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COMPACTION OF AWBA FUEL PELLETS WITHOUT BINDERS

(AWBA Development Program)

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AUGUST 1982

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PITTSBURGH, PENNSYLVANIA 15122-0079
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(AWBA Development Program)

R.G.R. Johnson

Contract DE-AC11-76PN00014

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FOREWORD

The Shippingport Atomic Power Station located in Shippingport, Pennsylvania was the first large-scale, central-station nuclear power plant in the United States and the first plant of such size in the world operated solely to produce electric power. This program was started in 1953 to confirm the practical application of nuclear power for large-scale electric power generation. It has provided much of the technology being used for design and operation of the commercial, central-station nuclear power plants now in use.

Subsequent to development and successful operation of the Pressurized Water Reactor in the Atomic Energy Commission (now Department of Energy, DOE) owned reactor plant at the Shippingport Atomic Power Station, the Atomic Energy Commission in 1965 undertook a research and development program to design and build a Light Water Breeder Reactor core for operation in the Shippingport Station.

The objective of the Light Water Breeder Reactor (LWBR) program has been to develop a technology that would significantly improve the utilization of the nation's nuclear fuel resources employing the well-established water reactor technology. To achieve this objective, work has been directed toward analysis, design, component tests, and fabrication of a water-cooled, thorium oxide-uranium oxide fuel cycle breeder reactor for installation and operation at the Shippingport Station. The LWBR core started operation in the Shippingport Station in the Fall of 1977 and will finish routine power operation on October 1, 1982. After End-of-Life core testing, the core will be removed and the spent fuel shipped to the Naval Reactors Expended Core Facility for detailed examination to verify core performance including an evaluation of breeding characteristics.

In 1976, with fabrication of the Shippingport LWBR core nearing completion, the Energy Research and Development Administration, now DOE, established the Advanced Water Breeder Applications (AWBA) program to develop and disseminate technical information which would assist U. S. industry in evaluating the LWBR concept for commercial-scale applications. The AWBA program, which is concluding in September, 1982, has explored some of the problems that would be faced by industry in adopting technology confirmed in the LWBR program. Information already developed includes concepts for commercial-scale prebreeder cores which would produce uranium-233 for light water breeder cores while producing electric power, improvements for breeder cores based on the technology developed to fabricate and operate the Shippingport LWBR core, and other information and technology to aid in evaluating commercial-scale application of the LWBR concept.

All three development programs (Pressurized Water Reactor, Light Water Breeder Reactor, and Advanced Water Breeder Applications) have been conducted under the technical direction of the Office of the Deputy Assistant Secretary for Naval Reactors of DOE.

Technical information developed under the Shippingport, LWBR, and AWBA programs has been and will continue to be published in technical memoranda, one of which is this present report.

ACKNOWLEDGEMENTS

I would like to acknowledge the invaluable assistance rendered by Mr. George White of Battelle Northwest regarding the techniques used by commercial fuel manufacturers in making fuel pellets without binders. I would also like to acknowledge the contribution made by Mr. Rich Hudson of the Mallinckrodt Co. who supplied the stearates used in this study.

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Highly active oxide fuel powders, composed of UO_2, UO_2-ThO_2, or ThO_2, were compacted into ultra-high density pellets without the use of binders. The objective of the study was to select the optimum die lubricant for compacting these powders into pellets in preparation for sintering to densities in excess of 97% Theoretical Density. The results showed that sintered density was a function of both the lubricant bulk density and concentration with the lowest bulk density lubricant giving the highest sintered densities with a lubricant concentration of 0.1 weight percent. Five calcium and zinc stearates were evaluated with a calcium stearate with a 15 lb/ft^3 bulk density being the best lubricant.

COMPACTION OF AWBA FUEL PELLETS WITHOUT BINDERS
(AWBA Development Program)

I. INTRODUCTION

Fuel pellets for the Light Water Breeder Reactor (LWBR) and Advanced Water Breeder Reactor (AWBA) programs have traditionally been fabricated with organic binders and die lubricants. This was necessary to achieve high densities and low porosity microstructures. Pellet densities in excess of 97% of theoretical were desired to maximize the fertile (ThO_2) fuel loading and to achieve dimensionally stable pellets.

Since commercial reactors operate with lower density pellets, commercial UO_2 pellets can be made without binders using a binderless precompaction technique known as slugging in conjunction with a highly efficient die lubricant. The study described in this report was undertaken to determine if the highly active LWBR/AWBA powders could be fabricated into fuel pellets using the commercial fabrication techniques without degrading the pellet density requirements.

II. BACKGROUND

Highly active powders were required for the LWBR/AWBA programs to reproducibly fabricate high density pellets. These powders were made highly active (activated) by milling in a high pressure air mill known as a micronizer (Reference (a)). This milling increases the surface area and lowers the bulk density of the powder. The lower bulk density reduces both flowability and compactibility. In the past, binders have always been needed to overcome these characteristics of activated powders and permit the compaction of high density pellets. The binder counteracts the effects of the milling operation by (1) increasing the bulk density of the material which improves compactibility, (2) increasing the flow characteristics which improves die filling, and (3) supplying the agent for welding the particles together.

Two different processing techniques were used to incorporate the binder which was a polyethylene glycol known as Carbowax 6000. These techniques were the liquid/solids and wet agglomeration processes and are described in detail in Appendix I.

The microstructures of LWBR and AWBA pellets were highly dependent upon the agglomerating techniques. Pores in wet agglomerated pellets were small and uniformly dispersed. Larger randomly dispersed pores were normally observed in pellets manufactured by the liquid/solids process, where the pores were formed by the dry blended die lubricant. If the agglomeration was not carefully controlled, high density press feed granules would be produced, which would not crush during compaction. As a result, the pellet microstructures would show the outline of the original granules delineated by interconnected pores. These interconnected pores represented undesireable zones of mechanical weakness and decreased thermal conductivity.

The objective of this study then was to provide a simpler process for making structurally sound, high density fuel pellets. The modified process would eliminate the operations and equipment for agglomerating with binders and for then removing the binders from the pressed pellets prior to the sintering operation.

III. EQUIPMENT AND MATERIAL

Certain specialized equipment essential to the fabrication of high density binderless pellets are described in this section. These include:

1) Micronizer - A Jet Pulverizer model with a 4 inch diameter grinding chamber. Operates with high pressure (100 psi) low humidity air.

2) Turbula Blender - A rotary blender with an adjustable cage capable of accommodating a wide variety of container sizes. Blender has three operating speeds controlled by pulley wheels and a belt. Blender operates with a gentle motion that does not harm the fragile powder granules.

3) Compacting Press - A 45 ton double action Dorst press. Dies were cylindrical and made from tool steel with a tungsten carbide-lined cylindrical cavity. Punches were cylindrical with tungsten carbide shanks mounted in tool steel bases, that were made to fit the appropriate die cavity with 0.001 inch maximum clearance.

4) Sintering Furnace - A cold wall controlled atmosphere furnace made by Vacuum Industries Div. of the GCA Corp., Summerville, Mass. The hot zone was lined with tungsten sheet and heated with tungsten elements. Maximum operating temperature was 2000°C in a flowing hydrogen atmosphere (50 CFH). Heating and cooling rates and soaking time and temperature were controlled by a microprocessor. Heating and cooling rates were controlled at 115°C/hr.

The materials used in this study consisted of fuel powders and organic lubricants. The fuel powders included oxalate precipitated ThO_2 activated by 2 pass micronizing, UO_2-ThO_2 made by 2 pass comicronizing U^eO_2 made by the Oak Ridge National Laboratory (ORNL) with ThO_2, highly enriched UO_2 made by 2 pass comicronizing ORNL U^eO_2 with U^dO_2 supplied by the Columbia plant of Westinghouse, and low enriched reactor grade U^eO_2 supplied by the Babcock and Wilcox Co. The other materials evaluated were the organic lubricants themselves. These were the stearates of calcium and zinc, which are metallic soaps widely used as lubricants and mold release agents in industry. Physical appearance and bulk densities of the stearates evaluated appear in Table I. The stearates were supplied by the Chemical Division of the Mallinckrodt Co.

IV. EXPERIMENTAL PROCEDURE

These separate fuel systems (UO_2, UO_2-ThO_2 and ThO_2) were evaluated for binderless pressing performance using 6 die lubricants at two primary concentration levels. Performance, which was evaluated over a range of pressed densities, was measured by the sintered pellet densities and by the integrity of the pellet microstructures. Also evaluated was the response of the powder with and without fines (-80 mesh material).

The process outline for binderless pressing is shown in Figure 1 where it is identified as the AWBA dry process. The as-received oxide fuel powders were initially blended together in a 4 quart V-blender in an operation called primary blending. Its purpose was to provide a homogeneous mixture of the two oxides when making a binary U^eO_2-ThO_2 composition or when diluting fully enriched (93.14% U-235) U^eO_2 with U^dO_2. Fully enriched U^eO_2 was never compacted in its virgin form but was diluted with U^dO_2 to provide a U^{235} enrichment between 9% and 33% for irradiation test loadings. Primary blending was not necessary when processing a single oxide system such as ThO_2 or low enriched commercial grade U^eO_2.

After blending, the powder was activated by micronizing. Each batch of powder was micronized twice to provide a powder with a surface area in excess of 5.0 sq. meters/gram (m^2/gm) as measured on a Micromeretics Model 2200 Surface Area Analyzer. The actual powder surface areas are listed in Table I. Experience has shown that two micronizing passes are necessary to obtain chemically homogeneous double oxide pellets with densities in excess of 97% of theoretical density (TD) (Reference (a)). To investigate the effect of micronizing upon powder compaction and sinterability when not using binders, one half of the 11.4% U^eO_2-ThO_2 batch was separated after the first micronizing pass and held for compaction without any further activation.

Since the commercial UO_2 powder is compacted into pellets without the use of binders, it was used as a control in this study against which the compaction behavior of the AWBA fuel powders was measured. Since this powder was to serve as a standard in the as-received condition, no micronizing activation was performed.

After micronizing (or in the unmicronized state for the commercial UO_2), the powder was slugged without organic additives into 1/2 inch thick 40% dense compacts (slugs) in a 1.25 inch diameter die. After ejection from the die, if the slugs were soft and friable, they were crushed by hand on the die table and reslugged, which substantially improved the green strength. All ThO_2 and UO_2-ThO_2 powder was double slugged. The UO_2 powders were more compactible and were sufficiently strong after a single slugging to be granulated.

TABLE I. PHYSICAL CHARACTERISTICS OF THE FUEL POWDERS AND THE LUBRICANTS AND A PELLET DENSITY TABULATION

A. Powder Surface Areas

1.	11.4% U^eO_2-ThO_2 - 1 pass	7.08 m^2/gram
2.	11.4% U^eO_2-ThO_2 - 2 pass	8.10 m^2/gram
3.	ThO_2	8.68 m^2/gram
4.	U^eO_2 - 16.7% U^{235}	5.12 m^2/gram
5.	U^eO_2 - 2.8% U^{235}	3.05 m^2/gram

B. Lubricant Properties

Identity	Type	Form	Bulk Density (lb/ft^3)
1. CA-38	Calcium Stearate	Granular	38
2. CA-22	Calcium Stearate	Fine Powder	22
3. CA-15	Calcium Stearate	Fine Powder	15
4. ZN-23	Zinc Stearate	Fine Powder	23
5. ZN-16	Zinc Stearate	Fine Powder	16
6. Sterotex	Glyceryl Ester of Stearic Acid	Fine Powder	37

C. Maximum Sintered Fuel Pellet Density Achieved with Each Lubricant (% TD)

	CA-38	CA-22	CA-15	ZN-23	ZN-16	Sterotex
1. 11.4% U^eO_2-ThO_2-1 pass	95.0	95.3	95.5	95.0	95.3	94.9
2. 11.4% U^eO_2-ThO_2-2 pass	96.7	-	97.3	-	97.0	-
3. ThO_2	97.0	-	98.0	-	97.7	-
4. U^eO_2-16.7% U^{235}	98.0	-	98.5	-	98.3	-
5. U^eO_2-2.8% U^{235}	-	-	97.3	-	96.7	-

The individual slugs (which weighed about 50 grams) were hand granulated through a 16 mesh sieve. The granulated powder batch was split in half, and the -80 mesh fines were removed from one of the two batches. The fines in each of the divided batches ranged between 16 and 26% of the batch weight. Each granulation condition was compacted into pellets to determine the effect of the fines upon compactibility. A flow diagram showing the experimental design used to evaluate each of the fuel systems is shown in Figure 2.

