2 in. (6.4 cm) and the LTI coatings were deposited from MAPP[9] gas, either undiluted or diluted with argon. For each particle batch, the buffer and LTI layers each had a thickness of 85 ± 5 μm. For each temperature and hydrocarbon concentration the strength first increased as the deposition rate was increased, reached a maximum, and then decreased with further increase in deposition rate. Decreasing the concentration of the hydrocarbons in the coating gas by adding an inert diluent decreased both the peak coated particle strength and the deposition rate at which

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[8]J. L. Kaae, "The Effect of Annealing on the Microstructure and the Mechanical Properties of Poorly Crystalline Isotropic Pyrolytic Carbons," *Carbon* 10(6): 691–99 (1972).

[9]MAPP gas is marketed by AIRCO, Inc., and consists primarily of methylacetylene and propadiene with alkanes as stabilizers.

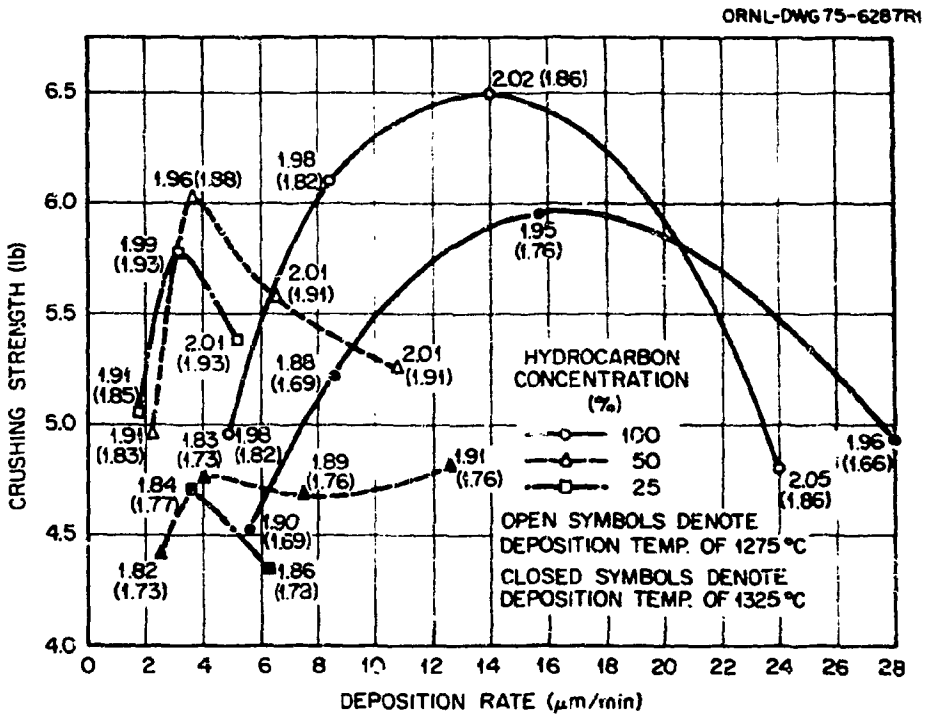

Fig. 3. Crushing Strength Versus LTI Deposition Rate for Biso-Coated ThO_2. The numbers accompanying data points are the observed and corrected LTI densities in g/cm^3. 1 lb force = 4.45 N.

it occurred. Figure 3 also shows that a decreased deposition temperature increases the strength of the particles.

A deposition rate correlation could be detected in these data because deposition rate ranged so widely and changes in the other variables were minimized. Since deposition rate itself is obviously not a material property, we tried to determine what structural attribute of the coating varied with deposition rate in a manner that would explain the relationship of strength with deposition rate. The most obvious assumption was that as the deposition rate varied, coatings having different densities were produced, and the supposition was that the strength varied according to the coating density. This was not the case, however, as can be seen by studying the trend in the LTI coating density values given alongside each data point in Fig. 3. The first value given is the observed LTI density as determined by the gradient liquid column technique, and the value given in parentheses is the corrected LTI density. Observed density refers to the density as conventionally measured. In this technique liquid infiltrates open pores, and the observed density value is larger than the actual bulk (geometric) density. The corrected density value is obtained by determining the fraction of the LTI coating volume that is open pores by use of 15,000-psi (103-MPa) mercury pycnometry and then using this value and the observed density to calculate the bulk coating density as follows: Corrected = (1 — volume fraction open pores) observed.

