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PLUTO FUEL ELEMENT FABRICATION AT LAWRENCE RADIATION LABORATORY

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AEC Research and
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PLUTO FUEL ELEMENT FABRICATION AT LAWRENCE RADIATION LABORATORY

(Title: Unclassified)

W. A. Sandholtz

February 8, 1965

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PLUTO FUEL ELEMENT FABRICATION AT LAWRENCE RADIATION LABORATORY

W. A. Sandholtz

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Livermore, California

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ABSTRACT

Development of an extrusion-based process for producing beryllia fuel elements for the Pluto nuclear powered ramjet is described. The goal was to extrude and fire hexagonal tubes to close dimensional tolerances without grinding. Sinterable beryllia powder was fueled by precipitating uranium and fuel additives from a single aqueous solution in which the beryllia had been slurried. After sintering, the fuel phase consisted of a phase-stabilized solid solution of urania (UO_2) with yttria and zirconia.

Principal operations consisted of slurrying and precipitation, filtration, drying and calcination, vehicle incorporation and mixing, extrusion, drying and vehicle burnout, and hydrogen sintering (including straightening).

Directions for continued development toward the "no grinding" goal are pointed out.

PLUTO FUEL ELEMENT FABRICATION AT LAWRENCE RADIATION LABORATORY

W. A. Sandholtz

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I. INTRODUCTION

Process development of the extrusion method of fabricating ceramic fuel elements for the Pluto reactors ended at the Lawrence Radiation Laboratory (LRL) early in 1963. Sufficient development work had been done earlier to permit the production of a core of Tory II-C elements at Coors Porcelain Company. However, further refinements in the process were desired. During Fiscal 1964 development of the process was continued at Coors Porcelain Company.

This report outlines the course of development at LRL, describes its final status, and suggests areas where further development was desirable.

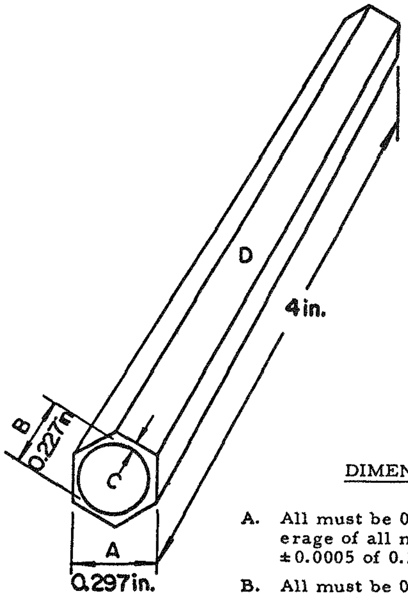
II. BACKGROUND

A. Fuel Element

The basic requirement was to develop a process capable of producing hundreds of thousands of tubular beryllia fuel elements. Near-theoretical densities and close dimensional tolerances were specified. Production of acceptable elements in good yield without resort to grinding of the sintered parts was set as the ultimate development goal.

Design for the Tory II-C reactor specified the basic fuel element dimensions indicated in Fig. 1. Smaller numbers of tubes of different dimensions were also required. Other criteria included chemical purity (especially concentrations of nuclear poisons), fuel phase composition and concentration, inclusions and voids, cracks, chips, surface finish, additional dimensional requirements, dimensional stability, and strength (modulus of rupture).

Refer to the Appendix for an outline of raw materials properties specified.



DIMENSIONS:

A. All must be 0.297 ± 0.001, average of all must lie within ±0.0005 of 0.297.

B. All must be 0.227 ± 0.002, average of all must lie within ±0.0005 of 0.227.

C. Must be 0.035 ± 0.003 for fueled tubes.

D. All surfaces 'D' must lie entirely within the space between two concentric hexagonal boundary surfaces whose dimensions 'A' are 0.2985 and 0.2955, respectively.

GLL-624-16505

Fig. 1. Pluto fuel element.

Inclusions and voids were located and measured by radiographic examination. Cracks, chips, and surface finish were observed with the aid of fluorescent penetrant dyes. Dimensional stability was checked after exposure of the tube to an elevated temperature cycle. Modulus of rupture was measured by a simple three-point beam loading of tubes at room temperature in standard test apparatus.

Early process work (Tory II-A) was done with simple urania-beryllia mixtures, However, by the time Tory II-C was in production the fuel phase was no longer urania alone but a phase-stabilized urania-zirconia-yttria combination of 1.06:1:1 molar ratio.

B. Process History

In the latter part of 1957, preliminary work with beryllia powders was started at LRL, Livermore, By early 1958, methods of addition of nuclear fuel (urania) to beryllia and the extrusion method of fabrication were under study. In mid-1958, the extrusion of thin-walled tubes began and the exploration of extrusion vehicles and mixing techniques was underway. Design of extruder die parts and development of the technique for precipitation of nuclear fuel from solution in situ with slurried beryllia powder (an operation termed "slurpitation") followed in late 1958.

During the last part of 1958, the basic process was laid out and the LRL production facility was planned.

Through early and middle 1959, the installation of processing equipment and the study of process parameters proceeded simultaneously. Enriched uranium was introduced to the process in late 1959.

After some process shakedown in January 1960, the facility started into a fuel element production schedule in February 1960. Following a period of production experience the decision was made to use the facility for process development rather than production. Changes in nuclear fuel composition and dimensional specifications introduced new processing problems.

Figure 2 presents two process flow charts. The left flow line (Fig. 2A) outlines the original process; the right flow line (Fig. 2B) outlines the terminal status of the process. The basic elements of the process remain unchanged; however, some processing steps have been added and some have been modified. Note that steps 3 (screening), 6 (calcining), and 9 (rubber milling) are not in the original process line.

Much of the equipment in the plant was sized and spaced to preclude any possibility of nuclear accidents. Note that fully enriched (93%) U^{235} was used. In those operations for which criticality-safe geometry was not possible, safe operating and handling procedures were detailed and rigidly enforced.

III. DISCUSSION

In this section the development of the LRL extrusion process is summarized step by step.

A. General

The basic extrusion process as first set up at LRL remained unchanged throughout development. Initial production efforts were frustrated mainly by control problems and process details. Two basic shortcomings of the initial process were identified: 1) the kind of batch mixing required to assure good tube properties was not well recognized; and 2) the necessity for sintering fuel elements in a restraining configuration became apparent only after the process was operating.

B. Raw Materials

1. Beryllia

UOX-grade powder from Brush Beryllium Company was used. It is largely in the submicron particle size range and contains about 200 ppm of spectrographic impurities. Its major anion impurity is sulfur (500 to 1000 ppm in the raw powder). Typically, it shows about 0.3% loss on ignition,