The die lubricants were added directly to the granulated powder. Two lubricant levels were initially tested: 0.1% and 0.25%, expressed as a weight percentage of the powder charge. After adding the lubricant in the prescribed amount to the powder, the individual containers were placed on a Turbula blender for blending (Figure 22). The blender was run at its slowest speed for one minute for all types of powder. A further study reported later in this report (Section V.2(a)) evaluated the effect of increasing the blending time to 2 and 4 minutes and introducing new lubricant addition levels of 0.05 and 0.15% using ThO_2 powder.

Press feeds representing the three fuel systems were compacted into right circular cylindrical pellets in carbide lined dies with die diameters of 0.307 or 0.383 inch. All pellets were compacted using double action pressing over as wide a range of pressed densities as could be achieved starting at 56% TD. Upper limits varied between 62 and 65% TD depending upon the fuel system being pressed. The upper limit was reached when pellets began to crack or break during pressing.

After compaction, all pellets were sintered in hydrogen with a dewpoint of -60°F at 1800°C for 12 hours in a cold wall tungsten lined batch furnace. Pellets were inspected for geometric dimensions, weights, physical appearance and yields. The geometric dimensions and weights were used to calculate individual pellet densities. Representative pellets were selected from the density series for metallographic evaluation.

V. EXPERIMENTAL RESULTS

1. UO_2-THO_2 SYSTEM

Figure 2 shows the experimental design that would permit each of the 6 lubricants (the 5 metallic stearates supplied by the Chemical Division of the Mallinckrodt Co. and Sterotex) to be tested with each attribute of interest using the 1 pass activated powder from the UO_2-ThO_2 system. After these pellets had been evaluated, the variables were reduced to include only the most promising lubricants and the most active of the micronized powder. The UO_2-ThO_2 system was chosen for the initial work since this was the fuel system used in the LWBR Core.

The sinterability curves (Figure 3) for the fully activated U^eO_2-ThO_2 powder (two micronizing passes), which used the lowest bulk density calcium stearate (15 lb/ft^3) tested, indicate excellent compactibility over a wide pressed density range with no cracks or laminations observed until the pressed density exceeded 63%. The highest sintered densities, which exceeded the 97% target over the entire pressed density range, were obtained using 0.1% calcium stearate. Powder compactibility was not affected by the -80 mesh fines at the 0.1% addition level as can be seen by the similarity of the curves for the -16/+80 and the -16 mesh material. Increasing the lubricant level to 0.25% lowered the sintered density about 1%. Retention of the -80 mesh fines at the 0.25% additive level also tended to have a negative effect upon the densification. These same conditions were tested using the other lubricants in each of the other fuel systems (UO_2 and ThO_2) with similar results, i.e., the 0.1% addition yielding the highest sintered densities with or without the presence of the fines. Owing to the similarity of the results with those seen in Figure 3, the sinterability curves representing conditions other than the 0.1% concentration level using the -16/+80 powder fraction are not shown for the other fuel types.

Each micronizing pass activates the powder by increasing the surface area. One micronizing pass does not sufficiently activate the powder to allow densification to 97% TD. The curves in Figures 4 and 5 clearly show the increase in powder activity imparted by the second micronizing pass. These curves also show that the ability of pellets to reach a high density is a function of the lubricant bulk density, i.e., the lower the lubricant bulk density, the higher the sintered pellet density.

Sterotex, when used as a dry lubricant without a binder, clearly does not have good lubricating properties (Figure 5). The number of crack free pellets that could be compacted at each green density was so low with the one pass micronized powder that Sterotex was not even tested with two pass powder. However, Sterotex always provided adequate die lubrication when used in powder in conjunction with a binder as can be seen in Figure 14, where the Sterotex was added in dry form to the granules after agglomeration in the LWBR process or was disolved in the oxylene solution during agglomeration in the process which produced the AWBA-1 pellets.

The calcium stearate with a bulk density of 22 lbs/ft^3 (CA-22) and the zinc stearate with a 23 lb/ft^3 density (ZN-23) were eliminated from further trials since the lower bulk density stearates were better performers as can be seen in Figures 4 and 5. The very high density calcium stearate identified as CA-38 was used throughout the study due to its high flowability. All the stearates except for this one were fluffy and had poor flowability. CA-38 was a free flowing granular lubricant with no tendency toward clumping. The superior flow characteristics were sufficient reason to include this lubricant in the remainder of the test.

(a) Metallographic Results

Photographs of representative pellet microstructures of UO_2-ThO_2 pellets made with CA-15 are shown in Figure 15. The photographs show that the original powder granules are retained in the pellet structure. The granules, which have been flattened but not crushed during compaction, are outlined by fine pores. The concentration of the pores increases as the lubricant content increases from 0.1% to 0.25%. These pellet structures were all acceptable for use in AWBA irradiation tests.

Photographs of the microstructures of UO_2-ThO_2 pellets made with the CA-38 and ZN-16 lubricants are shown in Figure 16. The structure of the pellets made with the ZN-16 lubricant is quite similar to that achieved with the CA-15 lubricant. The CA-38 produced lower density pellets containing discrete spherical pores larger than the pores produced by the other lubricants.

2. ThO_2 System

The thoria powder also exhibited good compactibility when evaluated with these three lubricants (ZN-16, CA-15, and CA-38) with both CA-15 and ZN-16 producing acceptable results at the upper end of the density range. Sintered densities of 98% were obtained at the upper end of the curve with CA-15 as can be seen in Figure 6. Microstructures of these pellets are seen in Figure 17 and are quite similar to the structure of equivalent UO_2-ThO_2 pellets except that the discrete pores produced by CA-38 are now interconnected by a network of fine but acceptable cracks.

(a) Relation Between Sintered Density, Lubricant Concentration and Blending Time

Lubricant levels (using CA-15) of 0.05, 0.10, and 0.15% were examined with blending times of 1, 2, and 4 minutes. Approximately equivalent density results were obtained for the 0.10 and 0.15% additions. Results were erratic for the 0.05% addition as apparently there is insufficient lubricant present for a uniform dispersion. The results are shown in Figures 7, 8, and 9. There is also a slight dependency upon blending time especially at the 0.10% addition level where densities increased as a function of blending time. The photomicrographs in Figure 18 show that the intragranular porosity increases directly with increasing lubricant concentration, but there are no visible changes in porosity distribution for increases in blending time.

(b) Large Diameter Thoria Pellets

Large diameter annular thoria pellets with a 1.033 inch outside diameter (OD) and a 0.538 inch inside diameter (ID) were also made. These pellets were compacted from a bindered powder in a die with a 1.25 inch OD using a 0.635 inch core rod to form the ID. These pellets were compacted to a length of 0.625 inch. A test series of binderless thoria pellets were compacted using this tooling to further examine the compactibility of the binderless powders. One tenth percent of the zinc stearate identified as ZN-16 was blended with -16 mesh granules of slugged thoria powder. The densification curve for these pellets is shown in Figure 10. The powder compacted well at pressed densities up to 60% TD although a springloaded upper punch to maintain topside pressure on the pellet during ejection from the die was required at 60% green density to suppress crack formation. The slope of the sinterability curve in Figure 10 indicates that sintered densities might have exceeded 97% had the powder been compacted at higher green densities, although the target density of 95% TD for these large diameter pellets was easily exceeded at all green densities.

3. UO_2 System

Two types of UO_2 were examined. One was a highly-enriched U^eO_2 made by micronizing a mixture of 17.8% $U^{235}O_2$ (93.14% U^{235}) and 82.2% U^dO_2 to provide a powder with an enrichment of 16.7% U^{235}. The other UO_2 was a commercially produced reactor grade (2.8% U^{235}) powder obtained from the Babcock and Wilcox Co., which was used as a standard against which to compare the compaction performance of the AWBA powders without binders.

The highly enriched double micronized UO_2 proved to be a very active powder with sintered pellet densities exceeding 97% over the entire green density range. In some cases, densities were well over 98% as can be seen in Figure 11. Over most of the range, the density curves were again ranked according to the lubricant bulk densities with the lowest bulk density lubricants producing the highest pellet densities. The photomicrographs in Figure 19 show the microstructures for each of the three lubricants tested. The structure of these pellets shows that shrinkage cracks were generated by the rapid densification of this highly active powder.

Since the program objective was to determine if the highly activated micronized powders would binderless press as well as the less active commercial UO_2 powder, the low enriched commercial UO_2 was not activated by micronizing, and, consequently, pellets did not reach as high a sintered density in all cases although densities did exceed 97% using CA-15 at the higher green densities (Figure 12). Only the CA-15 and ZN-16 lubricants were tested with the commercial UO_2 with calcium stearate again producing the highest density pellets. The microstructures shown in Figure 20 contain more fine porosity than seen in the pellets produced from the highly activated AWBA powders. The powder granules do not seem to be as clearly delineated by pores as was the case when using the AWBA powders. The increase in total porosity is consistent with the lower pellet densities.

4. Sintering Comparison

A compilation of the sinterability curves for each of the oxides examined using a 0.1% CA-15 stearate lubricant concentration with the -16/+80 powder fraction is presented in Figure 13. These curves allow a direct comparison of the sintering behavior of the binderless pellets in all the systems examined. The very high activity of the highly enriched UO_2 powder can readily be seen in this plot.

5. Comparing the Three Press Feed Techniques

A comparison can be made between the two bindered and one binderless process using two pass activated thoria powder. This comparison is shown in Figure 14 where the LWBR thoria powder was prepared for pressing by using the liquid/solids process shown in Figure 1. The AWBA-1 curve represents pellets made by the wet process, also shown in Figure 1. Finally, the AWBA-2 pellet density curve represents those pellets made by the AWBA binderless process and is one of the family of curves represented in Figure 6. Pellets made with binders consistently reached higher densities. It should be noted that these curves represent optimum densities reached by small test series of pellets. For instance, thoria seed pellets made for the LWBR core by the liquid/solids process had a mean density of 98.0% TD while those produced in the test series had a mean density of 99.0% indicating that it is not reasonable to expect densities as high as predicted by these curves under production conditions.

VI. GRAIN STRUCTURE

The development of grains in the range of ASTM 4 to 8 in thoria and thoria-base fuel pellets requires the addition of moisture to the hydrogen sintering atmosphere. This so-called "wet" hydrogen atmosphere generally consisted of totally saturated hydrogen at room temperature. The binderless pellets were all sintered in a dry hydrogen atmosphere with a dewpoint of -60°F, and therefore, grain growth was neither expected nor observed. The etched structure consisted of small uniformly distributed grains which were given an ASTM grain size rating of 11 to finer than 13. A typical grain structure for binderless pellets sintered in a dry hydrogen atmosphere is shown in Figure 21 at 400X.

VII. SUMMARY OF RESULTS

The study showed that the AWBA/LWBR pellet making process can be simplified by the binderless compaction of powders with calcium and zinc stearates. Compaction with these stearates can produce acceptable high density pellets from highly active powders. These pellets are free from rejectable cracks and internal defects such as pore clumping and excessive granule delineation. Fuel systems evaluated included the UO_2-ThO_2, ThO_2, and UO_2 systems. Densification of the pellets in each system was a function of the bulk density of the lubricant where the stearates with the lowest bulk densities produced the highest densities in the sintered pellets.

Maximum densities reached in each system using the lowest bulk density calcium stearate (15 lbs/ft^3) and zinc stearate (16 lb/ft^3) are as follows:

Fuel System	Lubricant	Pellet Green Density (% TD)	Maximum Pellet Sintered Density (% TD)
UO_2-ThO_2	Calcium Stearate	61	97.3
UO_2-ThO_2	Zinc Stearate	62	97.0
ThO_2	Calcium Stearate	64	98.0
ThO_2	Zinc Stearate	64	97.7
UO_2 (Highly Active)	Calcium Stearate	60	98.5
UO_2 (Highly Active)	Zinc Stearate	64	98.3
UO_2 (Commercial)	Calcium Stearate	65	97.3
UO_2 (Commercial)	Zinc Stearate	64	96.7

VIII. CONCLUSIONS

1. Highly active oxide fuel powders from the UO_2, UO_2-ThO_2, and ThO_2 systems can be compacted and sintered into high density (greater than 97%) pellets without the assistance of binders.
2. Calcium and Zinc stearates with low bulk densities (15 to 16 lb/ft^3) were the best die lubricants of those t sted as determined by sintered pellet densities.
3. The highest sintered densities were achieved with additive concentrations of 0.1%.
4. Powder granule distribution had little effect so long as the fines (-80 mesh material) did not exceed 25%.
5. Pellet microstructures were evaluated as being equivalent to structures observed in pellets made with binders with exception of grain size.
6. Grain growth was limited since the pellets were not sinterd in wet hydrogen.