As the deposition rate increased the observed gradient density values continually increased even at the highest deposition rates. Consequently the downward trend of the crushing strength versus deposition rate curves for the higher deposition rates does not appear to be related to a change in observed coating density. The data of Fig. 3 also show that crushing strength is not correlated with corrected coating density. A previously reported[10,11] apparent correlation of strength and density for densities below about 1.85 g/cm^3 does not appear to be general. Thus, there does not appear to be a direct relationship between either deposition rate or density and particle strength. Some other unidentified coating property must be influencing strength. Krautwasser et al.[12] have also recently concluded that there is not in general a correlation between particle crushing strength and LTI density. Their work does show that crushing strength is closely correlated with the concentration of larger pores (50 to 500 Å diam), whereas the overall LTI density is mainly controlled by the concentration of pores smaller than 50 Å diam. That is, even though the larger pores are relatively few they act as stress risers and initiate fracture. This is a very reasonable explanation why crushing strength is not correlated with overall coating density.

A relationship of deposition rate to crushing strength was also observed for particles coated under a limited set of conditions in the 5-in.-diam (12.7-cm) furnace, as shown in Fig. 4. For these coating batches, the LTI thickness, while usually near 75 μm, sometimes deviated from that value. When the thickness was not 75 μm, the strength value was normalized to the value to be expected for a thickness of 75 μm by use of the appropriate curve in Fig. 2. Statistical analysis of the data of Fig. 4 revealed that one could state with 95% confidence that particle strength increased with deposition rate for both annealed and unannealed Biso-coated ThO_2. It is important to point out that for this set of data the coating temperature and quantity of particles in the furnace (charge) were essentially held constant from run to run, with the different deposition rates being obtained by variation of two variables: the coating gas concentration and flow rate. Four different coating gas conditions were employed. These in order of decreasing deposition rate were as follows: high propylene flow [3.6 ft^3/min (1.70 liters/sec)] without diluent, high propylene flow with an equal amount of helium, low propylene flow [2 ft^3/min (0.9 liters/sec)] without diluent, and low propylene flow with an equal amount of helium. The data convincingly show that when this method is used to vary the deposition rate, crushing strength is correlated with the rate of LTI deposition.

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[10]J. L. Kaae, "Structure and Mechanical Properties of Isotropic Pyrolytic Carbons Deposited Below 1600°C," *J. Nucl. Mater.* 38: 42–50 (1971).

[11]J. L. Kaae, "Relations Between the Structure and the Mechanical Properties of Fluidized-Bed Pyrolytic Carbons," *Carbon* 9: 291–99 (1971).

[12]P. Krautwasser, H. Nickel, and K. Täuber, "Influence of Microporosity on Fracture Stress of Pyrocarbon Coatings," paper C1/3 in *3rd International Conference on Structural Mechanics in Reactor Technology*, held in London, Sept. 1–5, 1975, Vol. 1, Part C, Comp. by T. A. Jaeger, Commission of the European Communities, Luxembourg, 1975.