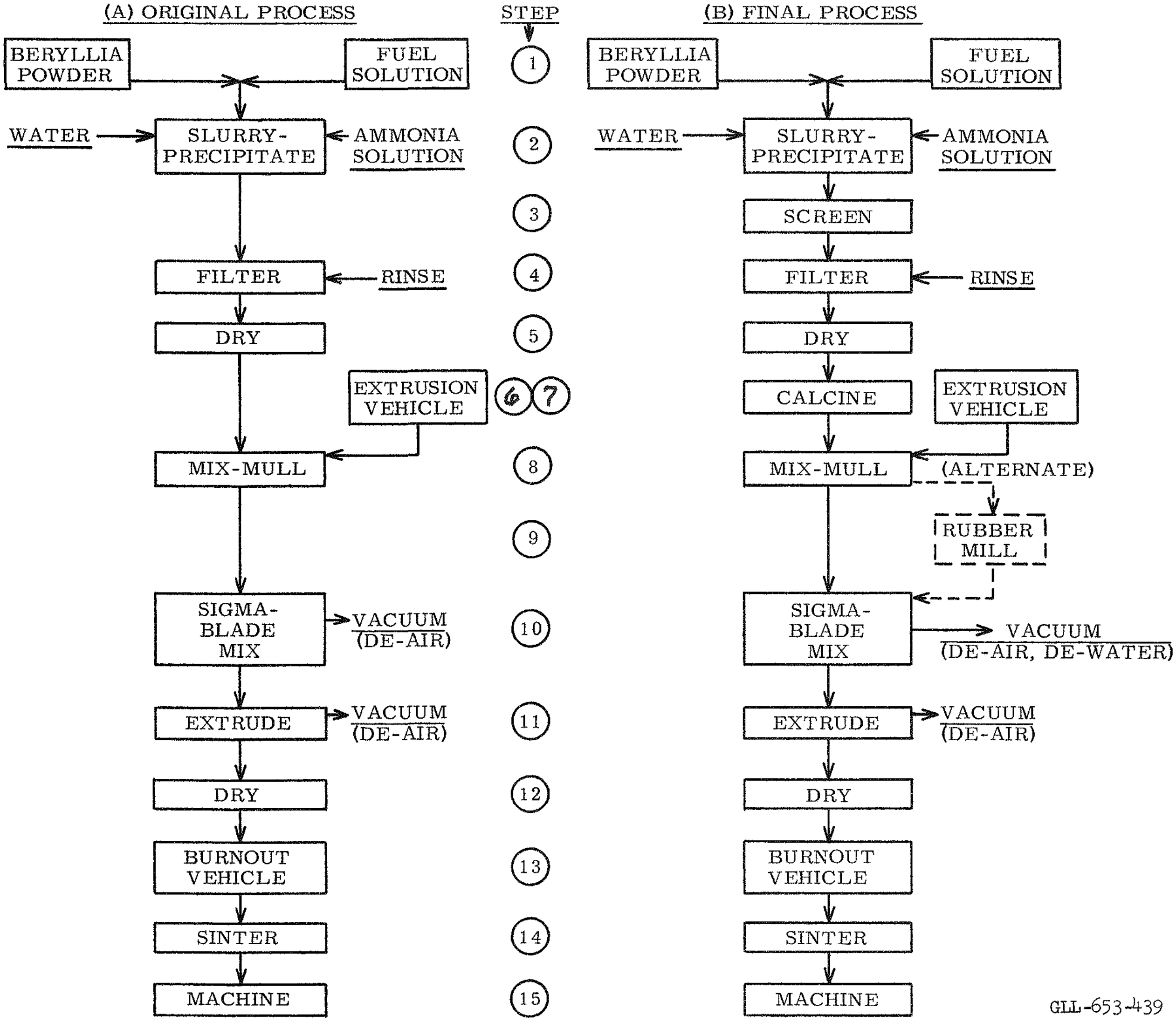


Fig. 2. Fuel element fabrication – LRL.

fires to about 2.97 g/cc density, and exhibits about 16% linear shrinkage as tested. Extruded and sintered UOX invariably shows preferred orientation, the c axes tending to align in the direction of extrusion.

2. Fuel Constituents

Production started with a fuel solution consisting solely of uranyl nitrate. Ultimately yttrium and zirconyl nitrates were added. These were precipitated with ammonia in the same manner as uranium alone.

C. Slurry-precipitation ("Slurpitation")

The first step in the process was the slurrying of BeO in deionized water. After 30 minutes of violent agitation, the slurry temperature rose to about 40°C and the powder was well dispersed.

At this point fuel solution was introduced to the slurry in amounts ranging from about 1 to 7% UO_2 by weight in the sintered piece. The fuel solution was a mixture of uranyl, zirconyl, and yttrium nitrates in acidified water. After thorough blending of the fuel solution with the slurry, ammonia solution was poured quickly into the mixture under violent agitation to precipitate the fuel components. The mix was taken to a pH of at least 9.5 to assure complete precipitation.

Experiments were run on coprecipitating the fuel before slurrying with beryllia, but the process reverted to "slurpitation" of the beryllia mixed in fuel solution at room temperature. Slurpitated mixes were more filterable and easier to make in the LRL equipment than the coprecipitated ones.

The elevation of mix temperature from mixing and chemical energy from the precipitation reactions was not controlled. No subsequent process variability has been attributed to the uncontrolled aspects of the slurpitation, however.

Criticality-safe requirements imposed a long, narrow geometry on the mix tanks. After some mixing studies, a baffle system was installed in the mix tanks and both paddles and screw-type agitators were mounted on the axially mounted vertical mixer shafts. Shaft speeds of 1000 rmp produced excellent mixing.

D. Screening

Because the UOX beryllia sometimes contained foreign material, it was desirable to screen the "slurpitate" before filtering. Small vibratory screens (50 mesh) were designed and installed at step 3 of the process with good results.

E. Filtering

The filters were cylindrical baskets of criticality safe geometry in which were inserted cylindrical paper filters.

Filtration was rapid and trouble-free. The filtration problems were mechanical, having to do with assembly, disassembly, and unloading of the filters rather than their process function.

Early in the history of the process, the rinse water was ammoniated to preclude the possibility of solubilizing some of the precipitated uranium with carbonate ion present in the rinse water.

F. Drying

Step 5 (Fig. 2) relied on electric rod heaters inserted vertically down the axis of the cylindrical filters for drying. Water vapor removal was by natural convection up the filter tube. Initially, these dryers were intended to completely dewater the filter cake in preparation for incorporating the extrusion vehicle. Finally, the rod heaters were used only to dry the filter cake sufficiently for clean removal from the filter. This change was made for several reasons. Of greatest importance was the acidity of the filter cake; low pH batches cracked more in extrusion than neutral batches. The low pH of the batches resulted from hydrolysis of ammonium nitrate which was not removed by rinsing. The rod heaters were incapable of pyrolyzing nitrates. In addition, the rod heaters did a nonuniform drying job in a non-reproducible manner.

G. Calcination

This step was added to the process to bring the filter cake to a uniform state for subsequent operations. After partial drying (step 5), the cake was charged in stainless steel cans to a tunnel kiln where it dwelled at 950 to 1000°F in air for about 8 hours. This dried the cake completely, pyrolyzed nitrogenous materials, and dehydrated the fuel precipitates to the oxides. The result was an essentially neutral blend of beryllia and fuel oxides with uniform processing characteristics.

H. Extrusion Vehicle

The extrusion vehicle was reformulated several times because of other process changes.

Before production operations began, a variety of vehicle constituents were examined. These included potato and corn starches, glycerine, dextrine, Carbowax 4000, Carbopol 934 (a starch), gelatin, sodium carboxymethyl cellulose, carboxymethyl cellulose, alginates, polyvinyl alcohol, polyvinylpyrollidone, ethyl cellulose, paraffin emulsion, polyvinyl acetate emulsion, stearic acid, and deionized water. The LRL plant went into production with a polyvinylpyrollidone-glycerine-water vehicle containing smaller amounts of polyvinyl acetate and paraffin.

Water content was reduced during the processing of each batch. Initial water content was set to give good plastic mixing properties. Final water content was set for good extrusion properties. The final water content in the mix was controlled as a function of the amount of fuel in the mix. A typical relationship between "total combustibles" (vehicle content) and fuel loading for a given range of extrusion pressure is illustrated by Fig. 3.

At termination of LRL development activities the vehicle composition was:

	BASIS: 100 parts beryllia	
Binder-Thickner:	Methyl cellulose (Dow 400 cps Methocel)	3 parts
Lubricant:	Colloidal graphite solids (Aquadag, 22% C in water)	5 parts
Plasticizer:	Water	48 parts
	TOTAL	56 parts

This vehicle was dried during batch mixing to about 29 parts water for extrusion. The vehicle ingredients were homogeneously blended before addition to the BeO.

The most significant development changes in the vehicle were elimination of water-insoluble organic constituents (e.g., polyvinyl acetate) and the substitution of a particulate lubricant (graphite) for liquid lubricants.

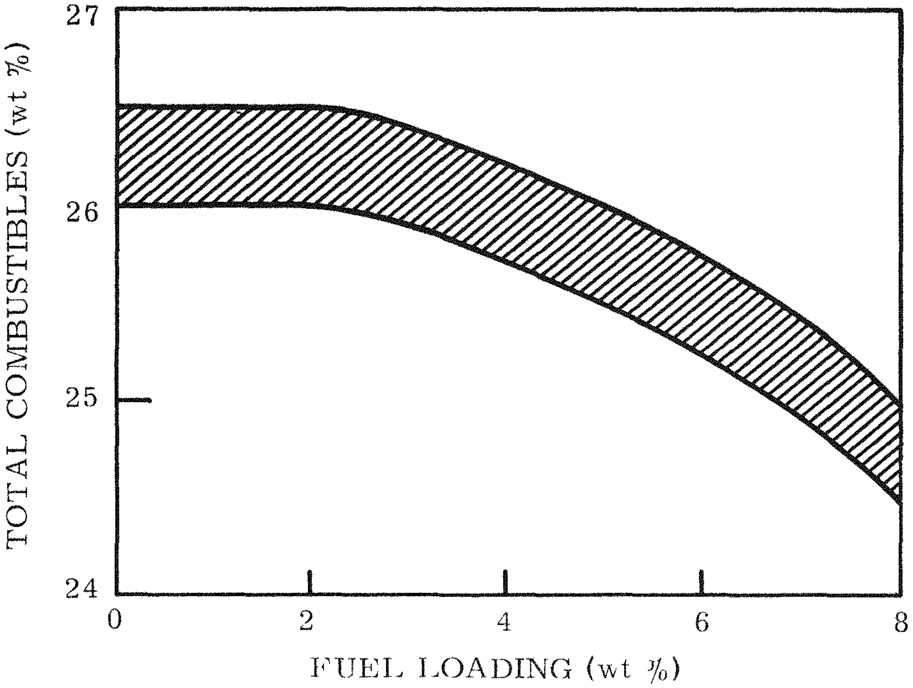


Fig. 3. Total combustibles vs weight percent fuel. Note: combustibles values between these curves result in ramhead pressures between 10,000 and 12,000 psi for coprecipitated UO_2 - ZrO_2 - Y_2O_3 fuel.