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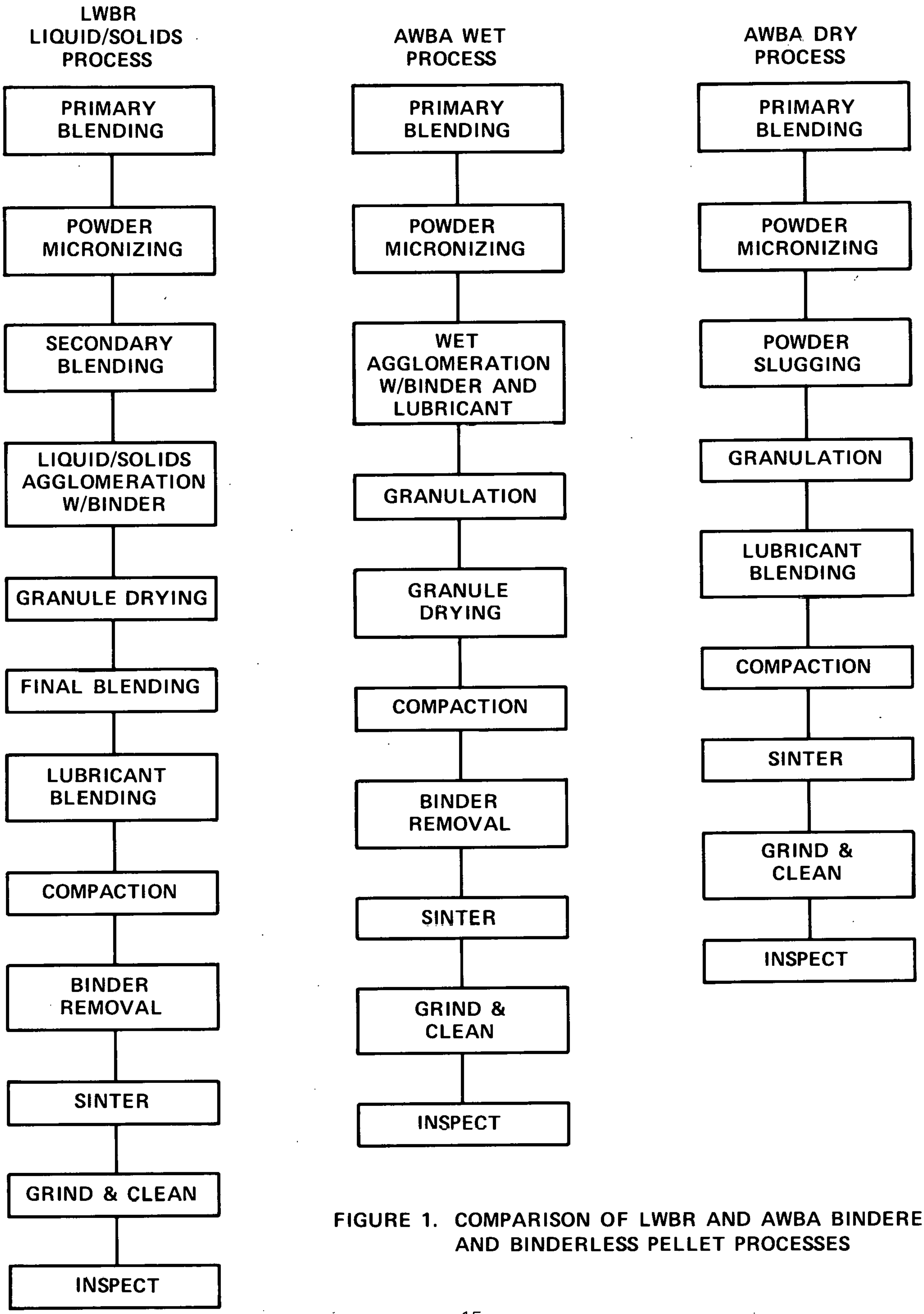


FIGURE 1. COMPARISON OF LWBR AND AWBA BINDERED AND BINDERLESS PELLET PROCESSES

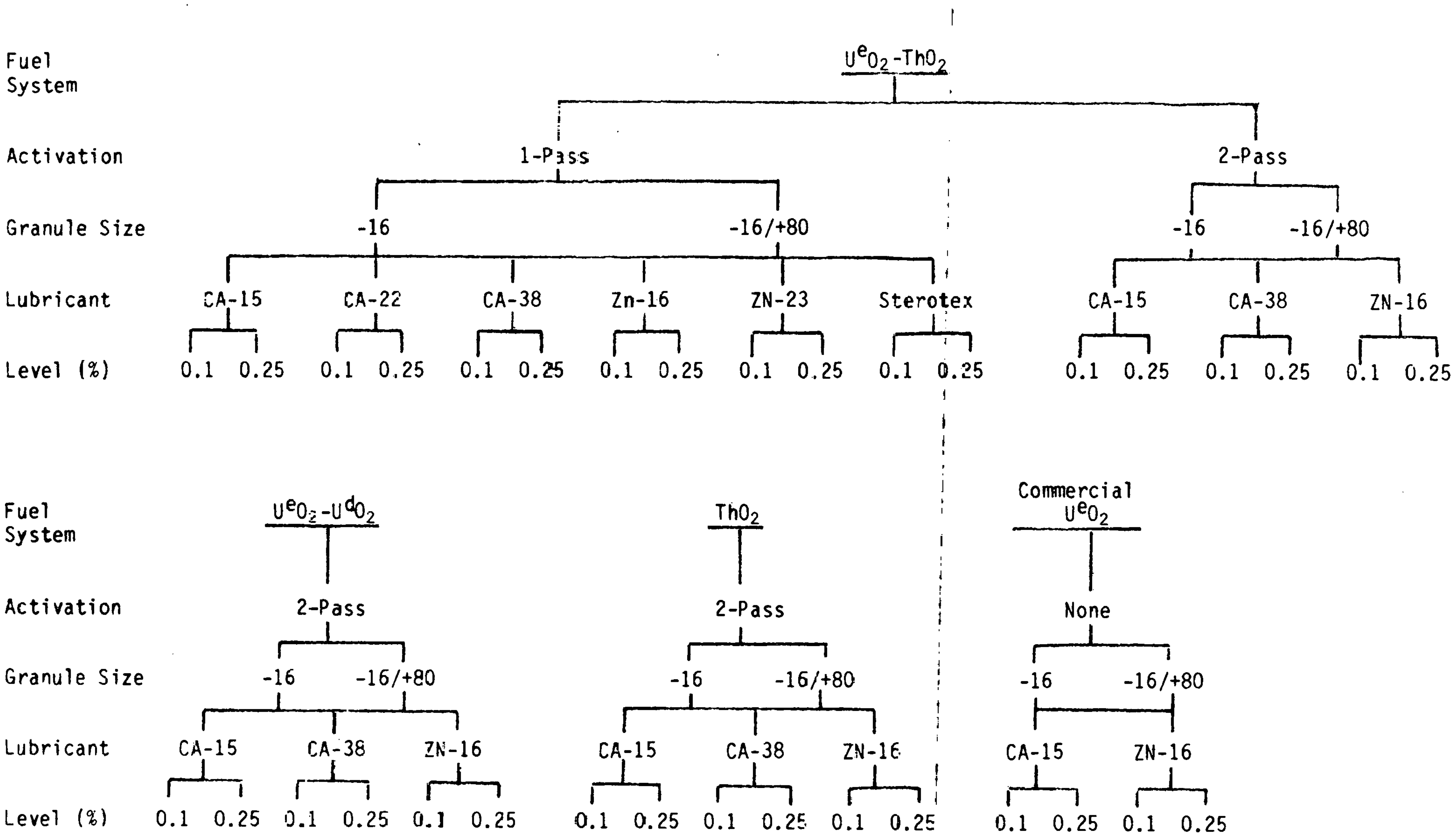


FIGURE 2 EXPERIMENTAL DESIGN

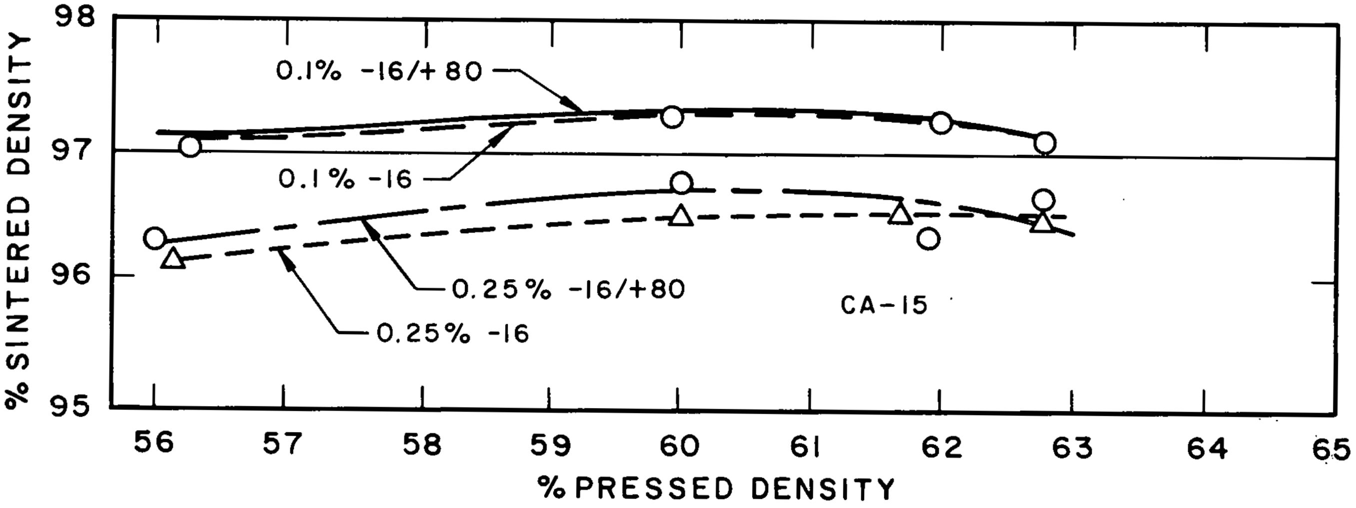


FIGURE 3. SINTERABILITY CURVES: 11.4% U^EO_2-ThO_2 - 2 PASS ACTIVATION. LOW BULK DENSITY CALCIUM STEARATE LUBRICANT TO EVALUATE LUBRICANT CONCENTRATION AND POWDER GRANULE SIZE.

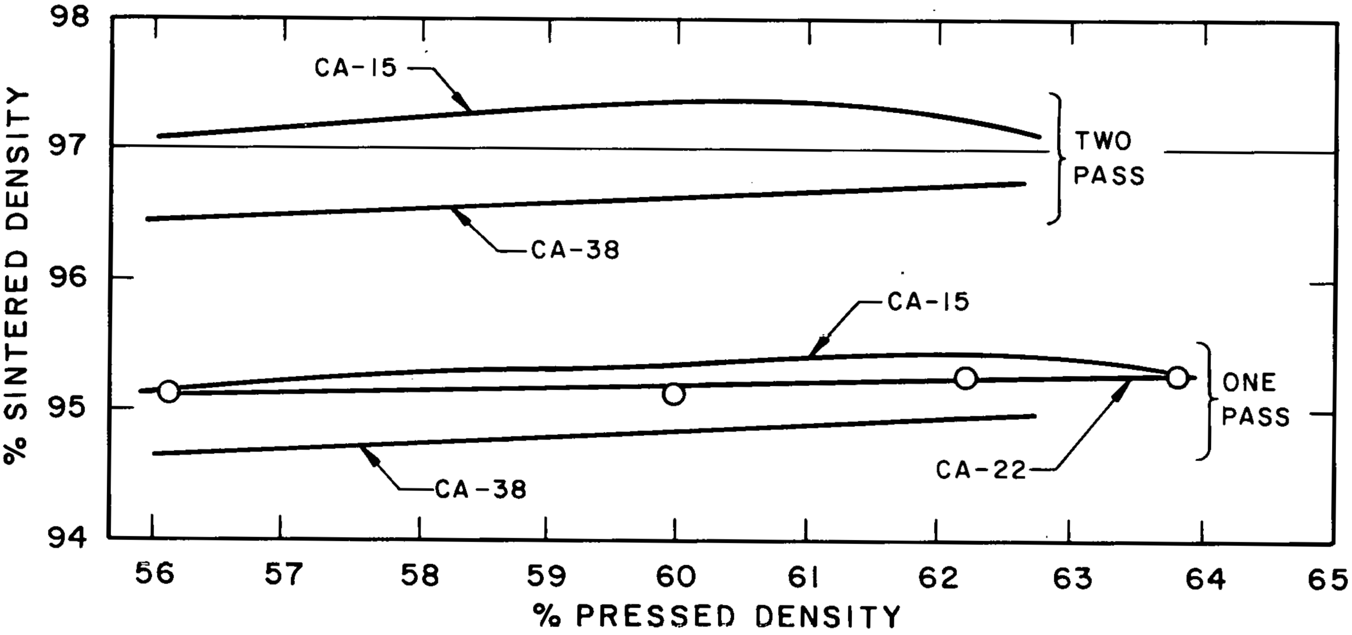


FIGURE 4. SINTERABILITY CURVES: 11.4% U^EO_2-ThO_2 1 & 2 PASS ACTIVATION, -16/+80 POWDER FRACTION COMPARSION OF CALCIUM STEARATE LUBRICANT WITH THREE BULK DENSITIES AT THE 0.1% CONCENTRATION