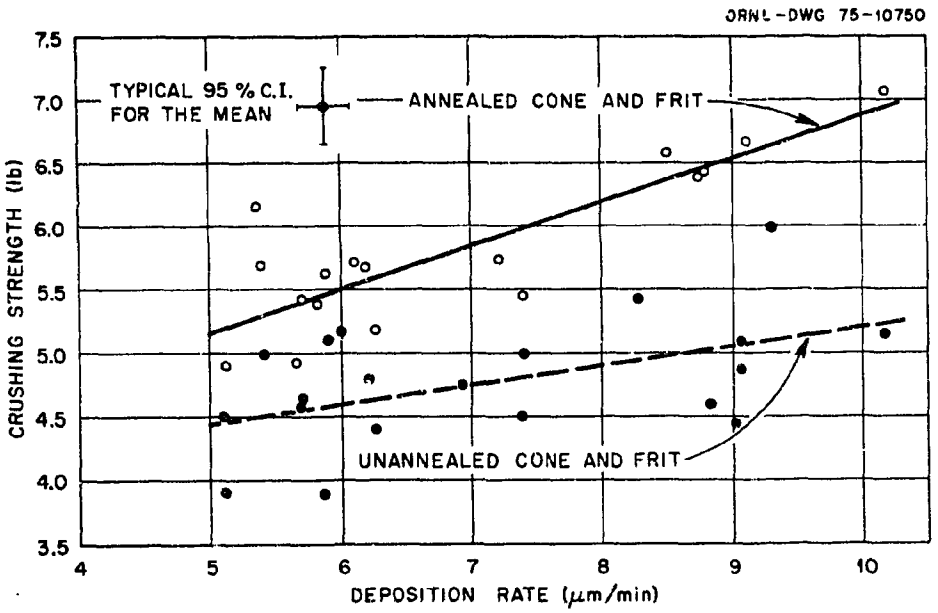

Fig. 4. Crushing Strength Versus LTI Deposition Rate for Biso-Coated ThO_2. 1 lb force = 4.45 N.

As before, crushing strength was not correlated with coating density. Typical properties for the above four coating conditions are given in Table 1 for annealed and unannealed particles. Note for the last three rows in the table that coating density is practically constant even though deposition rate and crushing strength vary. The correlation between crushing strength, LTI deposition rate, and coating thickness is graphically shown in Fig. 5. The strength contours shown in the figure were calculated from a multiple regression equation for annealed particles coated with the cone and frit. This method of presenting the data should be of value in predicting the strength of coating batches provided both the coating thickness and deposition rate are known and the deposition rate is varied only by the restricted set of variables stated.

For a broader range of coating conditions the correlation with deposition rate breaks down. In a later and separate carefully controlled experiment using the same coating furnace on 43 coating runs, strength and deposition rate did not correlate. In this experiment, in addition to the variations in coating gas concentration and flow rate, the deposition rate was also varied by altering the furnace charge or temperature. This is the same experiment that showed the good correlation with coating thickness in Fig. 1, and the ranges of experimental conditions have been described in conjunction with that correlation. It was clear from analysis of the data that the observed batch-to-batch variation in strength could not be explained solely by deposition rate. In other words, the manner by which a particular deposition rate was achieved was of considerable importance. For example, when a high deposition rate was obtained by use of a small charge of particles to the furnace, the resulting coating strength was not nearly as large as one would anticipate from Fig. 4.

Table 1. Properties of Annealed and Unannealed[a] Biso-Coated Particles for a Limited Set of Deposition Conditions

Gas Flow, cfm(liters/sec)		LTI Deposition Rate	LTI Coating Density[b], g/cm³		Crushing Strength[c]	
C_3H_6	He	(μm/min)	Observed	Corrected	(lb)	(N)
2 (0.9)	2 (0.9)	5.9	1.90 (1.88)	1.79 (1.78)	5.3 (4.8)	23.6 (21.4)
2 (0.9)	0	6.2	1.99 (1.96)	1.86 (1.83)	5.6 (4.6)	24.9 (20.5)
3.6 (1.7)	3.6 (1.7)	7.4	1.98 (1.94)	1.87 (1.84)	5.9 (5.0)	26.2 (22.2)
3.6 (1.7)	0	9.3	2.00 (1.95)	1.86 (1.81)	6.4 (5.5)	28.5 (24.5)

[a]Property values in parentheses are for unannealed particles.

[b]Observed density refers to the density as convenventionally measured by the gradient liquid column technique. Liquid infiltrates open pores and the measured density is larger than the actual bulk (geometric) density. Corrected density is obtained by determining the fraction of the coating volume that is open pores by use of mercury pycnometry and then using this value and the observed density to calculate the bulk coating density as follows: Corrected = (1 − Volume fraction open pores) observed.

[c]The 95% confidence intervals on the mean were 0.3 to 0.4 lb (1.3–1.8 N).