At the pressures used in later extrusions (around 20,000 psi) a binder was not essential. Beryllia was extruded with only water as the extrusion vehicle. The extrusion was plastic and had mechanical strength. It could be handled and fired to full density. It showed extreme dilatancy,* however, indicating that water was migrating under pressure. Methocel, then, acts not only as a binder, but also as a thickener, effectively increasing the viscosity of the water and retarding its migration.

A solid lubricant (i. e., graphite) was considered because it should be less migratory than liquids, thus staying in the areas where needed and not collecting at shear interfaces. If extruder performance is a criterion, this

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*The dilatancy was observed in extrusion as a pressure-rate phenomenon in which extrusion speed gradually slowed and stopped even at maximum available pressure. After a short period of pressure release, another length of material could be extruded before flow again slowed and stopped.

model is reasonable. Graphite-lubricated, Methocel-thickened batches gave the best ceramic surfaces, least surging in extrusion, and best dimensional control seen.

The extrusion vehicle represents a prime potential area of development for improving the process. Future development should include a comprehensive joint survey of possible binder-thickeners and placticizers which are not volatile under batch formulation and would preclude plasticizer evaporation, condensation, and transport in batch storage.

I. Mixing, General

At termination, mixing batches for extrusion was the most critical operation in the process. Future development of the process will be sensitive to improvements in producing mixed batches with measurable, uniform, predictable extrusion properties.

Mixing fixes the "extrudability" of the batch. The LRL mixing procedure was developed largely by "cut and try" methods using performance in the extruder and the quality of the extruded material as the criteria of mixing performance. Pitfalls in this approach are: (1) the difficulties in distinquishing between batch and extruder variables, and (2) the difficulties in ascribing tube properties to the batch or to the extruder — or both. Actually, the properties of the mix and the operating characteristics of the extruder are interrelated and not readily separable.

J. Mix-Muller

The muller consisted of a covered, flat-bottomed, stainless steel bowl about two feet in diameter in which two heavy mixing wheels rotate. It was used for breakup of calcined filter cake and primary mixing of extrusion vehicle with ceramic oxides. The muller was incapable of producing homogeneous mixtures and was difficult to clean. As a result, additional mixing was required (cf. section III K), and foreign inclusions from previous batches were sometimes noted. Future process development would probably eliminate this device from the process.

K. Sigma-Blade Mixing

This was the primary method of mixing extruder batches until late in the history of the process. The Baker-Perkins sigma mixer is a heavy duty mixer-kneader in which horizontal-axis blades mix, turn, knead, and shear the charge. It has an 18-quart nominal capacity, and is water jacketed and controlled at 20°C. It operates at 60 rpm on one blade and 30 rmp on the other and is equipped with a tight lid and a vacuum system capable of maintaining 28 inches of vacuum in the mixer. During the early stages of process development, the sigma was not recognized as a principal source of extrusion property variability. As the method of operation of the sigma evolved, however, its effects on the extrusion properties became evident. When storage conditions and working temperatures, times, and vacuum were brought under control, large fluctuations in batch extrudability disappeared.

Ultimately, the sigma cycle was divided into two sections. First, the wet mass (from the muller) was mixed for 60 minutes in a wet plastic state to produce a homogeneous mass. Then, the moisture content was drawn down slowly at fixed temperature and vacuum over an extended time (about 2 hours) to optimum extrusion consistency. These batches were fairly predictable in performance, but inhomogeneities persisted and were manifested in occasional surging from the extruder. It was shown that batch particles dried under vacuum were drier at their surfaces than inside and that small particles were drier than large. Small and uniform batch particle size was the goal. The batches were stored in controlled temperature and humidity to preclude inhomogeneity caused by water evaporating from the batch and recondensing within the container.

The introduction of the rubber mill (section III L) altered the role of the sigma mixer. It then became merely a device for breaking up and blending the rubber milled material.

L. Rubber Mill

This was a 5-horsepower laboratory mill consisting of a pair of parallel horizontal chrome-plated steel rolls with an adjustable gap between them. The rolls were counter-rotating at a speed ratio of 1.4/1, fixed roll/adjustable roll. The adjustable roll is run at 36 feet per minute surface speed. Both rolls were 6-inch diameter with 12-inch faces having 10-inch working space between guides. The rolls were run with about a 0.030-inch gap between them. They were internally cooled by water flow adjusted over a range of about 2 to 6 gallons per minute, depending on the amount of heat being generated in the batch.

The rubber mill, in limited trials, did the most intensive mixing job of any device tried in combining extrusion vehicle with ceramic body. The high shear and high pressure obtainable on the mill served to break down, impregnate, and work the batch rapidly and effectively. Milling had a marked effect on extrusion properties. Much drier materials were extrudable after milling than after sigma mixing. These dry materials, in the green extruded state, were dimensionally superior to the sigma-mixed batches.

With sigma-mixed batches, compacting and extrusion followed by rework and re-extrusion invariably led to lower extrusion pressures and more uniform rates. Similar effects were observed in rubber-milled batches, even though rubber milling does a more intensive mixing job than sigma mixing. The implication is that equilibrium mixing was not achieved before extrusion.

Continued development would include studies of roll speeds and spacing, mixing times and temperatures, and batch consistencies and controls.

M. Batch Consistency

The value of simple measurements of vehicle content and "plasticity" or "extrudability" of the extrusion mix was recognized. Such measurements were not easy to find or develop, however.

At the termination of LRL activities, a design for a so-called "pre-extrusion tester" had been completed. The tester was a miniature extruder instrumented to give precise material pressure and extrusion rate data. The tester's performance was to be correlated with that of the extruder to get reliable extrudability measurements on gram quantities of extrusion mix.

Other measurements were tried. In mid-1960, a number of experiments were run to correlate a simple penetrometer (Shore A-2) measurement with extrusion pressure. The correlation was not good. At that time the pH effect on batch consistency was unrecognized and was an uncontrolled variable. Retrospectively, a re-evaluation of penetrometer measurements as a potential simple in-process control would be justified. Penetrometer hardness measurements of the batch sheet on the rubber mill might be an effective method of following the course of the mill cycle.

An attempt was made to measure batch consistency in-process with an instrumented sigma-blade mixer. A standard Baker-Perkins mixer (size 4) was coupled to a Plastagraph, an instrumented drive for measuring and recording input torque to the mixer shaft. The device was to trace a running record of batch consistency as measured by the torque required to drive the mixer. This proved fruitless because of the nature of the mix which does not remain a continuous plastic mass throughout the mix cycle. As the mix dried, approaching extrusion consistency, it fragmented into a particulate mass (sand- or gravel-like). In this stage, the sigma mixer essentially tumbles the loose mass with little, if any, shear or mixing action on the separate particles. This tumbling action rendered the Plastagraph reading meaningless.

Control of batch consistency developed along other lines. The extrusion quality of the mix was judged largely visually. In general, the finer the particle size, the better the extrusion. The best extrusion batch was almost powdery. Accurate knowledge of the total vehicle content is essential. This was taken as a partial measure of batch consistency. With good control of the earlier steps in the process plus adequate temperature, time, and vacuum controls on the sigma mixer, water content of the batch was a reasonable indicator (±5%) of extrusion pressure. Through most of the LRL operations a "total combustibles" figure was obtained by burning out all volatile and combustible materials in a batch sample. This was accurate, but time consuming. Rapid means of accurately determining moisture content would be helpful in the process. The burnout of combustibles has been considerably expedited by redesign of the burnout facility, but it is still not a good in-process measure.

N. Aging and Storage

Mixed batches were stored in sealed polyethylene bags at a constant temperature of 72°F for not less than 24 nor more than 148 hours before extrusion. This permited the moisture content to equilibrate within the batch but avoided appreciable water loss from the batch.

O. Extrusion

Extrusion problems remaining at termination were well understood with the exception of curvature (camber).

Extrusion quality and extruder performance in the initial process were unpredictable. Improvements required study and involved other operations (especially mixing) previously discussed.

The axes of the extruder and the take-away belt were inclined about 10° from horizontal in early efforts to develop an air conveyor to air-float the extrusion through the drier. Part of the problems were peculiar to this near-horizontal extrusion.

1. The Extruder

The mixed batch was charged to a ram extruder and deaired for 1 minute at 30 mm Hg absolute pressure before extrusion. The extruder was capable of applying pressure up to 24,000 psi. The mix was forced through an annular opening whose cross-sectional dimensions were about 25 percent larger than finished tube size. Operating pressures ranged from 10,000 to 20,000 psi; higher pressures were required at startup.

The most persistent extrusion flaws were transverse cracks and camber. The cracking problem was complex and yielded bit by bit to a number of improvements. Vehicle improvements helped solve this problem; so did extruder improvement.