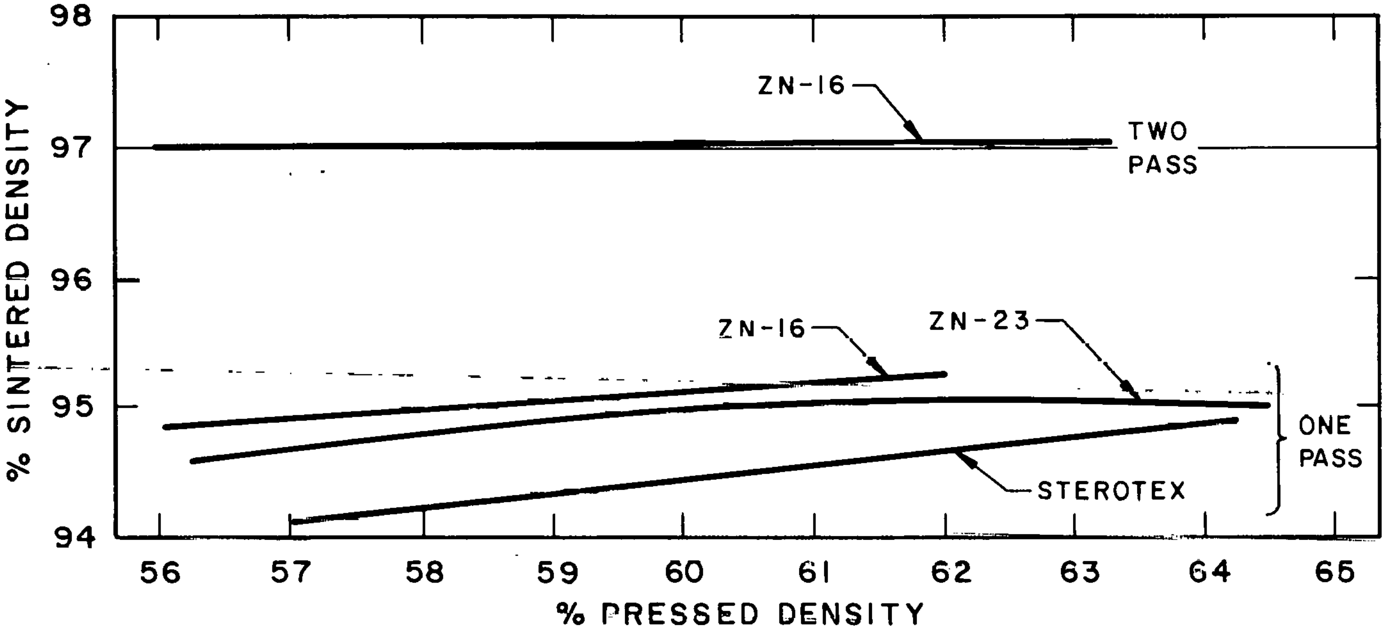


FIGURE 5. SINTERABILITY CURVES: 11.4% $U^EO_2-ThO_2$ 1 & 2 PASS ACTIVATION, -16/+80 POWDER FRACTION COMPARISON OF ZINC STEARATE AT TWO BULK DENSITIES AND STEROTEX, ALL AT 0.1% CONCENTRATION

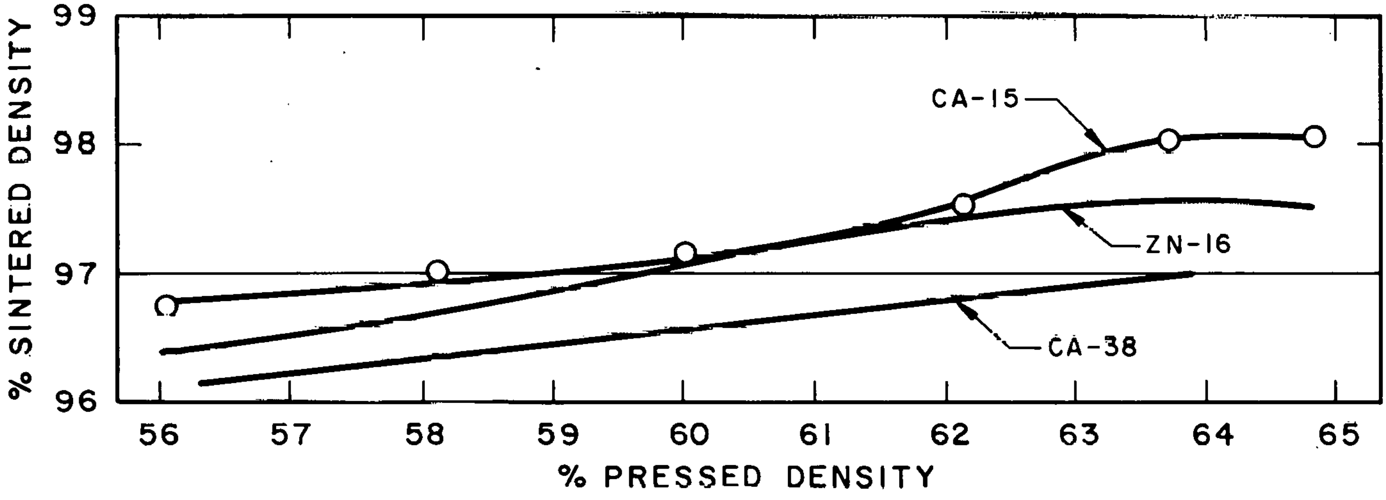


FIGURE 6. COMPARISON OF THE SINTERABILITY OF ThO_2 POWDER (-16/+80 FRACTION) USING THREE STEARATE LUBRICANTS AT 0.1% CONCENTRATION

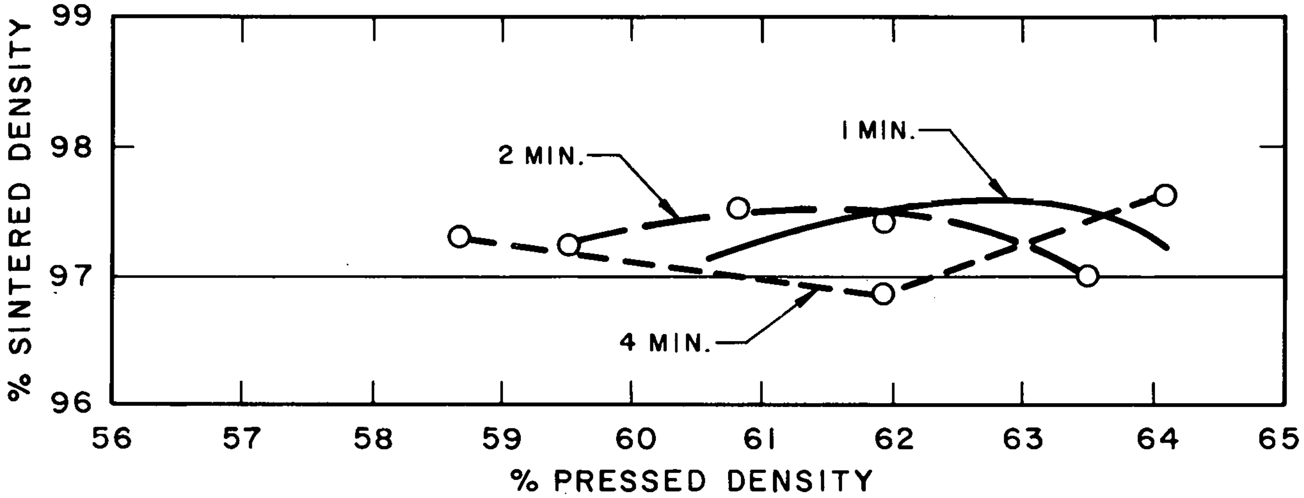


FIGURE 7. COMPARISON BETWEEN LUBRICANT BLENDING TIME AND CONCENTRATION USING A -16/+80 FRACTION OF ThO_2 POWDER. LUBRICANT CONCENTRATION: 0.05% CA-15 STEARATE

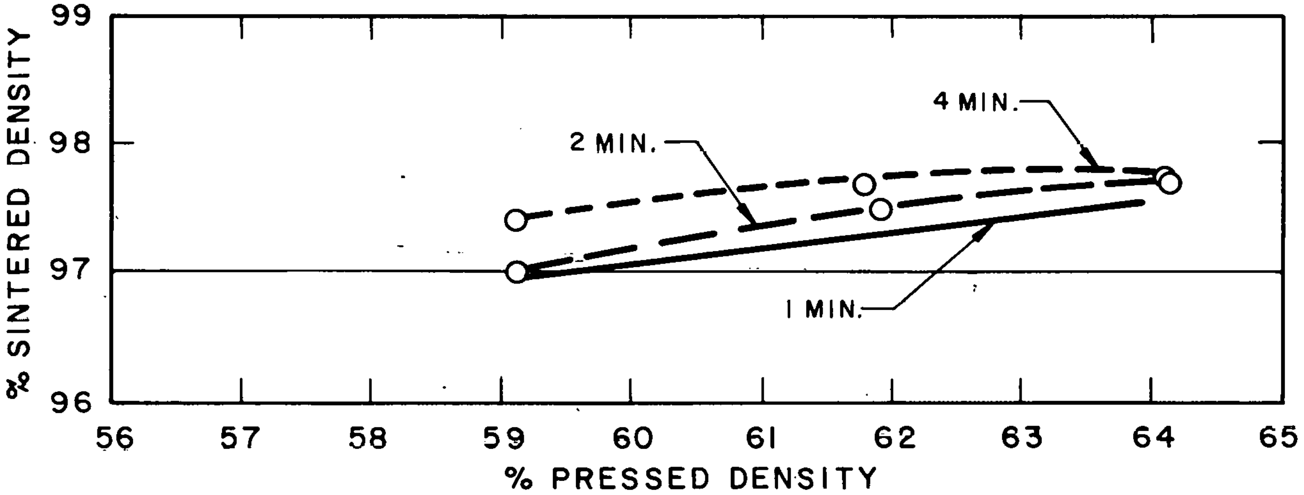


FIGURE 8. COMPARISON BETWEEN LUBRICANT BLENDING TIME AND CONCENTRATION USING A -16/+80 FRACTION OF ThO_2 POWDER. LUBRICANT CONCENTRATION: 0.10% CA-15 STEARATE