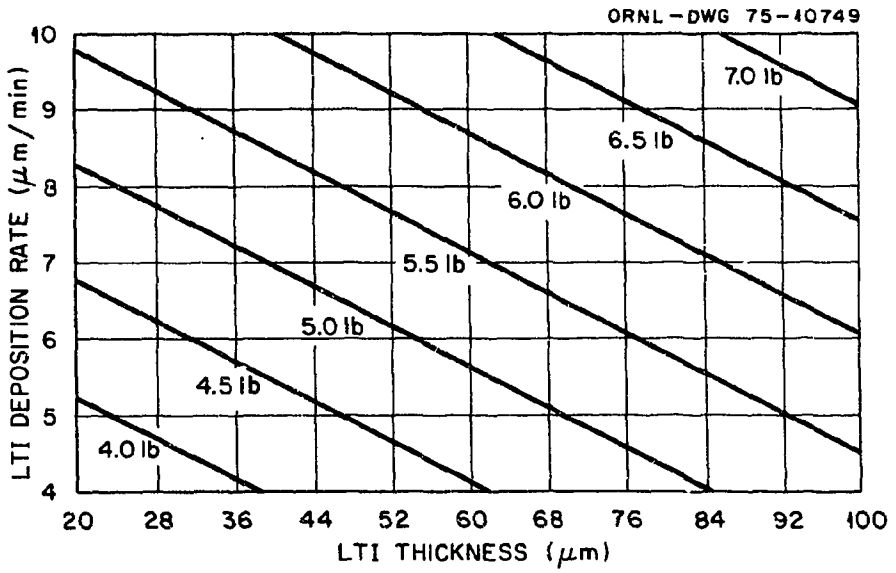

Fig. 5. Crushing Strength Contours for Annealed Biso-Coated ThO_2. Contours are for 17.8, 20.0, 22.2, 24.5, 26.7, 28.9, and 31.1 N.

In addition to correlating average batch strength with coating conditions as described above, the influence of individual particle dimensions on strength was investigated on particles from a single coating batch. Two Biso-coated thoria batches were investigated in this manner. Each particle used in the study was radiographed and separately measured to

obtain the dimensions of the kernel and each coating layer before determination of crushing strength. Figure 6 shows that the large within-batch variation in crushing strength [typical standard deviations are about 1 lb (4.4 N) for Biso-coated ThO_2] is not explained by within-batch variation in outer coating thickness, even though the average strength for a batch of particles correlated well with such thickness. The least squares fit shown in Fig. 6 does have a positive slope very similar to the curves of Fig. 2, but this explains only a small fraction of the within-batch variation in strength. Attempts to correlate individual particle strength values with other particle attributes such as buffer thickness, overall coating thickness, particle and kernel diameter, and particle shape were equally unsuccessful. Also unsuccessful was an attempt to correlate crushing strength with $b^2 - a^2$ where b and a are the outer and inner radii of the outer coating layer. It had been thought that such a correlation might exist since the stress S of the outer coating at rupture is reported[13] to be given by $S = F/\pi(b^2 - a^2)$, where F is the applied force. The multiple regression approach was also unsuccessful at explaining the major fraction of the within-batch variation in strength. Obviously some unknown factor is responsible for the observed strength variation. The variation could be simply the result of the inherent variability of the material. That is, the crushing strength depends on the size of a flaw that occurs within or near the region of high stress. Krautwasser's[12] work suggests that this is true.

Others[14] have found that particle strength depends on the density of the buffer coating. Our results confirm this finding, as can be seen in Fig. 7. The upper two curves in the figure are for annealed and unannealed Biso-coated ThO_2 having typical fertile particle coating thicknesses, while the bottom curve is for annealed Biso-coated ThO_2 but with coating thicknesses similar to those of Triso-coated fissile particles (i.e., 50-μm-thick buffer and 35-μm-thick LTI). In each case particle strength increases with buffer coating density, with the rate of increase being about 0.7 lb (3.1 N) for each 0.1 g/cm^3 increase in density. Buffer density values are for the buffer layer after deposition of the LTI layer and thus include the carbon that infiltrated the buffer layer during deposition of the LTI coating.

The buffer density varies from one particle to another over a range of about 0.1 g/cm^3 even for a given coating run.[15] Thus, one might speculate that the within-batch particle-to-particle variation in crushing strength shown in Fig. 6 was the result of differing buffer densities.

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[13]W. Delle, K. Prittler, G. Haag, and H. Schiffers, *Estimate of Stresses in the Coating of Compressed Coated Particles*, JÜL-569-RW, Institute of Reactor Materials, Jülich, Germany, January 1969; Scientific Translation Service, Ann Arbor, Michigan, Order No. 7510.

[14]Personal communication with J. Holder, C.E.N., Grenoble, France, April 24, 1975.

[15]W. J. Lackey et al., "Microsphere Coating," *Gas-Cooled Programs Annu. Progr. Rep. Dec. 31, 1972*, ORNL-4911, pp. 33–44.