The greatest effort in extruder development was in die design, and marked improvements in extruder performance were realized thereby.

a. Die Parts, Mechanical Design and Fabrication. At first, the precision maintained in die part fabrication was inadequate to assure extrusion of fuel elements within specified tolerances. A program of die design, material and machining specifications, vendor selection, and part inspection was inaugurated to bring die part fabrication under adequate control. This extensive effort resulted in major improvements in the die system and an understanding of die parts fabrication which would permit further improvements in future die designs.

All die parts, with the exception of the forming die itself, were made from 4340 steel, chrome plated. The hexagonal dies were made of tungsten carbide (Kennametal K96).

The die assemblies used at LRL were assembled, installed, and clamped in position from the exit end of the extruder; that is, from the outside. Extrusion operating pressure tended to disassemble the die configuration; i.e., to separate mating surfaces. High assembly stresses were required to insure integrity of the die configuration under load. The final die design reversed this situation; that is, the working pressure forced mating surfaces together rather than apart. Figure 4 is an assembled view of this die design. Means are still needed for accurately centering the extrusion mandrel in the extrusion die. Although die parts were machined with the best tolerances achievable with good precision shop practice, most assemblies showed die-mandrel eccentricities in excess of 1 mil as a result of accumulated machine tolerances.

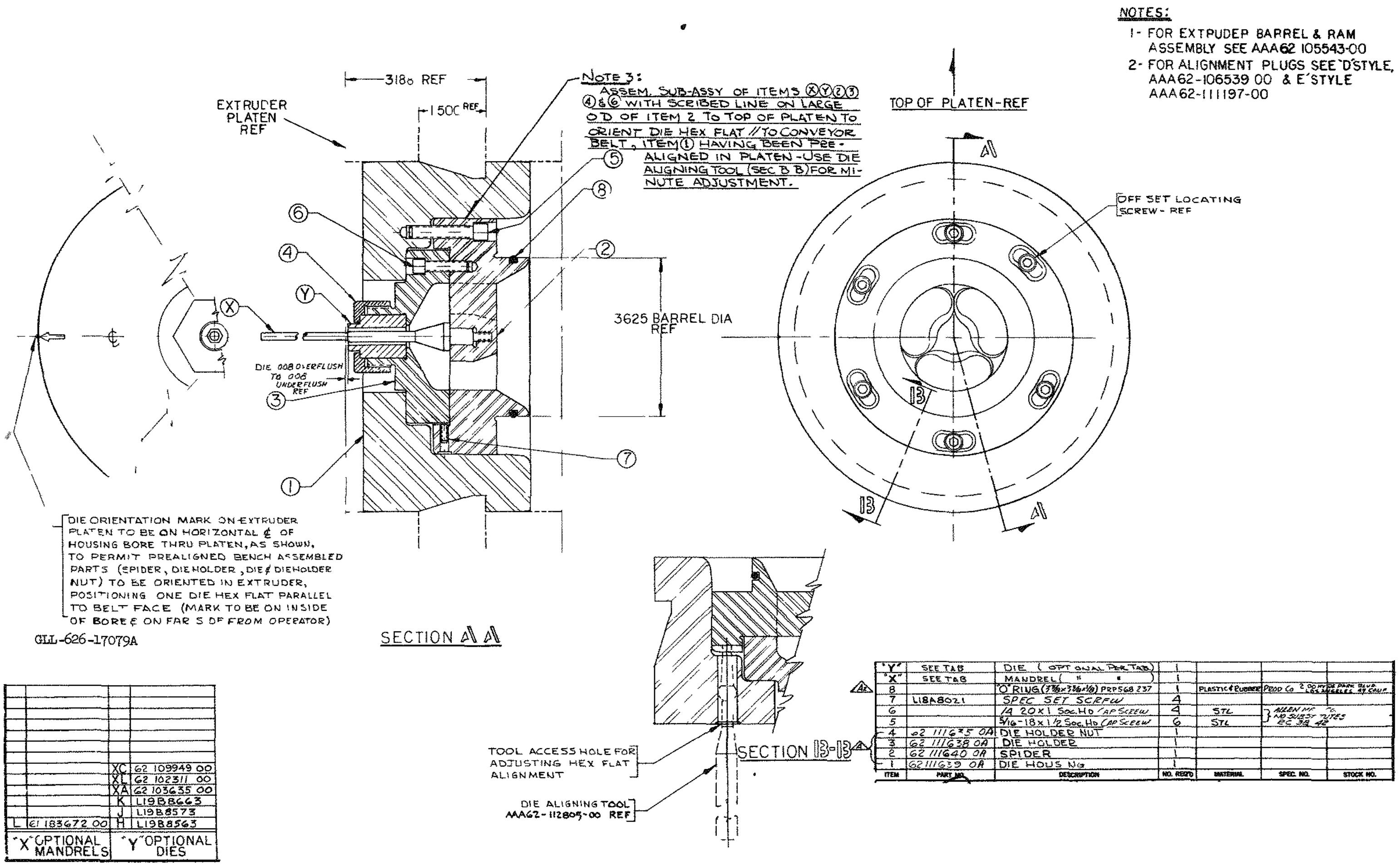
b. Die Parts, Processing Characteristics. Early experimentation showed that extrusion dies with short lands (1/4 inch) and long lands (2-1/2 inch) performed poorly. A land length of 1.342 in. was standardized (Fig. 5). The hexagonal corners in early dies were sharp (0.003 in. corner radius); feathering and cracking on the extrusion were observed with these dies. Improvement resulted when 0.025 inch corner radius (see Fig. 5) was adopted.

Die lands with smooth finishes (4 to 8 microinches) produced higher proportions of cracked tubes than dies with rougher lands. As a result, final specifications called for a 30 to 50 microinch finish on the die lands. No good hypothesis explaining this behavior was developed.

The reducing section from the extruder barrel to the die was part of the die holder. Early experiments with the size of included angle in this conical section indicated a 120 degree angle to be optimum.

The extrusion mandrel protruded far enough from the die to carry the extrusion to the take-away belt. Flexing and sag of the tube at this point was thus prevented. Mandrel, die holder, and other dimensions were fixed so that the extrusion always released from the finishing section of the mandrel before it released from the die, i. e., the mandrel diameter was reduced slightly upstream from the die exit. This was intended to reduce cross-flats dimensional variability at the cost of greater bore variability. If the external surfaces of the tube were to be ground after sintering anyway, it would be desirable to reverse the situation and accept greater variation across flats than in the bore.

c. Working Pressure. The original 75-ton extruder at LRL was capable of ramhead pressures of 12,000 psi. Developments showed that higher working pressures produced better extruded dimensions. A 120-ton extruder capable of ramhead pressures up to 24,000 psi was installed, and the higher pressures were an important factor in the development of the process. The best dimensional results were realized at extrusion pressures from 19,000 to 24,000



NOTES:

1- FOR EXTRUDER BARREL & RAM ASSEMBLY SEE AAA62 105543-00

2- FOR ALIGNMENT PLUGS SEE "D" STYLE, AAA62-106539 00 & E STYLE AAA62-111197-00

NOTE 3: ASSEM. SUB-ASSY OF ITEMS ⓍⓎ②③④ & ⑥ WITH SCRIBED LINE ON LARGE O D OF ITEM 2 TO TOP OF PLATEN TO ORIENT DIE HEX FLAT // TO CONVEYOR BELT, ITEM ① HAVING BEEN PRE-ALIGNED IN PLATEN - USE DIE ALIGNING TOOL (SEC B B) FOR MINUTE ADJUSTMENT.

DIE ORIENTATION MARK ON EXTRUDER PLATEN TO BE ON HORIZONTAL ℄ OF HOUSING BORE THRU PLATEN, AS SHOWN, TO PERMIT PREALIGNED BENCH ASSEMBLED PARTS (SPIDER, DIEHOLDER, DIE & DIEHOLDER NUT) TO BE ORIENTED IN EXTRUDER, POSITIONING ONE DIE HEX FLAT PARALLEL TO BELT FACE (MARK TO BE ON INSIDE OF BORE ℄ ON FAR S DF FROM OPERATOR)

"X" OPTIONAL MANDRELS		"Y" OPTIONAL DIES	
		XC	62 109949 00
		XL	62 102311 00
		XA	62 103635 00
		K	L19B8663
		J	L19B8573
L	61 183672 00	H	L19B8563

ITEM	PART NO.	DESCRIPTION	NO. REQ'D	MATERIAL	SPEC. NO.	STOCK NO.
"Y"	SEE TAB	DIE (OPTIONAL PER TAB.)	1			
"X"	SEE TAB	MANDREL (" ")	1			
8		"O" RING (3 3/16 x 3 3/16 x 1/8) PRP568 237	1	PLASTIC & RUBBER	PROD Co 2 00 HYDE PARK BLVD. LOS ANGELES 47 CALIF	
7	L18A8021	SPEC SET SCREW	4			
6		1/4 20 X 1 Soc. Hd CAP SCREW	4	STL	ALLEN MF Co. NO SUBSTITUTES EC 38 42	
5		5/16-18 x 1 1/2 Soc. Hd CAP SCREW	6	STL		
4	62 111635 0A	DIE HOLDER NUT	1			
3	62 111638 0A	DIE HOLDER	1			
2	62 111640 0A	SPIDER	1			
1	62 111639 0A	DIE HOUSING	1			

Fig. 4. Extruder die assembly.