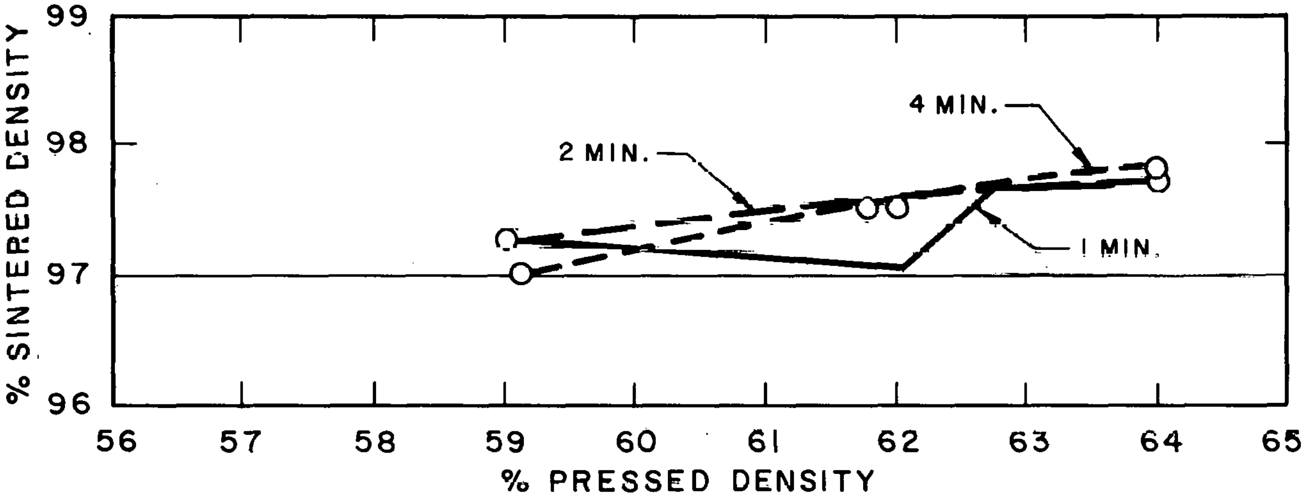


FIGURE 9. COMPARISON BETWEEN LUBRICANT BLENDING TIME AND CONCENTRATION USING A -16/+80 FRACTION OF ThO_2 POWDER. LUBRICANT CONCENTRATION: 0.15% CA-15 STEARATE

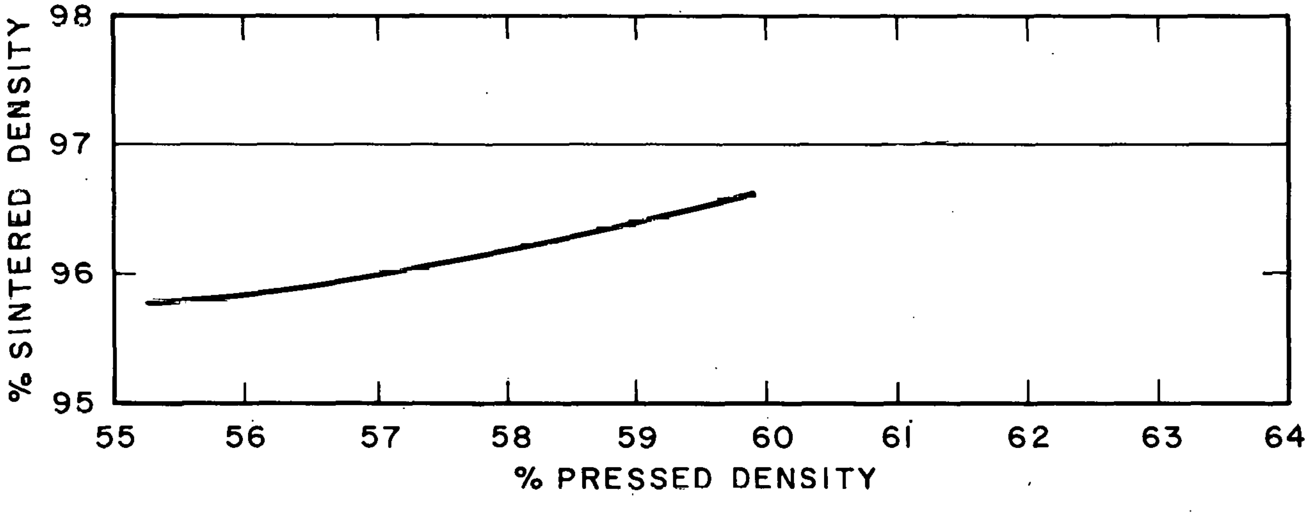


FIGURE 10. SINTERABILITY CURVE FOR THE FABRICATION OF LARGE DIAMETER ANNULAR ThO_2 PELLETS (1.25 IN. OD X 0.635 IN. ID) USING 0.1% ZN-16 STEARATE WITH A -16 MESH GRANULATED ThO_2 POWDER

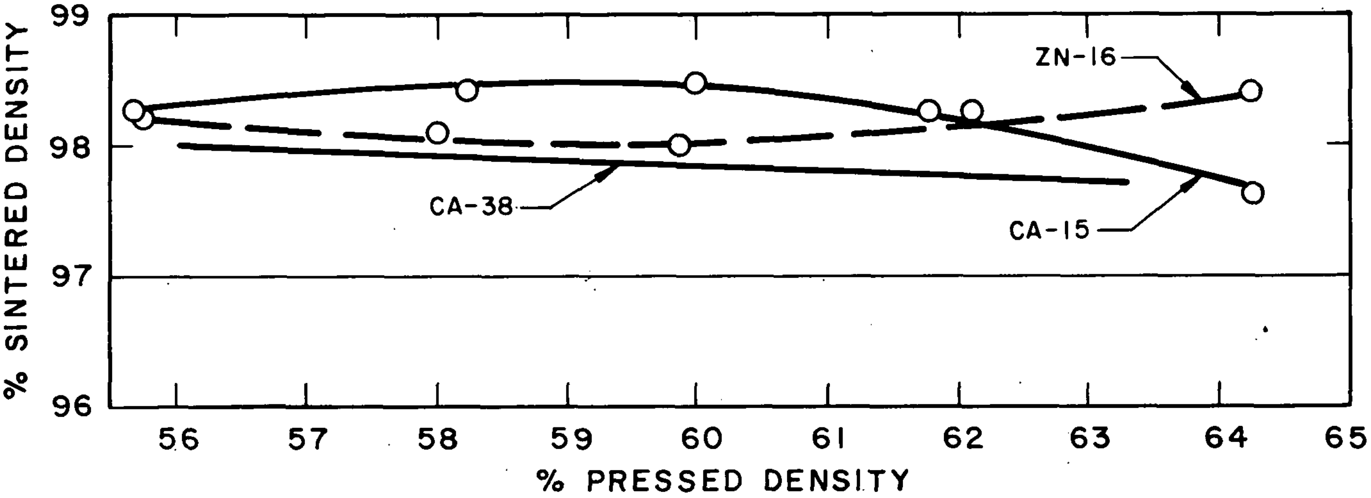


FIGURE 11. COMPARISON OF THE SINTERABILITY OF HIGH ENRICHED UO_2 (14.7% U-235) USING THREE STEARATE LUBRICANTS AT 0.1% CONCENTRATION. TWO PASS ACTIVATION, -16/+80 POWDER FRACTION

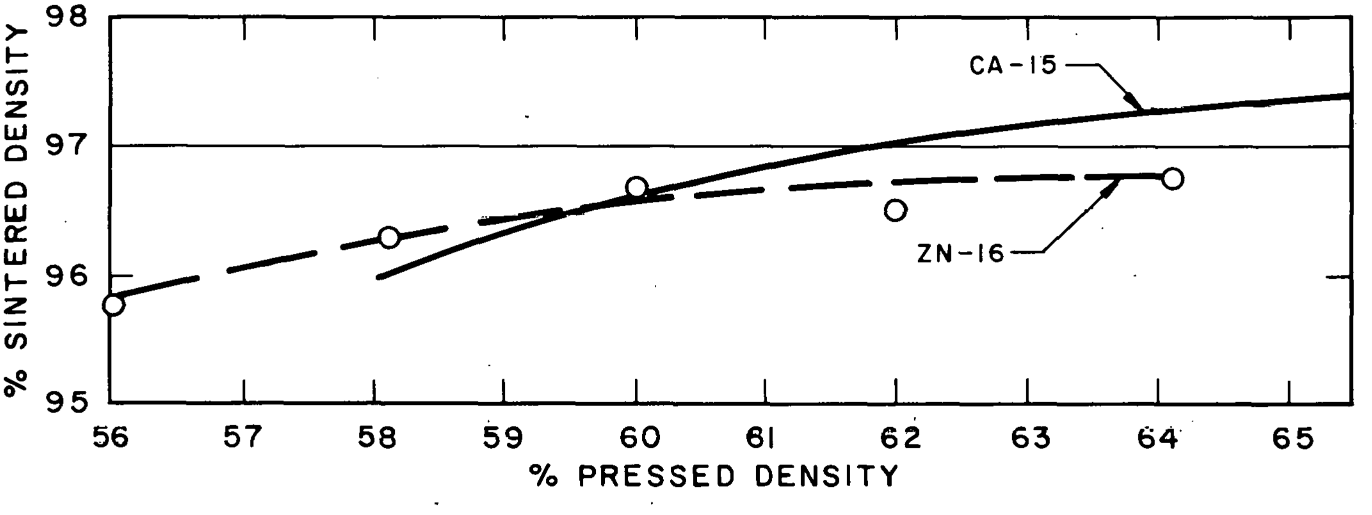


FIGURE 12. COMPARISON OF THE SINTERABILITY OF LOW ENRICHED COMMERCIAL UO_2 (2.84% U-235) USING TWO STEARATE LUBRICANTS AT 0.1% CONCENTRATION. NO ACTIVATION, -16/+80 POWDER FRACTION.

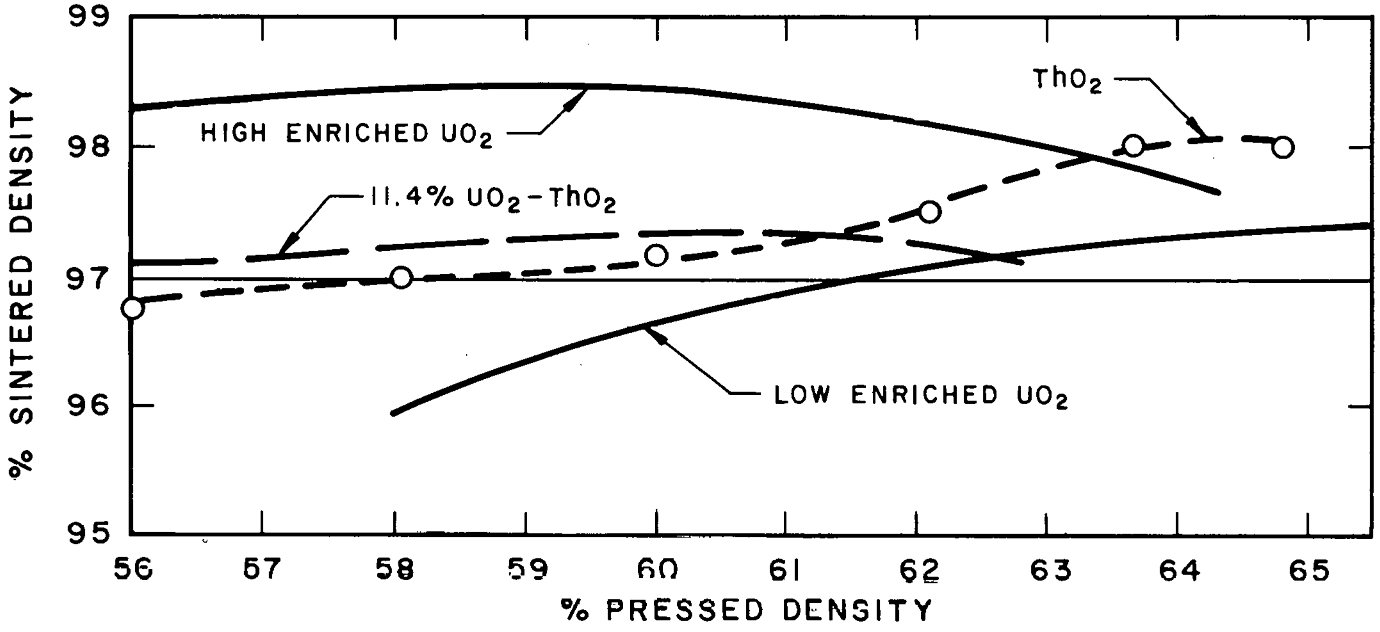


FIGURE 13. SINTERABILITY CURVES FOR THE INDICATED OXIDES USING CA-15 STEARATE AT THE 0.1% CONCENTRATION USING THE -16/+80 POWDER FRACTION