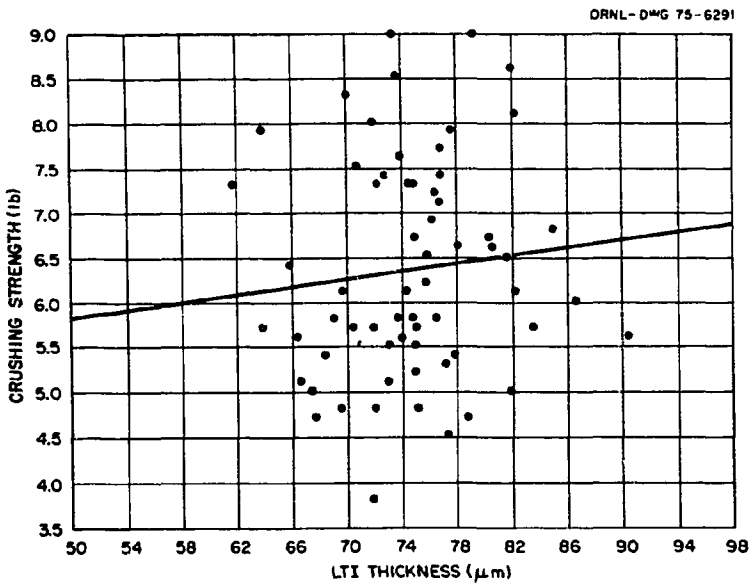

Fig. 6. Inability to Explain Particle-to-Particle Variation in Crushing Strength by Use of Within-Batch LTI Thickness Variation for Biso-Coated ThO_2.

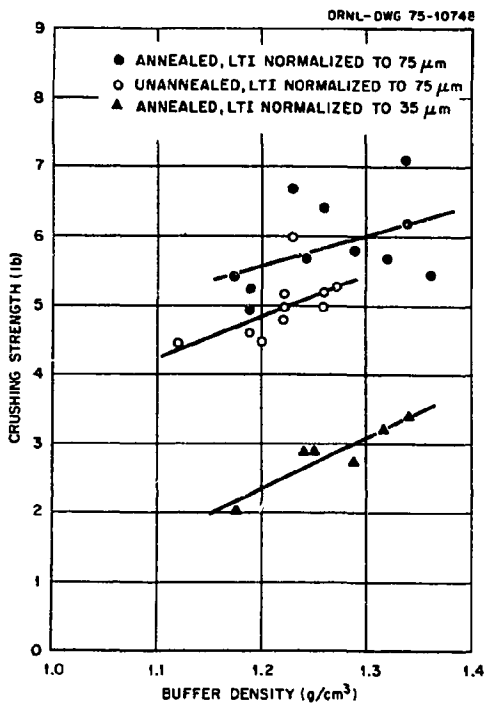

Fig. 7. Effect of Buffer Coating Density on Strength of Biso-Coated ThO_2. 1 lb force = 4.45 N.

However, this does not appear to be the case, since the observed within-batch variation in buffer density is considerably smaller than that required to account for all the within-batch variation in particle crushing strength.

Triso-Coated Particles

The Triso-coating design will be employed for the uranium-bearing fissile particles. This design consists, from the kernel outward, of a porous carbon layer, a dense carbon layer (inner LTI), an SiC layer, and a dense carbon layer (outer LTI). The reference fissile kernel is obtained by loading uranium onto an ion exchange resin[16] followed by heat treatment. The heat treatment consists of two steps; a low-temperature treatment termed carbonization and a high-temperature treatment termed conversion. After resin carbonization to 600 to 800°C kernels consists of a carbon matrix with small UO_2 inclusions. By heating in the range 1600 to 1800°C any desired fraction of UO_2 can be converted to UC_2 via the simplified reaction $UO_2 + 4C \rightarrow UC_2 + 2CO$. The carbonization and conversion processes are described further by Johnson et al.[17]

Particle strengths after the various steps involved in Triso coating are shown in Fig. 8. Note that at the inner LTI stage of processing the crushing strength is about 3 lb (13 N) and appears to depend on the extent to which the UO_2 is converted to UC_2 and also upon the LTI thickness. After coating with SiC there is less spread in the data, and the thickness of the SiC layer appears to account for the batch-to-batch variability in strength. Note that particle strengths actually decrease when the SiC layers are applied. Application of the outer LTI coating significantly enhances the particle strength. Interestingly, after the outer LTI coatings were applied the strongest and weakest batches were also the strongest and weakest at the SiC layer stage of processing, indicating that the strengths of the SiC layer influences the strength of the fully coated particle. Note that annealing of fully Triso-coated resin particles is not desirable since it results in a decrease in strength, which is particularly large for particles coated with the conical gas distributor.