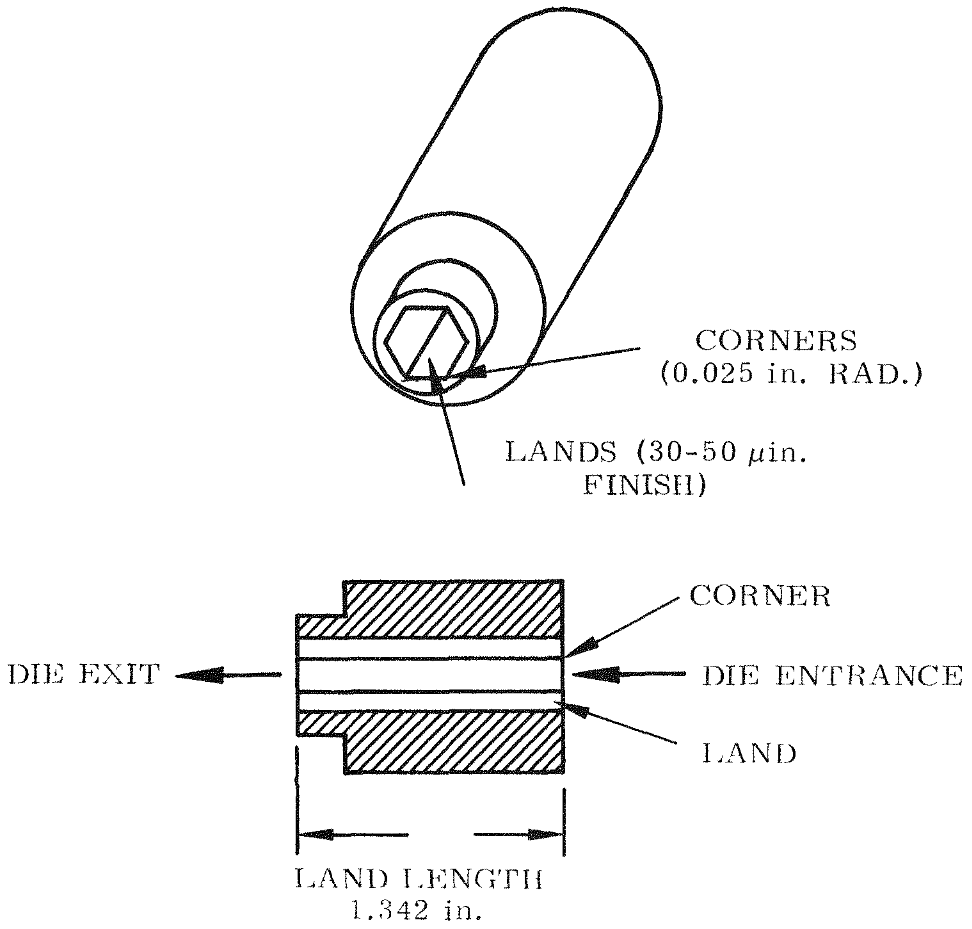


Fig. 5. Extrusion dies (no scale).

psi at the ramhead. These pressures with Methocel-graphite-water vehicle showed dimensional control approaching the limits of die and gaging accuracies; hence, a need for increased extrusion pressures is not foreseen.

2. The Tube Cutter

The original LRL process incorporated a camera iris type of cutter for scoring the tubes as they emerged from the die. This type of cutter was used until development terminated. It was improved, however. Initially, a stationary cutter was expected to score the moving tube and retract without noticably perturbing the material flow. The tube was "upset," however. That is, a section of each tube was expanded by compression of the moving extrusion against the momentarily stationary cutter blades. "Upset" was eliminated when a traveling cutter assembly was installed. The assembly oscillated parallel to the travel of the take-away belt. The cutting portion of the cycle was synchronized with the belt travel.

Minor improvements would make this cutter entirely adequate. The operator should be able to center the cutter iris on the extrusion axis and adjust depth of cut during operation. The cutter should be separately driven and timed more precisely with the belt travel.

3. Tube Take-Away

A vexing problem associated with horizontal extrusion was removal of extruded tubes from the extruder without damage or destortion. Substantial improvements in tube dimensional stability were realized from harder extrusions (higher extrusion pressures) and vehicle improvements. But the tube must be removed under stress-free conditions if dimensions are to be preserved. The interrelationships among batch, extruder, and take-away varibles were complex. The predominant transverse cracking flaw yielded only to improvements in all three areas. The same can be said of dimensional control.

In the initial process the take-away belt was driven at constant speed. Extrusion rate was matched to belt speed by manual adjustment, relying on visual observation. This was unsatisfactory because the operator was neither able to observe unacceptable mismatches of speed nor to compensate accurately for them. With a cross-flats tolerance band only 2 mils wide, any variation in belt speed which either compressed or stretched the extrusion by more than about 1% resulted in cross-flats distortion of about 2 mils. Sensitive automatic control was required. Three schemes were considered:

1. Speed matching
2. Tension control
3. Size control

The first two schemes were tried. In future development of horizontal extrusion the third method should be explored.

Speed matching was unsuccessful. The problems of sensing and measuring the extrusion rate accurately without distorting the tube virtually eliminated further consideration of speed matching. A "floating table" method was proposed as the most attractive scheme for sensing extrusion rate. It was proposed that the extrusion be picked up by a short "frictionless" belt section which would sense its speed and send a speed control signal to the main take-away belt drive. The system was built and tried but had a serious inherent shortcoming. This scheme assumed no mechanical connection between the main belt and the extruder and that the extrusion was a rigid body; neither of these assumptions was true. First, the extrusion itself made a direct mechanical tie between belt and extruder. Second, the extrusion was soft and plastic. These factors combined meant that any error in speed match which inadvertently entered the system tended to perpetuate itself. That is, for example, if the tube was inadvertantly stretched by the take-away belt (too high take-away speed), the speed sensing belt was then measuring the speed of the stretched tube (not the tube as extruded) and accordingly called for an erroneous take-away speed.

A bench model of a dancer roll system was designed and built to demonstrate that small (ounce) variations in belt tension could be sensed readily by the equilibrium position of a dancer roll. A tension control system was built which made dramatic improvement in dimensional control (see Fig. 6). The largest perturbing factor in the tension control system was the manual removal of tubes from the belt. The actions of snapping tubes at the score marks and removing them from the belt fed error signals into the tension control system. Further development of this tube take-away system would involve means for eliminating such error signals.

In controlling tension, a variable is removed which affects tube size, but tube size is not actually controlled. An improved system would operate on a size measure. A system is visualized in which the take-away belt would be speeded or slowed in response to an "oversize" or "undersize" signal from a continuous cross-flats measuring device. This would eliminate stretching (undersize) or compression (oversize) of the extrusion.

A minor problem remaining in the take-away system was the belt. Fiber glass belts were used exclusively, but they wore rapidly and their drag characteristics changed with time. This affected the tension control system.

Better functioning of the take-away belt would be realized if the drier (including the belt) were cut to minimum length for adequate drying.

4. Camber Control

A variety of identified causes produced camber in the extrusion. These include:

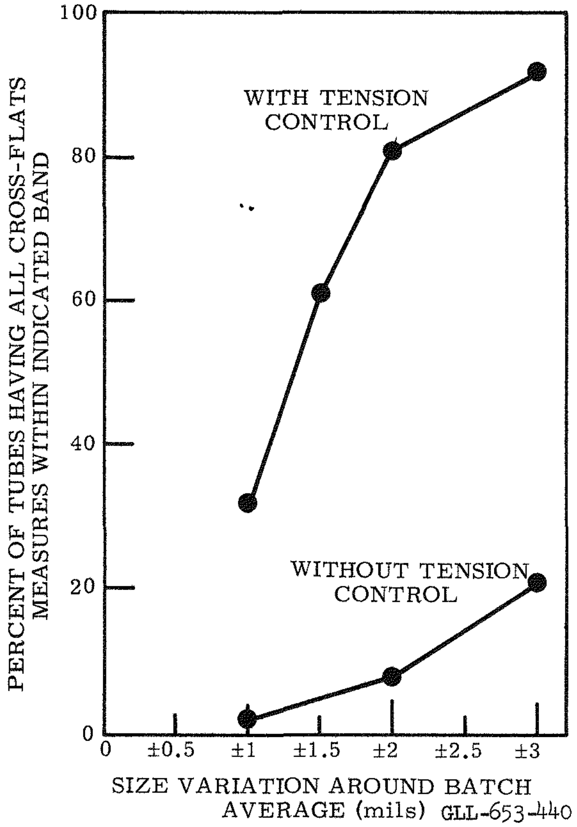


Fig. 6. Size control in extrusion with similar unfueled beryllia batches. These data are from sintered tubes.

a. Die-mandrel eccentricity.

b. Improper pickup of the extrusion by the take-away belt.

c. Too high tension on the extrusion. (In the extreme this will also cause cracking.)

d. Unbalanced drying of the extrusion.

e. Partial plugging of the extrusion die.