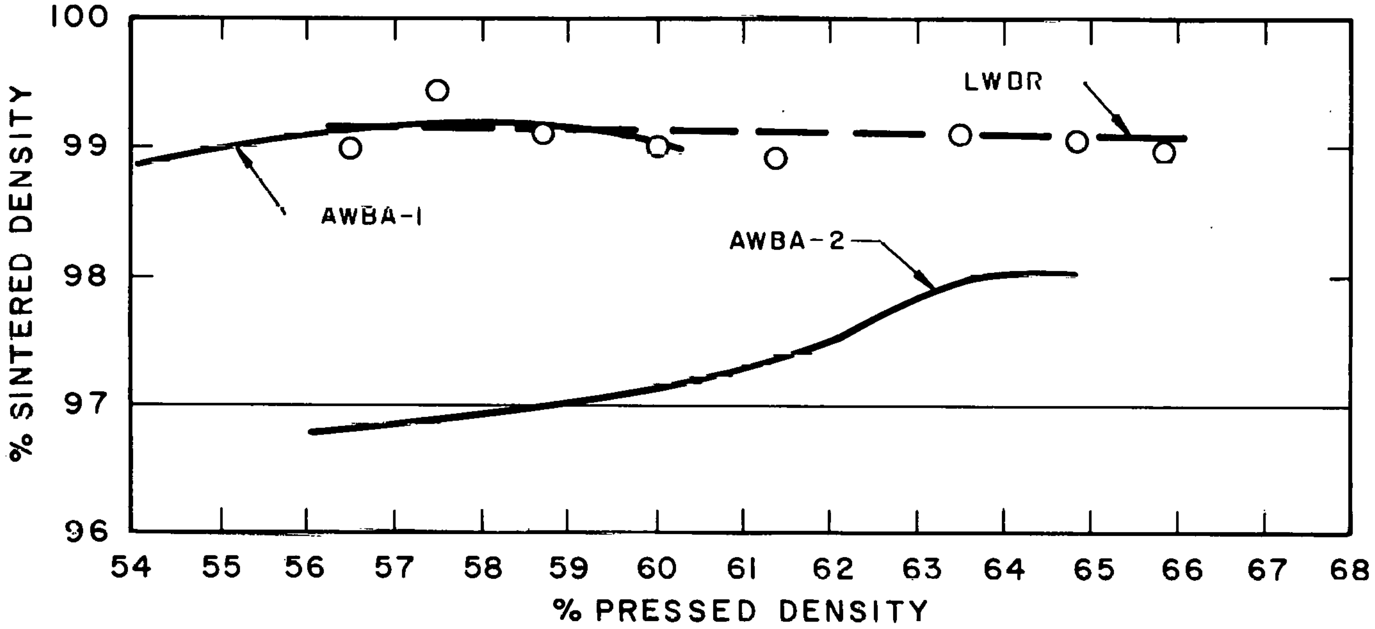
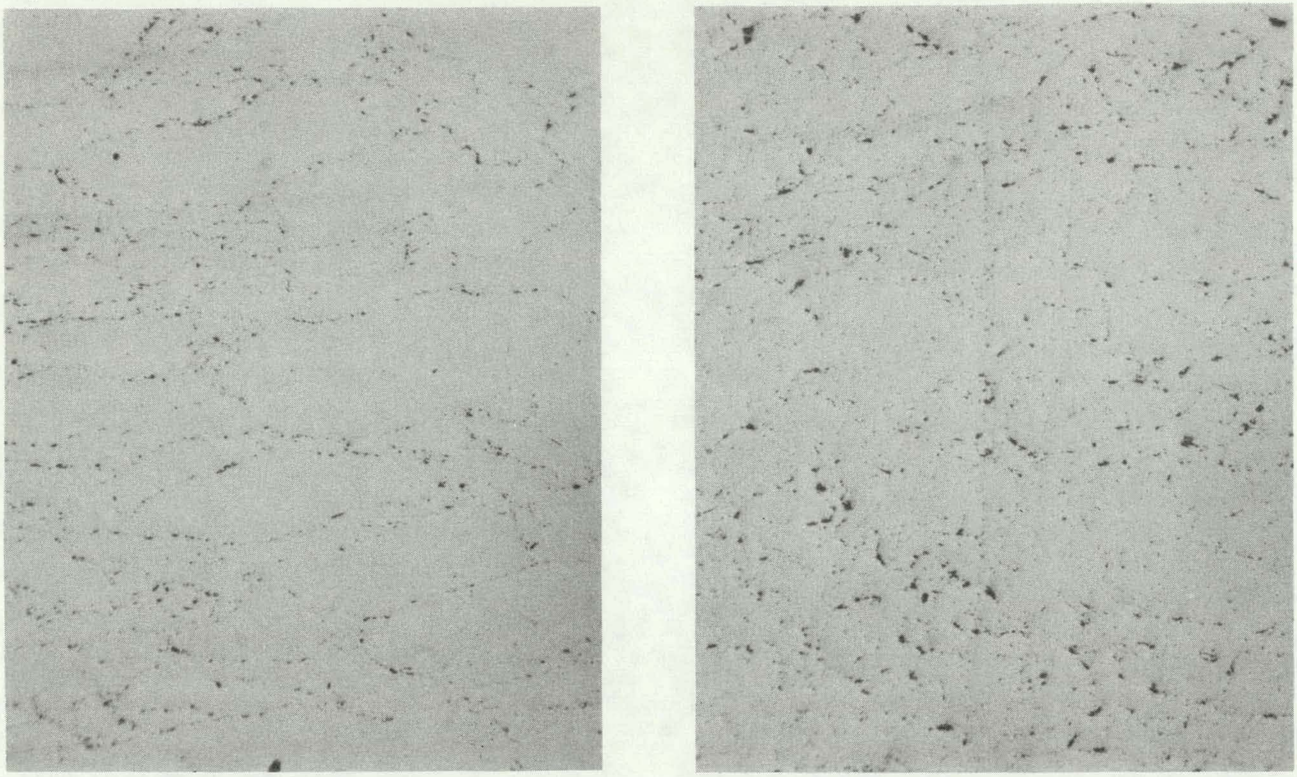


FIGURE 14. SINTERABILITY OF ThO_2 USED FOR THE LWBR CORE PELLETS AND AWBA TESTS. AWBA-1 AND LWBR PELLETS MADE WITH CARBOWAX 6000 BINDER. AWBA-2 PELLETS MADE WITHOUT BINDER USING CA-15 STEARATE.



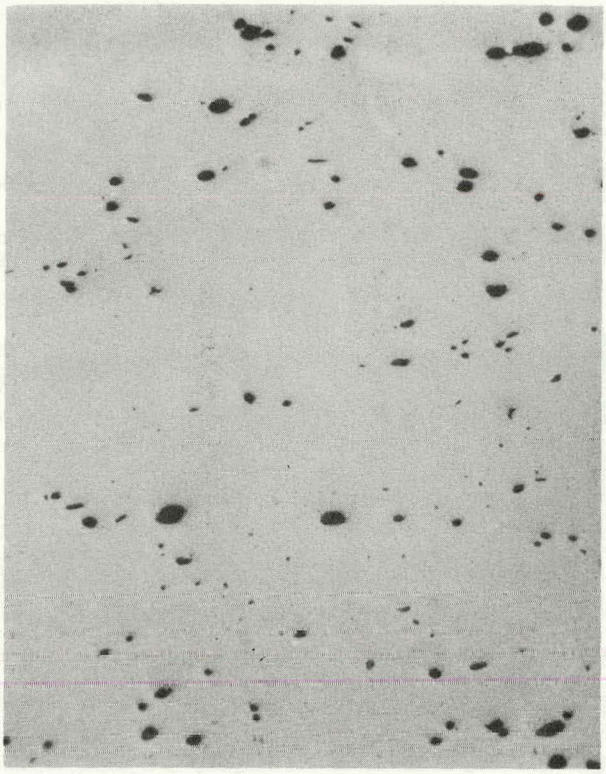
(a) 0.1% ADDITION 50X
-16/+80 POWDER

(b) 0.1% ADDITION 50X
-16 POWDER

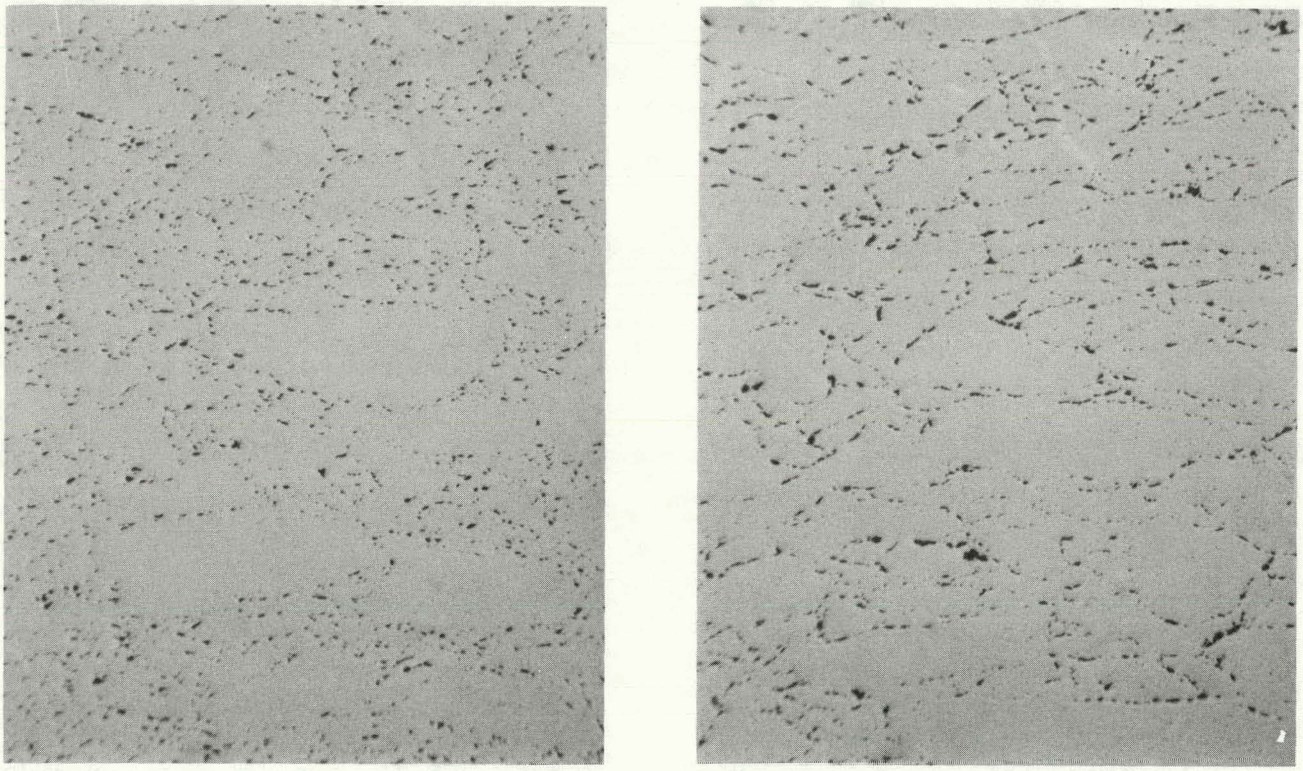
(c) 0.25% ADDITION 50X
-16/+80 POWDER

(d) 0.25% ADDITION 50X
-16 POWDER

FIGURE 15. MICROSTRUCTURES OF 11.4% UO_2-ThO_2 PELLETS MADE WITH CALCIUM-15 STEARATE



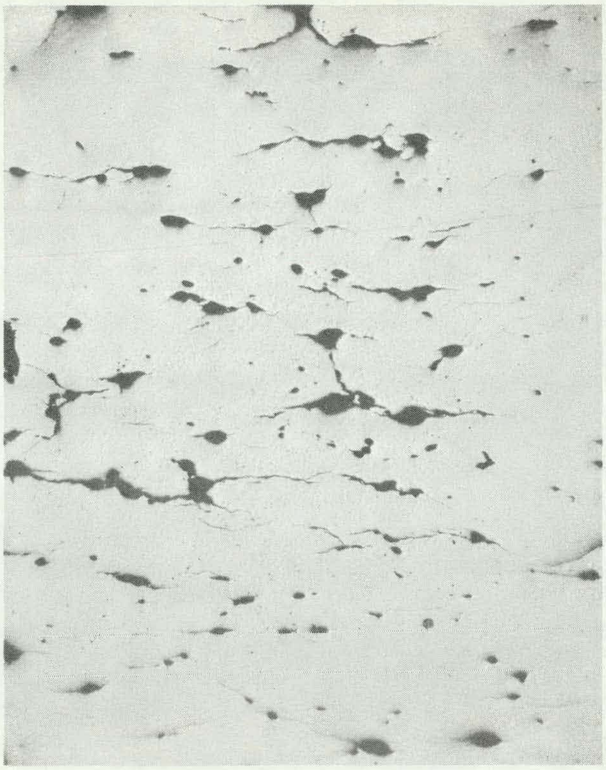
(a) 0.1% CA -38 STEARATE 50X
-16/+80 POWDER