The observed decrease in strength on annealing Triso-coated particles was surprising since annealing had increased the strength of carbon-coated particles. Since all particles are eventually annealed at 1800°C when the fuel rod is annealed, it was of interest to pursue the adverse effect of annealing on Triso-coated particle strength. Since the data of Fig. 8 indicated that seemingly small differences in the strength of the SiC layer appreciably influenced the strength of the fully coated Triso particle we decided to study the influence of various annealing treatments

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[16]P. A. Haas, *HTGR Fuel Development: Loading of Uranium on Carboxylic-Acid Cation Exchange Resins Using Solvent Extraction of Nitrate*, ORNL-TM-4955 (September 1975).

[17]D. R. Johnson, W. J. Lackey, and J. D. Sease, *The Effects of Processing Variables on HTGR Fuel Kernels Fabricated from Uranium-Loaded Cation-Exchange Resin*, ORNL-TM-4989 (August 1975).

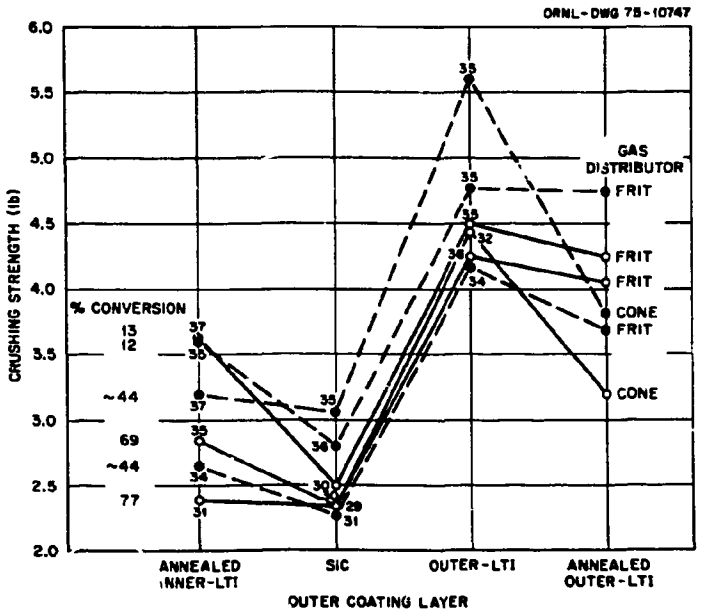

Fig. 8. Change in Particle Crushing Strength During Fabrication of Triso-Coated Amberlite Resin. Numbers by the points give the layer thicknesses in micrometers. 1 lb force = 4.45 N.

on the strength of particles coated only through the SiC layer. The study included 12 SiC coated particle batches. Variables of interest were annealing temperature from 1630 to 2000°C, annealing times from 12 to 420 min, details of the deposition process such as temperature and rate, coating furnace size [2.5 vs 5 in. (6.4 vs 12.7 cm)], cone or frit gas distributor, and resin or ThO_2 kernels. Results clearly showed that the crushing strength of each particle batch decreased on annealing, and the decrease was larger for higher annealing temperatures. Annealing time had only a very small influence. The decrease in strength was larger for particles coated with the conical gas distributor. Typically, the strength decrease caused by annealing at 1800°C was about 0.5 lb (2.2 N) for particles coated with the cone and about 0.2 lb (0.9 N) when the frit gas distributor was used. Even the smaller value is statistically significant since 95% confidence intervals for the mean strength were less than 0.1 lb (0.4 N).

From the standpoint of cracking coatings during penumatic particle transfer or other handling operations, particles having been processed only through SiC coating appear most fragile from the data of Fig. 8. It is noteworthy that complete postcoating characterization can probably be performed without removing the particles from the furnace at this stage of processing, so this potential handling problem can likely be avoided.