In addition, we speculate that diametrically asymmetrical flow patterns became established in the die system at times (for unidentified reasons) resulting in asymmetrical flow rates around the mandrel which in turn gave curvature in extrusion. Small perturbations of flow rate would produce marked curvature. For example, to produce 0.020 inch camber in a piece 6 inches long and 0.4 inch in diameter, the axial direction flow rate at the longest element would be only about 0.2 percent greater than that at the shortest element. (See Fig. 7.) Such a mechanism may account for the camber still observed in extrusion but not attributable to the recognized causes. Future development of the extrusion process should include a study of ultrasonic extrusion as a means of keeping the flow pattern symmetrical.

The problem of getting and maintaining concentricity of the extrusion die and mandrel needs study. Improvements resulting from closer machine tolerances were helpful. Nevertheless, reasonable shop tolerances and assembly problems place a practical limit on the concentricity which can be assured.. The plug gauges used were themselves tolerance limited. Methods are needed for closer gauging of concentricity.

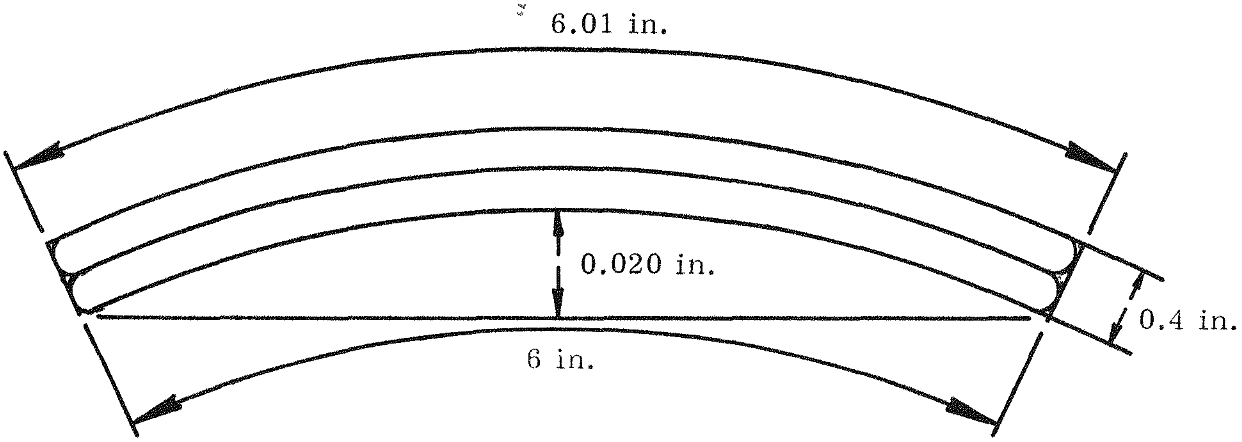


Fig. 7. Length of tube elements and camber. There is no scale and curvature is assumed circular.

Distortion resulted if the plastic extrusion either dropped or was lifted onto the take-away belt. This was eliminated by making the pickup section of the belt adjustable so that it could be precisely matched to the extrusion.

Successful operation of the tension control system eliminated tension curves and cracks in the extrusion. These flaws arose when the belt was over-driving, stretching the extrusion and causing it to crack and/or part unevenly at the scores. In this way, the first tube elements to part were stretched least and the last elements most, resulting in a curved tube.

5. Slump

When tube wall thickness was reduced about 25%, "slump" appeared in the extruded tubes. "Slump" was a distortion of the cross section in which the measurement between the vertically opposed pair of tube flats (as extruded) was consistently less than those between the other pairs of flats. In the final extrusions slump was not always evident, varying from 0 to about 0.5 mil.

P. Drying

The original drier in the LRL process included both radio-frequency and radiant heating. Radio-frequency heating was ultimately abandoned. A radiant tunnel drier divided into a number of zones with individual controls was installed. Partial drying, sufficient to permit handling, was accomplished in about 3 minutes dwell time at temperatures up to 375°F.

Q. Vehicle Burnout

This was a batch operation. Tubes were burned out in air at about 800°C for 3 hours. The temperature up-ramp took the tubes from room temperature to 800°C in about 7 hours.

R. Sintering

Tubes were fired in dry hydrogen between 1600 and 1750°C for 4 hours at temperature. The kilns were heated by tungsten resistance elements. A typical sintering cycle is illustrated in Fig. 8. The basic sintering conditions were prescribed by the ceramic properties of the fuel elements and were varied only slightly. Straightness within 3 mils over a 4-inch tube span was the goal in sintering and was achieved with 85 to 90% success.

Transverse shrinkage ranged from about 20% in unfueled tubes to about 23% in fueled tubes containing 7% UO_2. Axial shrinkage was slightly less.

1. Kiln Furniture

The sintering operation was used to straighten curved (cambered) tubes. Studies were made of kiln furniture geometries and the sintering cycle as it related to straightening.

The original LRL process was designed to sinter tubes standing on vertical pins, but camber distortion occurred during sintering. Molybdenum and tungsten kiln furniture were then designed to straighten tubes in sintering. The approach was to fire the tubes horizontally as they lay in straight grooves machined in molybdenum sheets. The grooves were V-shaped in cross section with a 120 degree included angle between the sides. Each tube was held in conformity with the groove by its own weight and the weight of a 3/16 inch diameter straight tungsten rod inserted in the tube bore. This effected considerable straightening. There were limitations due both to the kiln furniture and the shape of the unsintered piece, however. Slight creep and distortion of the grooved molybdenum plates were noticed in addition to normal machining aberrations; these factors did not seriously affect tube straightening, however. The overriding factors in tube straightening were the dimensional and ceramic qualities of the extruded tube. If a uniform green tube was cambered in a simple planar curve of long radius (15 mils camber over a 4-inch span implies a radius of curvature of about 133 inches), it could be straightened well

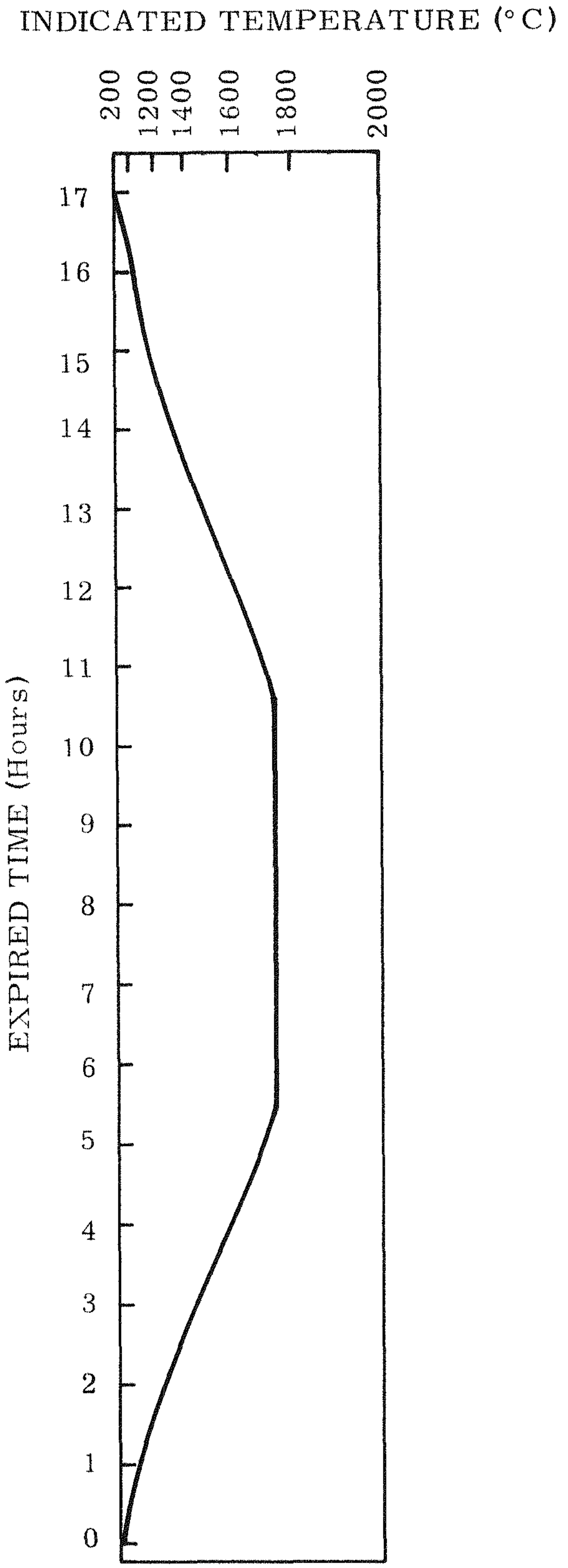


Fig. 8. Typical sintering cycle, batch kiln.

in sintering. If the green tube had compound curves, or undulating surfaces, or appreciable twist, or a sharp curve (even if restricted to a small fraction of the tube length), it was difficult to straighten.