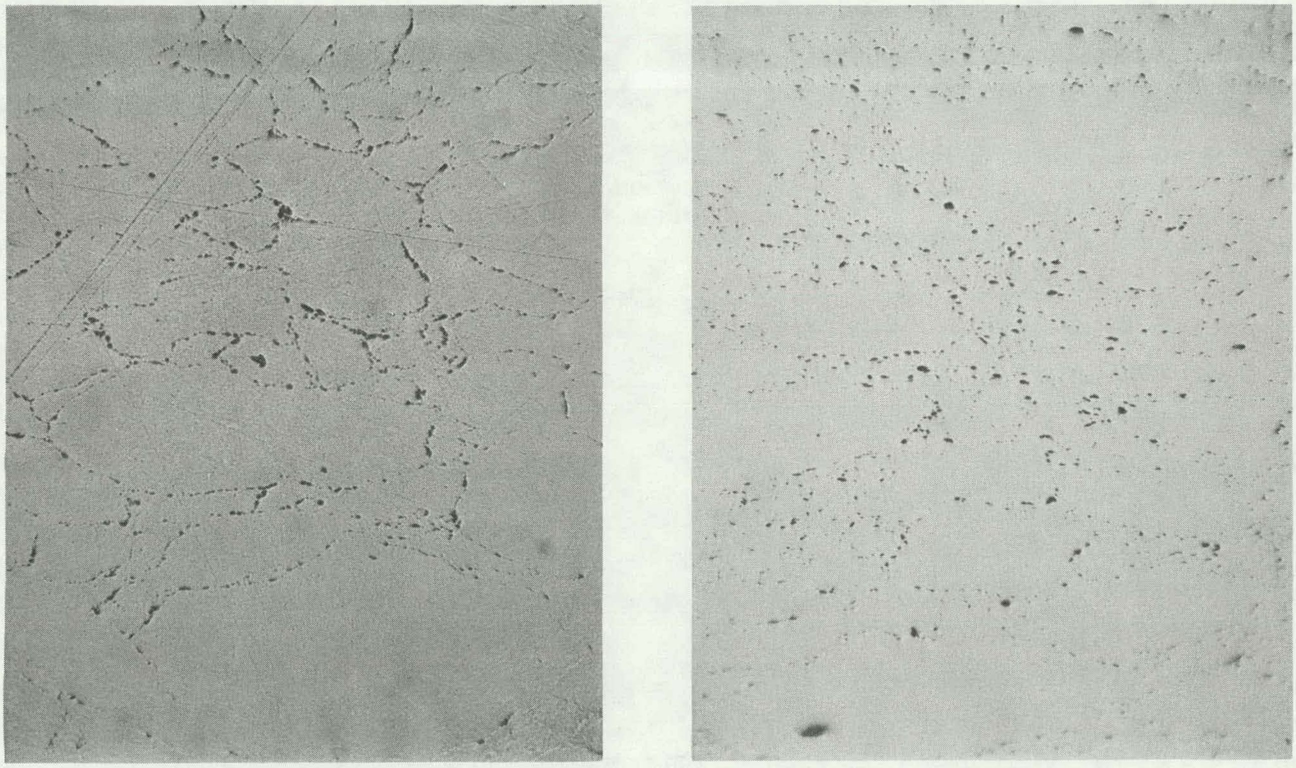
(b) 0.1% ZN -16 STEARATE 50X
-16/+80 POWDER

(c) 0.1 % ZN -16 STEARATE 50X
-16 POWDER

FIGURE 16. MICROSTRUCTURES OF 11.4% UO_2 - ThO_2 PELLETS MADE WITH TWO DIFFERENT STEARATES.



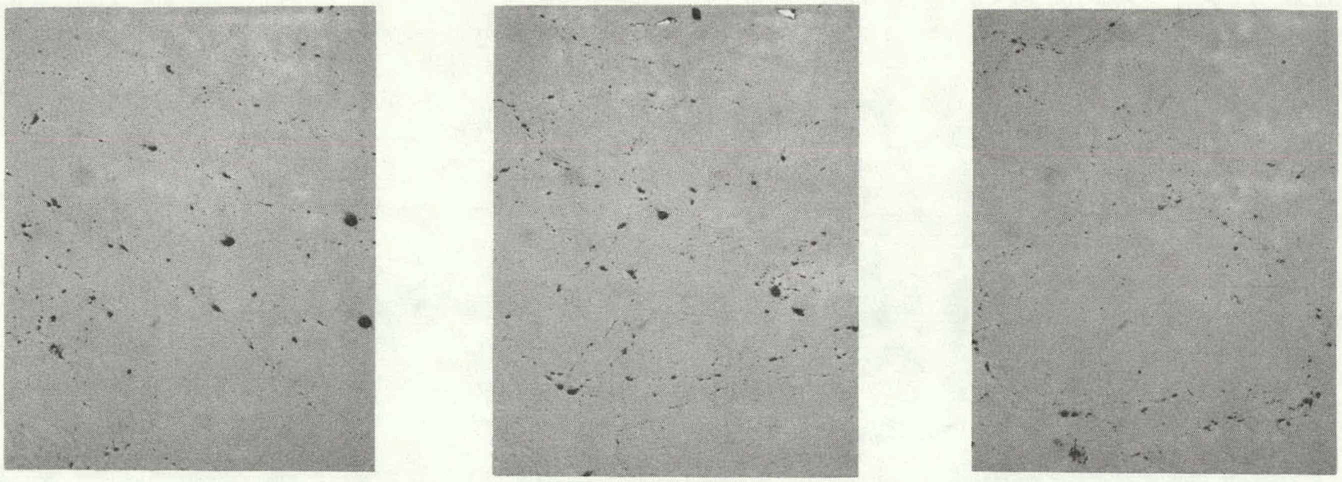
(a) 0.25% CA-38 STEARATE 50X
-16/+80 POWDER



(b) 0.1% CA-15 STEARATE 50X
-16/+80 POWDER

(c) 0.1% ZN-16 STEARATE 50X
-16 POWDER

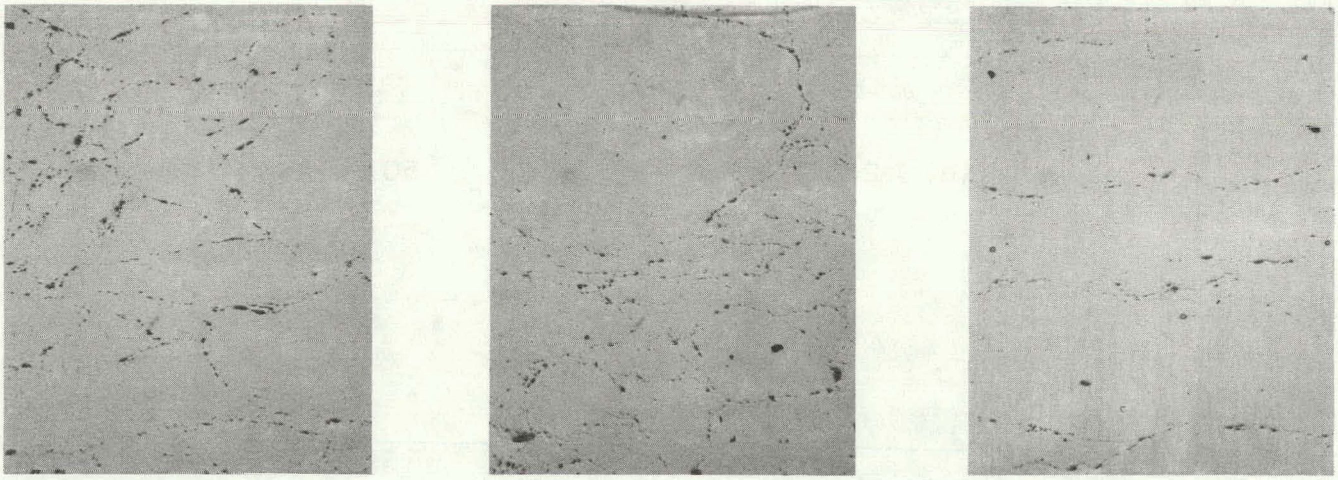
FIGURE 17. MICROSTRUCTURES OF ThO_2 PELLETS MADE WITH THREE DIFFERENT STEARATES.