To date, only a limited effort has been directed toward altering the SiC coating process to produce stronger coatings. The available data indicate that crushing strength of unannealed particles decreased about 0.3 lb (1.3 N) when the deposition temperature was increased from 1600 to

1700°C. The grain size of the SiC was larger for the higher deposition temperature, which probably is the reason for the reduction in strength. Crushing strength appeared to increase on increasing the SiC deposition rate from 0.1 to 0.2 μm/min, but this observation is based on limited data.

There is considerable particle-to-particle variation in crushing strength for a given batch of Triso-coated particles. Typically, the within-batch standard deviation is 0.5 lb (2.2 N). Attempts to correlate individual particle strength with other particle attributes in a manner like that previously described for Biso-coated particles were rather unsuccessful. Outer coating and SiC coating thickness variations explained only a small fraction of the crushing strength variation.

Kernels

Up to this point only coated particles have been discussed. However, kernel strength may be important for at least two reasons. First, coated-particle strength may be correlated with kernel strength; such a correlation for Biso-coated particles is indicated in Fig. 8 by the conversion percentages, which determine kernel strength. Also, strong kernels are desired to minimize breakage during transport and other handling operations.

The kernels for the fertile particles consist of ThO_2 prepared by the sol-gel process.[18] Such kernels are very spherical and dense and appear almost glasslike.[19] Their crushing strength is about 18 lb (80 N), and no damage has been observed during numerous pneumatic transfer and other handling operations.

The reference kernel for fissile particles is obtained by carbonizing and converting uranium-loaded resin microspheres. The crushing strengths for 13 carbonized batches of resin-derived kernels have been measured. The resin source for 10 of the batches was Amberlite[20] while the other three batches were derived from Duolite[21] resin. The crushing strengths for the Amberlite batches varied from 3.4 to 6.5 lb (15–29 N), with the average being 5.2 lb (23 N). Two Duolite batches had strengths of 3.1 and 2.8 lb (13.8 and 12.5 N), which are less than those of any of the Amberlite batches. A third Duolite batch, for which the uranium was loaded onto the resin by an alternate chemical flowsheet, exhibited a strength of only 1.0 lb (4.4 N). In the case of the Amberlite material the batch-to-batch variation in strength was caused by differing uranium

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[18]P. A. Haas, *Process Requirements for Preparing ThO_2 Spheres by the ORNL-Sol-Gel Process*, ORNL-TM-3978 (December 1972).

[19]P. A. Haas and W. J. Lackey, *Improved Size Uniformity of Sol-Gel Spheres by Imposing a Vibration on the Sol in Dispersion Nozzles*, ORNL-TM-4094 (May 1973).

[20]Amberlite IRC-72, Rohm and Haas.

[21]Duolite C-464, Diamond Shamrock.

and moisture contents of the resin at the beginning of the carbonization process. These variables are correlated with strength in Fig. 9. The contour lines in the figure were obtained by multiple regression analysis, which showed at the 99% level of confidence that both uranium and moisture contents influenced strength.

The above data were for carbonized material for which the maximum processing temperature was 800°C. On heating this material in the range 1600 to 1800°C some of the UO_2 is converted to UC_2. Strength measurements were made for such converted kernels. As shown in Fig. 10, crushing strength and percent conversion correlate; the more nearly complete the conversion reaction, the weaker the kernels. Because of the presence of finely divided UO_2 or UC_2 both the carbonized and converted kernels oxidize or hydrolize rapidly on exposure to air, and for this reason kernels are kept under argon during fabrication. However, the apparatus for measuring crushing strength was not in an inert-atmosphere glove box, and thus it was expedient to expose the kernels to air for up to 30 min while their strengths were being determined. Thus oxidation or hydrolysis could have altered kernel strength. To determine the extent of any such changes in strength the apparatus was enlcosed on several occasions with a plastic bag equipped with gloves and purged with argon. Measurements made under argon gave very similar results for carbonized as well as converted kernels, giving confidence that exposure to air did not compromise the validity of the kernel strength data.

Carbonized particles appear sufficiently strong to withstand particle transfer operations without damage, and pneumatic particle transfer of carbonized resin has been accomplished many times with no detectable damage. Converted kernels are weaker, and thus some particle cracking could conceivably occur during handling. Converted kernels ranging from 15 to 75% conversion have been pneumatically transferred into the coating furnace many times, and extensive data show that the vast majority and perhaps all of the particles do not crack. More detailed examination is in progress to determine if any minor deterioration such as abrasion or cracking of say 1 particle in 10^4 occurs during handling operations.