The cost of cutting and maintaining straight and true V-grooves in molybdenum plates was high. For this reason, other geometries were attractive. One such geometry is a simple cylindrical sleeve. The sleeve is relatively easy to machine true and holds its shape well with firing. In this configuration the tube rests horizontally on a consecutive pair of its six edges with the intervening tube wall unsupported. This wall carries the weight of the tungsten straightening rod resulting in a slight bulging of the wall. The sleeve occupies considerable kiln space.

A rod configuration in which two adjacent faces of a tube were supported by a pair of parallel horizontal molybdenum rods was briefly evaluated. It had the sleeve's disadvantage of making only line contact with the tube, thus permitting the internal straightening rod to distort the cross section slightly, but it was conservative of kiln space and relatively light weight. It had the sleeve's advantage of being easy to machine, and functioned well.

2. Kiln Atmosphere

Throughout LRL operations dry hydrogen was used as the kiln atmosphere. In the ceramic insulated kilns the hydrogen picked up some moisture which raised its dew point. High dew points (50-60°F) resulted in poor densification of the fuel elements, especially for unfueled BeO. High dew points were not a problem in the small batch kilns which were all-metal and not ceramic insulated, however.

3. Tube Straightening

The straightening of tubes as it relates to kiln furniture, tube shape, and sintering cycle was studied with the following conclusions:

a. Tube straightening occurred rather abruptly over a narrow temperature range (about 1000 to 1200°C) before densification was well underway. (See Fig. 9.)

b. Most densification in fueled material occurred in the 1200 to 1600°C region. Little densification occurred in long soak periods at 1700°C or above. (See Fig. 10.)

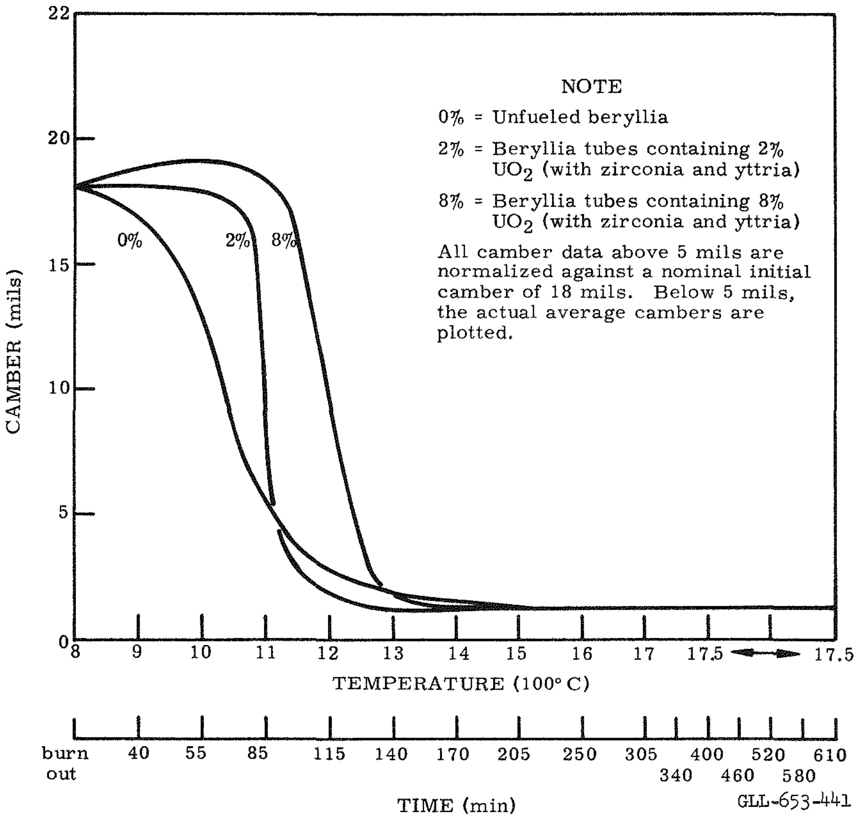
c. The presence of fuel altered both straightening and densification behavior. Fueled tubes started to straighten and densify at higher temperatures than unfueled, but both processes went more rapidly in the fueled material once they were started. (See Figs. 9 and 10.)

d. Straightness after firing was dependent on the weight of the rod inserted in the bore only below about 50 grams and independent of the rod camber at least up to 0.004 in of rod camber.

e. Straightening was nearly independent of groove cambers (at least up to 5 mils of groove camber over a 6-inch span).

f. Final sintered tube cambers were independent of unsintered tube cambers up to about 0.017 inch of unsintered camber over a 3-1/2 inch span.

g. Few tubes straightened to less than 1 mil camber over a 3-1/2 inch span regardless of the straightness of the groove and mandrel. Tube faces did not conform smoothly to the kiln furniture. Rather, depending on the number and kind of localized tube distortions, the tube flat actually contacted the furniture at only a few points. This implies that the dimensional quality of the tube must be inherently good on a point-to-point basis. Straightening in sintering was effective only on simple, planar, gentle curves, and then to a limited extent.



NOTE

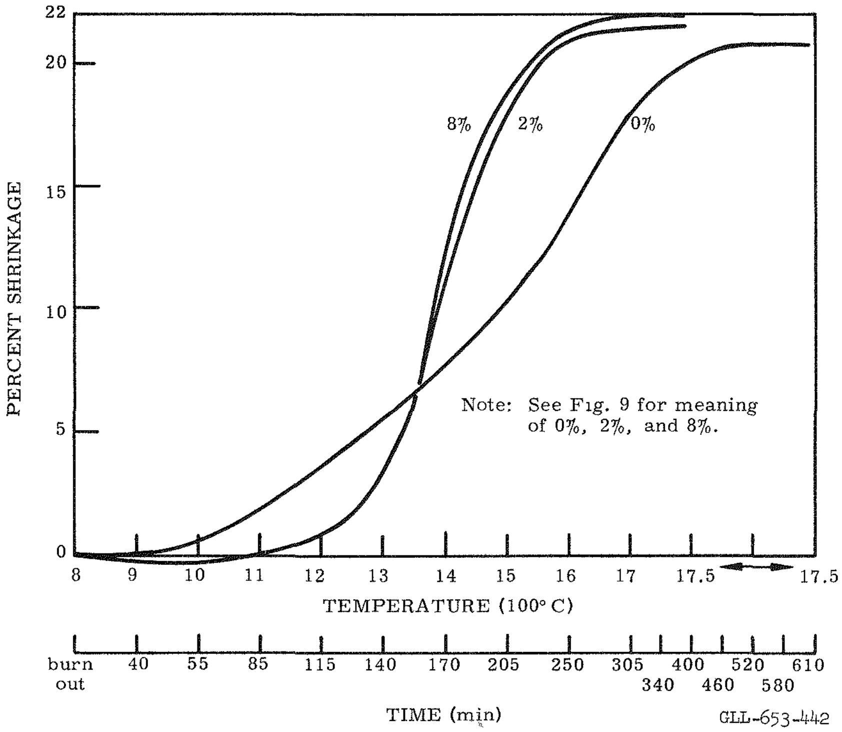
0% = Unfueled beryllia

2% = Beryllia tubes containing 2% UO_2 (with zirconia and yttria)

8% = Beryllia tubes containing 8% UO_2 (with zirconia and yttria)

All camber data above 5 mils are normalized against a nominal initial camber of 18 mils. Below 5 mils, the actual average cambers are plotted.

Fig. 9. Camber vs temperature in sintering.



Note: See Fig. 9 for meaning of 0%, 2%, and 8%.

Fig. 10. Cross-flat shrinkage vs sintering temperature.

h. After sintering, tubes were straightened by refiring under load. This was a cumbersome process, however. Weights of 80 to 130 grams per inch of tube length were required to reduce camber to less than 2 mils when tubes were refired at 1800°C for 1 hour.

i. As higher extrusion pressures became available, drier batches were extruded, yielding increased bulk green densities. There were indications that the more dense tubes were difficult to straighten. Green density can be lowered without sacrificing extrusion pressure by incorporating additional burnable solids (e.g., graphite) in the vehicle.

S. Machining

Work at LRL included no experience in machining of sintered fuel elements. Brief experience with the grinding of green tube surfaces was encouraging. Tubes as extruded and dried were readily ground to yield good sintered surfaces. The tooling and procedures for doing this on a production scale were not considered, however. The value of green machining is dependent on uniform green density in the tube. That is, if a piece of uniform dimensions has local variations in green density, and is sintered to a uniform density, unequal local shrinkages will result in dimensional distortions. Until uniform green density is assured, development of green machining as a process cannot be justified.