(a) 0.05% ADDITION 105X
1 MIN. BLENDING

(b) 0.05% ADDITION 105X
2 MIN. BLENDING

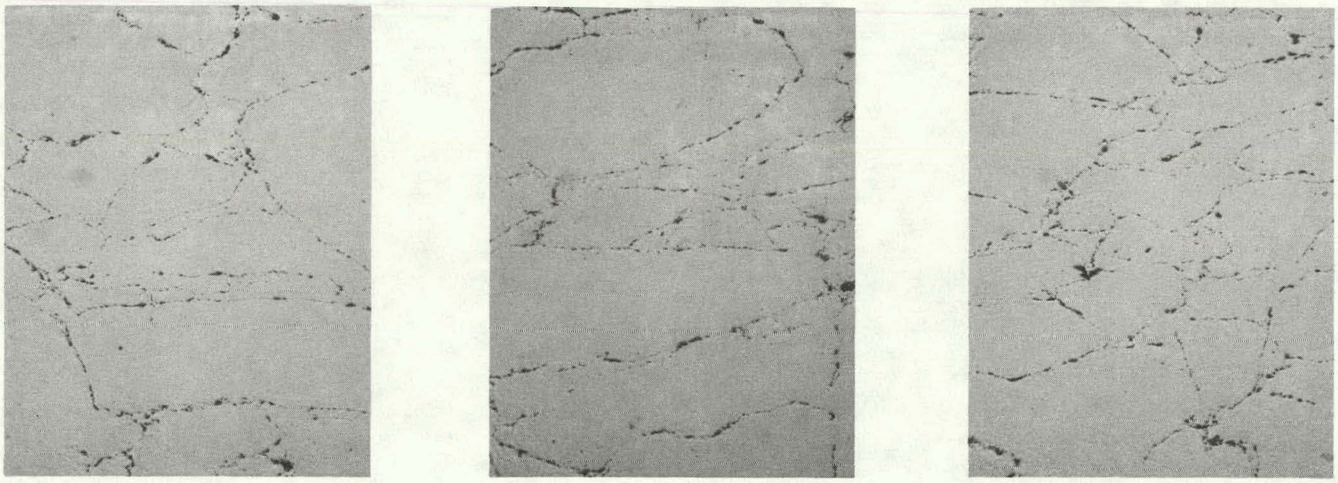
(c) 0.05% ADDITION 105X
4 MIN. BLENDING



(d) 0.1% ADDITION 105X
1 MIN. BLENDING

(e) 0.1% ADDITION 105X
2 MIN. BLENDING

(f) 0.1% ADDITION 105X
4 MIN. BLENDING

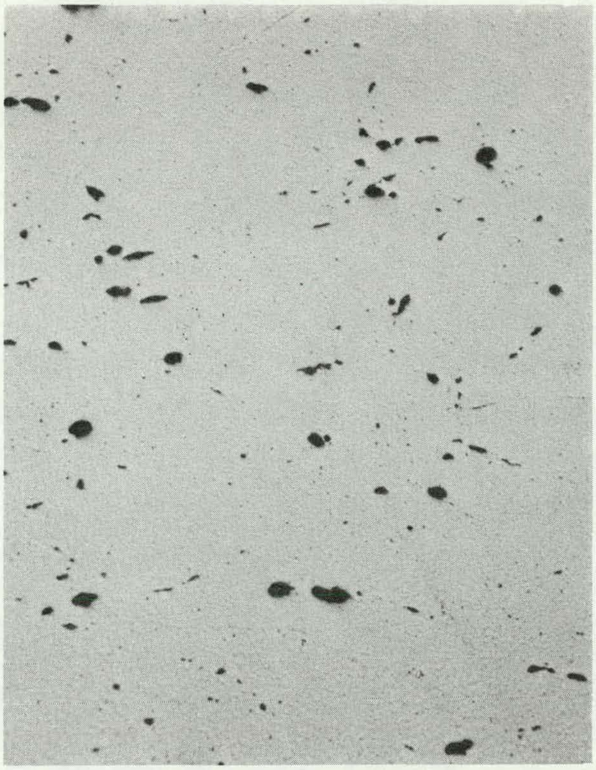


(g) 0.15% ADDITION 105X
1 MIN. BLENDING

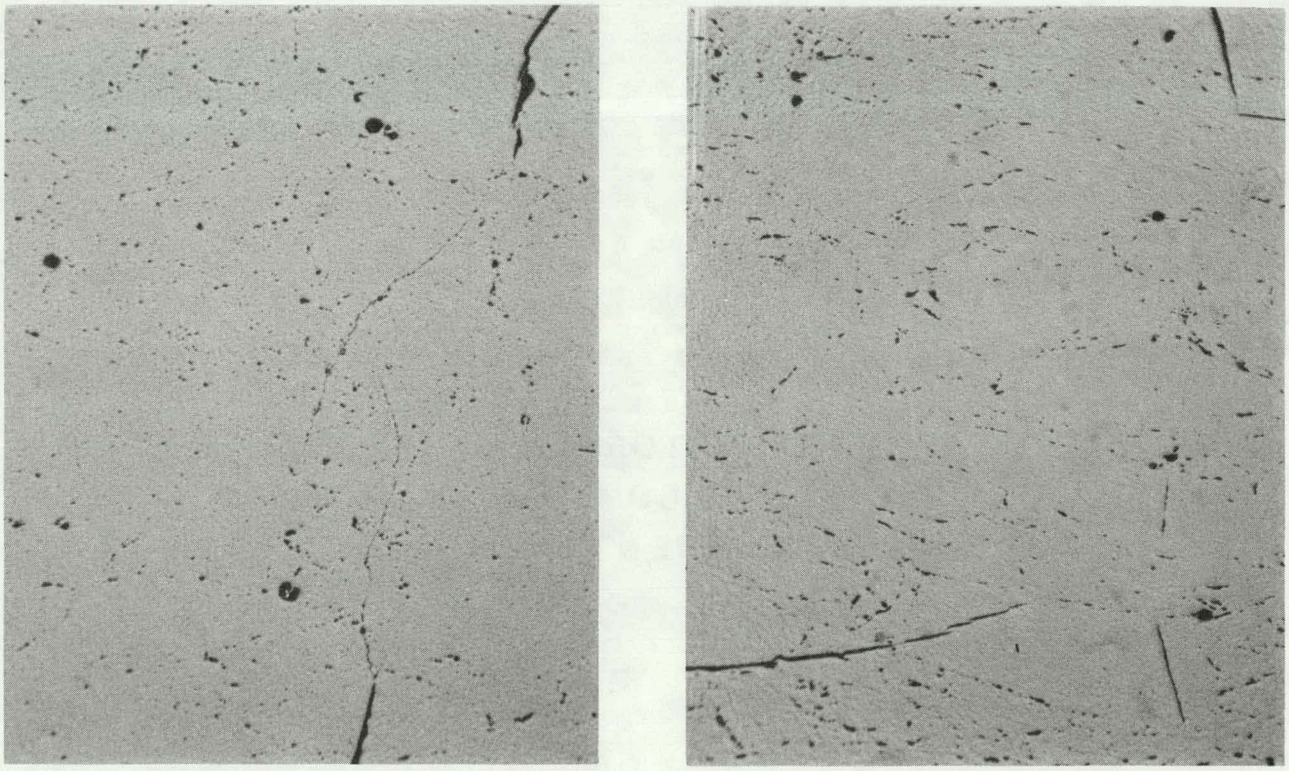
(h) 0.15% ADDITION 105X
2 MIN. BLENDING

(i) 0.15% ADDITION 105X
4 MIN. BLENDING

FIGURE 18. MICROSTRUCTURES OF ThO_2 PELLETS MADE WITH CA-15 STEARATE LUBRICANT



(a) 0.1% CA -38 STEARATE 50X
-16/+80 POWDER



(b) 0.1% CA -15 STEARATE 50X
-16/+80 POWDER

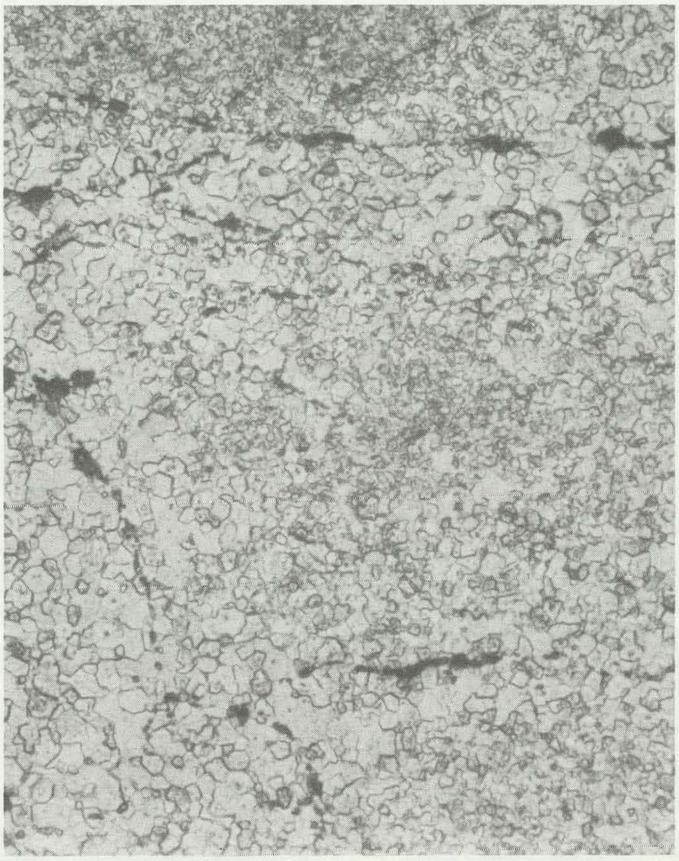
(c) 0.1% ZN -16 STEARATE 50X
-16/+80 POWDER

FIGURE 19. MICROSTRUCTURES OF HIGH ENRICHED UO_2 PELLETS MADE FROM MICRONIZED POWDER WITH THREE DIFFERENT STEARATES.

(a) 0.1% CA -15 STEARATE 50X
-16/+80 POWDER

(b) ZN -16 STEARATE 50X
-16/+80 POWDER

FIGURE 20. MICROSTRUCTURES OF LOW ENRICHED UO_2 PELLETS MADE FROM COMMERCIAL POWDER WITH TWO DIFFERENT STEARATES.



ETCHED 400X

FIGURE 21. GRAIN STRUCTURE IN ThO_2 PELLET SINTERED IN DRY HYDROGEN

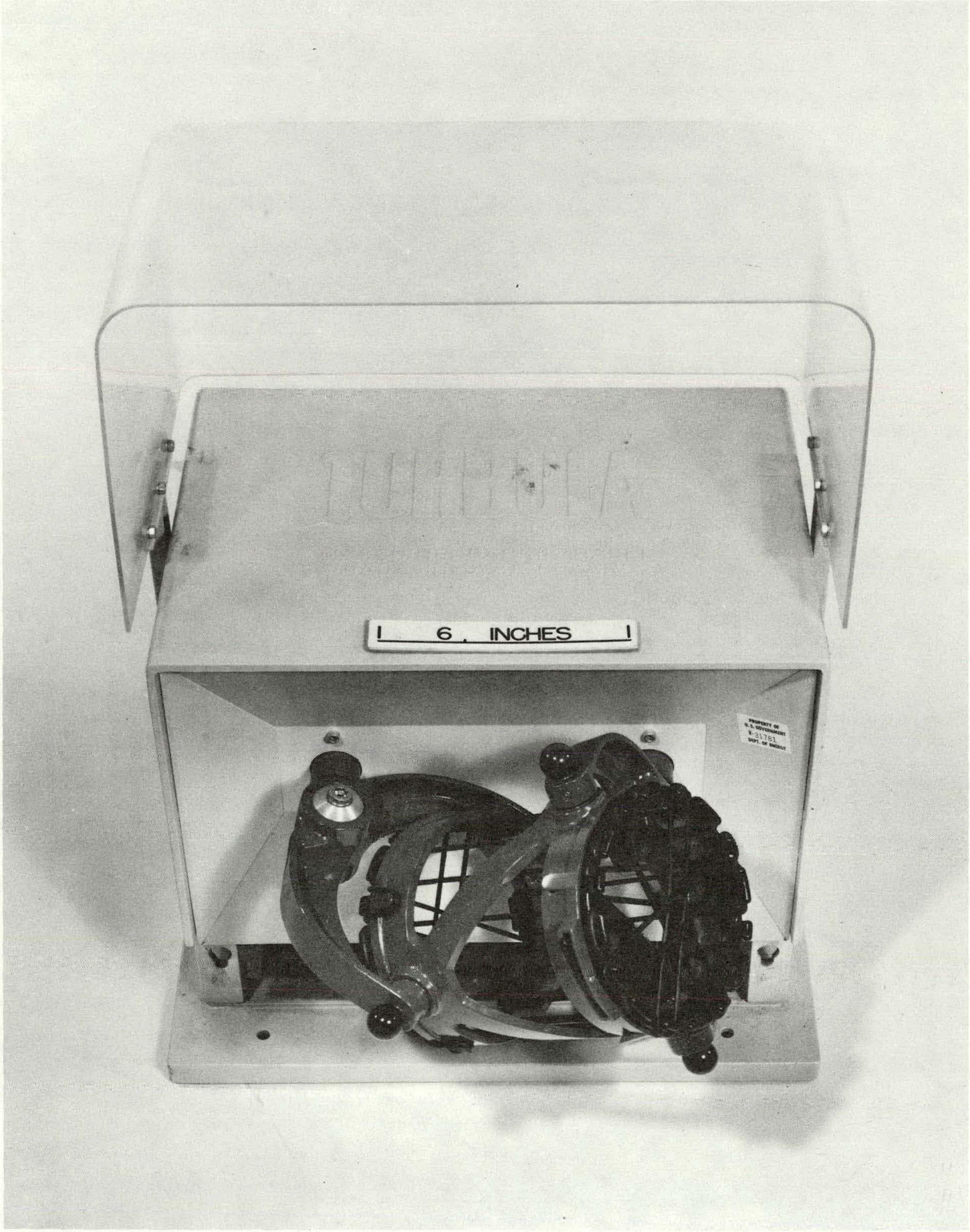


FIGURE 22. TURBULA BLENDER

APPENDIX I

TECHNIQUES FOR AGGLOMERATING FUEL POWDERS WITH BINDERS

The binder used throughout the LWBR and AWBA programs was a polyethylene glycol known as Carbowax 6000. Binder contents were on the order of 1.25% of the powder weight. The die lubricant, which was needed to reduce both the interparticle frictional forces and the frictional forces between the particles and the wall of the die, was a dry powdery organic stearate known as Sterotex. Two different agglomerating techniques were used to incorporate the binder. These techniques are described below and are shown in the processing sequence in the flow diagram in Figure 1. It will be noted that all three processes start with blending and micronizing operations to ensure that the powder is homogeneous before and after activation.

The LWBR fuel powders were agglomerated with the Carbowax binder in a liquid/solids blender (Reference (b)). This type of blender uses a spraying technique to wet the powder with a solution of the binder while it is being tumbled. The powder is granulated within the blender by chopping it with a bladed intensifier bar. The solvent for the binder is a mixture of dichlorodifluormethane and methylene chloride known commercially as Oxylene M-6. The Sterotex die lubricant could not be added with the binder during liquid/solids agglomeration and was dry blended into the granulated powder prior to pressing. The liquid/solids agglomerating technique was capable of handling the large batch sizes required for the manufacture of LWBR core fuel.

The smaller batch sizes used for the manufacture of AWBA development and irradiation test pellets were better accommodated by a technique known as wet agglomeration (References (c), (d), (e)). In the wet process, the dissolved binder and lubricant are added to the fuel powder in a mixer bowl with a sufficient quantity of the Oxylene solvent to form a very fluid slurry. A planetary mixer then agglomerates the slurry until the resulting mixture is dry enough to granulate through a suitably-sized sieve. The granulated press feed material produced by the wet technique had a very uniform dispersion of the binder and die lubricant. This press feed material was highly compactible and would generally produce sintered pellets having microstructures containing only small uniformly dispersed pores, which is the desired microstructure.

Although the agglomeration process could be controlled so as to produce soft fully compactible granules, both the wet and liquid/solids processes were time consuming. Both processes also encountered limiting criticality restrictions on batch size due to the Oxylene solvent which is a neutron moderator. Finally, the presence of the binder requires an additional furnacing operation prior to sintering to debinder the pellets, which adds substantially to the manufacturing costs.

Owing to the large amount of binder present (1.25%) a special furnace is required for debindering. This furnace uses a flowing CO_2 atmosphere to remove the binder without oxidizing the pellets at a temperature of 925°C. This furnace needs a programmable heating rate to prevent too rapid heating of the pellets through the binder decomposition temperature (around 400°C).