CONCLUSIONS

1. The crushing strength of annealed and unannealed Biso-coated ThO_2 particles was increased greatly with increasing outer coating thickness.
2. The strength of Biso-coated ThO_2 was increased by annealing the coated particles at 1800°C and by increasing the inner coating density.
3. There was little difference between the strength of particles coated with conical or porous plate gas distributors with the exception that the strength of Triso-coated particles decreased more on annealing where the coatings had been deposited with the conical gas distributor.
4. Within-batch particle-to-particle variation in strength for fertile and fissile coated particles could not be accounted for by indivdual geometrical particle attributes such as coating thicknesses, particle diameter, and particle shape.
5. Carbonized resin kernels are strong and have strengths averaging 5 lb (22 N). Their strength depends on both uranium and moisture content.

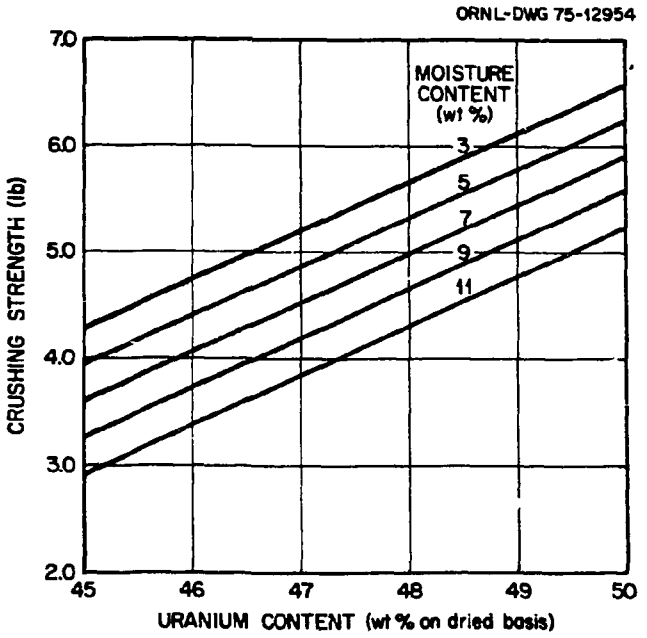

Fig. 9. Influence of Uranium and Moisture Contents on the Strength of Carbonized Resin Kernels. 1 lb force = 4.45 N.

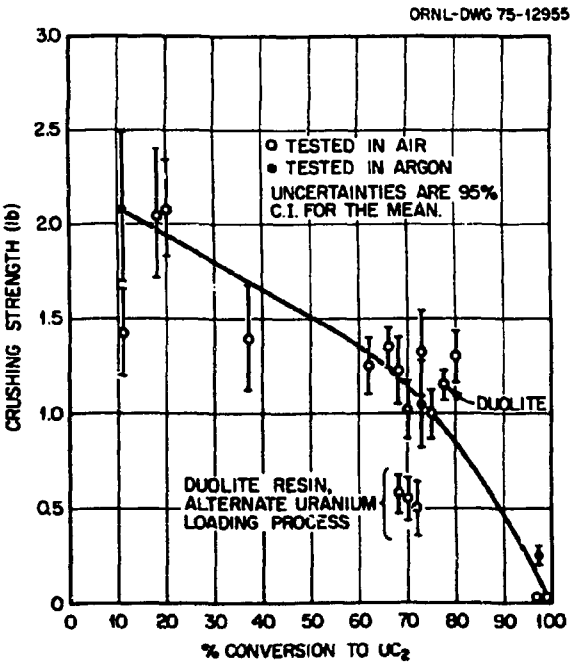

Fig. 10. Influence on Crushing Strength of the Extent to which the UO_2 of Resin-Derived Kernels is Converted to UC_2. Data are for Amberlite resin unless otherwise noted. 1 lb force = 4.45 N.

6. The strength of converted kernels decreases from about 2 lb (9 N) for particles having 15% of the oxide converted to carbide to about 1 lb (4 N) for 75% conversion.

7. The strengths of fertile and fissile coated particles as well as uncoated kernels appear adequate to allow fuel to be fabricated without excessive particle damage provided that care is taken to produce high-quality particles. The simple crushing test employed here is very useful in determining particle quality.

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