T. Scrap Recovery

Little was done on scrap recovery during LRL operations. Scattered attempts were made to recycle materials within the process without using special grinding or processing equipment. With the final extrusion vehicle (graphite-methylcellulose, see above), an unextrudable batch was rewetted and extruded with excellent results.

The press of processing problems on virgin material prevented scrap recovery studies at LRL. Scrap recovery was recognized as essential to ultimate process economy, however. Not only must the scrap grinding process be developed, but the processing characteristics of the resultant product must be determined.

The conceptual simplicity of reconstituting dry extruder scrap (without removal of the vehicle) urges further consideration of this possibility. In the past, the mixing operations were inadequate to break up and rewet dry extruder scrap. A further difficulty was that the vehicle formulations contained materials which were not rewettable or redispersible to their initial state. A number of approaches suggest themselves:

1) Intensified mixing action in rework.
2) Intensified wetting action.
3) Vehicle reformulation with more wettable constituents or with nonvolatile materials.

The rubber mill should be explored for reworking batches. Autoclave rewetting under elevated temperature and pressure should be explored.

A completely nonvolatile vehicle would probably eliminate the rework difficulty.

IV. TERMINAL STATUS

At the time that this work was terminated, the process was not fully developed. The directions for continued development were well defined, however.

The final extrusion at LRL (extrusion 1463) was the best effort. In Fig. 11 is shown the close cross-flat dimensional control achieved in this final batch. Note that dimensional control is not as close after sintering. Even so, more than 80 percent of the sintered tubes had an hexagonal cross section true within ±1 mil of the average; 95 percent were true within ±1.5 mils of the average. These figures can probably be improved further with continued development of mixing and extrusion techniques.

Both internal and external tube surfaces in Extrusion 1463 were of excellent quality. Examination with fluorescent surface penetrant revealed very few flaws. Radiographic examination showed inclusions. These were not attributable to process parameters but to contamination from previous batches or extraneous materials.

The principal problem remaining in LRL tube fabrication was camber. A maximum camber curve for extrusion 1463 (Fig. 12) shows that only 78% of the tubes were cambered less than 5 mils. Straight extrusions will yield straight sintered tubes. Development work would still be needed to insure straight extrusions.

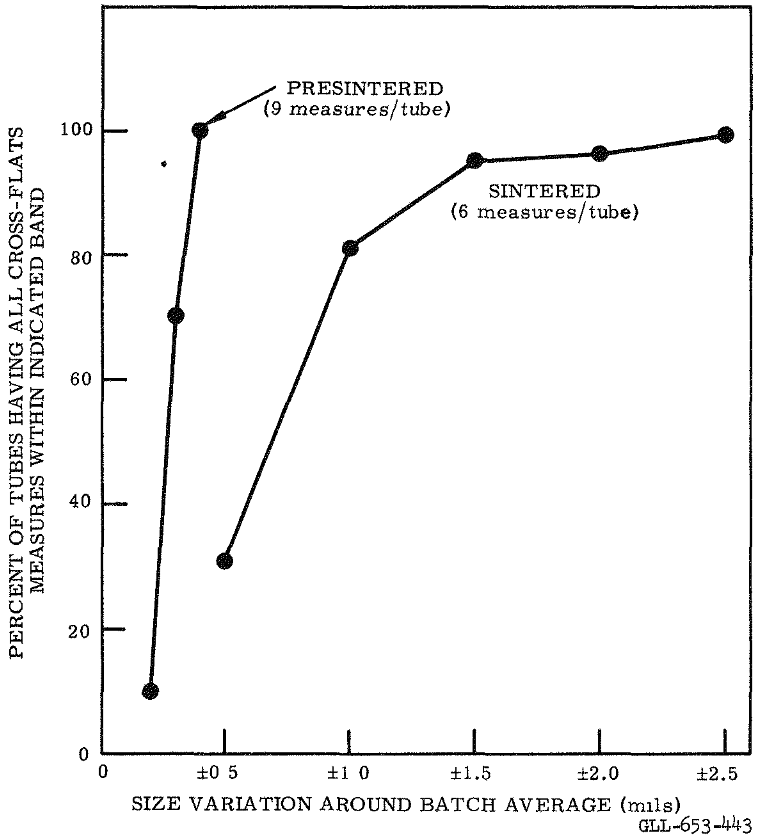


Fig. 11. Size control, unfueled beryllia extrusion 1463.

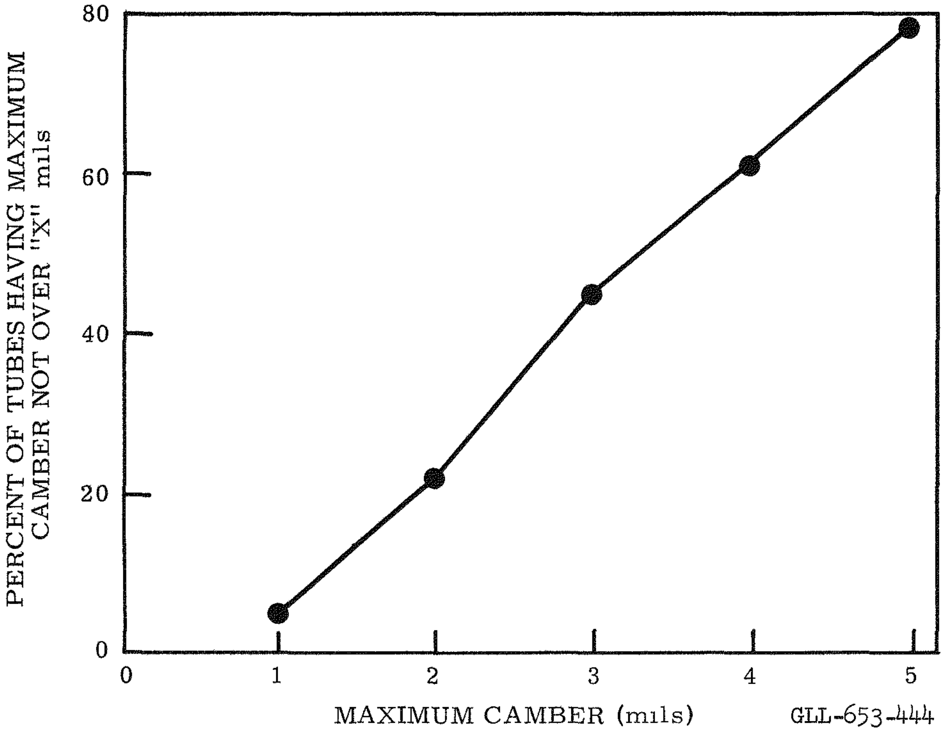


Fig. 12. Camber distribution, extrusion 1463.

V. CONCLUSION

A standing goal in LRL extrusion process development was the production of fuel elements on size without grinding. Substantial progress was made in that direction and continued development would see additional progress. At this point, however, insufficient data exist to predict the optimum balance between the costs of precision in the extrusion process and machining costs.

VI. BIBLIOGRAPHY

Most of the work described in this report is discussed in more detail in the Pluto Programmatic Report I (UCRL-5515) and Pluto Quarterly Reports, beginning September 1959 (UCRL-5699).

More background information on the properties of fuel elements may be found in:

(a) UCRL-7036, pt. 3, "Technology of Ceramic Components," by A. J. Rothman, August 1962.

(b) UCRL-6941, "The Pluto Program," June 1962.

(c) UCRL-7272, "Tory II-C Safety Analysis Report," April 1963, ed. by E. Goldberg.

(d) Other forthcoming UCRL reports on properties and behavior of Pluto fuel elements.

VII. APPENDIX

Outline of Raw Materials Properties Specified for the Pluto Fuel Element Fabrication Process

1. Beryllia. Loss on ignition, chemical purity (especially sulfur and nuclear poisons), powder uniformity, sinterability, and shrinkage were specified.
2. Aqueous uranyl nitrate. Concentration, free acid, chemical purity, and suspended solids content were specified.
3. Aqueous yttrium nitrate. Concentration, free acid, chemical purity, (especially nuclear poisons), and suspended solids content were specified.
4. Aqueous zirconyl nitrate. Concentration, free acid, chemical purity, (especially nuclear poisons), and suspended solids content were specified.
5. C. P. reagent grade ammonium hydroxide solution and deionized water were also used in the process.
6. Low ash organic materials and collodial graphite were used in the extrusion vehicles.

VIII. ACKNOWLEDGMENTS

The LRL process was initially designed and developed by J. B. Cahoon, Jr. Others who contributed to its development were G. D. Snider, R. N. Tully, J. N. Dexter, L. D. Christensen, R. L. Clendenen, A. J. Haecker, Jr., B. Rubin, and the writer.

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