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URANIUM-PLUTONIUM MIXED-OXIDE FUEL FABRICATION FOR THE DEUTERIUM CRITICAL ASSEMBLY "DCA" IN JAPAN

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H. AKUTSU, T. MUTOH, K. KAWASHIMA, K. NARUKI

Tokai Works,
Power Reactor and Nuclear Fuel Development Corporation,
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Abstract

The construction of the prototype heavy water reactor "FUGEN" is currently progressing on the schedule to be critical in 1976. Prior to this reactor operation, the reactor physics examination are carring out with the Deuterium Critical Assembly "DCA" since 1973. A new facility, the Plutonium Fuel Fabrication Facility (PFFF) of PNC which is mainly devoted to mass-production, was constructed to meet the plutonium fuel fabrication requirement for PNC's FBR and ATR projects.

At the "ATR line" of this PFFF, the fabrication of the plutonium fuel elements for DCA had been carried out since Mar. 1972, and 92 fuel assemblies with the total mixed-oxide amounting to as much as 10 tons were completed in Feb. 1973.

It was the first experience in Japan that, such a large amounts of mixed-oxide was fabricated as the fuel assemblies composed of three kinds of fuel rods according to both the isotopic charactor and the plutonium enrichment, totaling about 2600 fuel rods.

This fabrication program completed on the schedule demonstrating the designed capacity of the facility and gave us much

information and experiences concerning the production technology of mixed-oxide fuel, which form the solid base to the next program of the production of about 18 tons of mixed-oxide fuel as 100 assemblies for the prototype reactor "FUGEN".

The experiences and some informations obtained during the production works for DCA fuel fabrication are described in this report.

1. Introduction

The Power Reactor and Nuclear Fuel Development Corporation (PNC) is constructing a prototype heavy water reactor, named "FUGEN". One of the features of this reactor is an applicability of a "Plutonium Self-Sustaining Cycle", in which natural uranium enriched with plutonium (recovered from its own spent fuel) is to be fueled. Use of plutonium not only gives relief to an enriched uranium burden in a fuel material, but also lessens the problem caused by the positive void coefficient in reactor operation. Thus, since plutonium plays an important role in this type of the reactor, especially from the view point of reactor safety and its economy, many development works are being performed on the reactor physics with a "Deuterium Critical Assembly (DCA)".

For this DCA works, about 10 tons of mixed-oxide fuels were fabricated at the ATR line of the Plutonium Fuel Fabrication Facility of Tokai Works, PNC, in the period from Mar. 1972 through Feb. 1974. It was the first experience in Japan that such an amount of mixed-oxide fuel had been produced. Thus, this fabrication work gave us great experience and information concerning the production of mixed-oxide fuel, which lead us to form the confidence for the production of about 18 tons of mixed-fuels for "FUGEN". This paper describes the outline of works for DCA fuel fabrication.

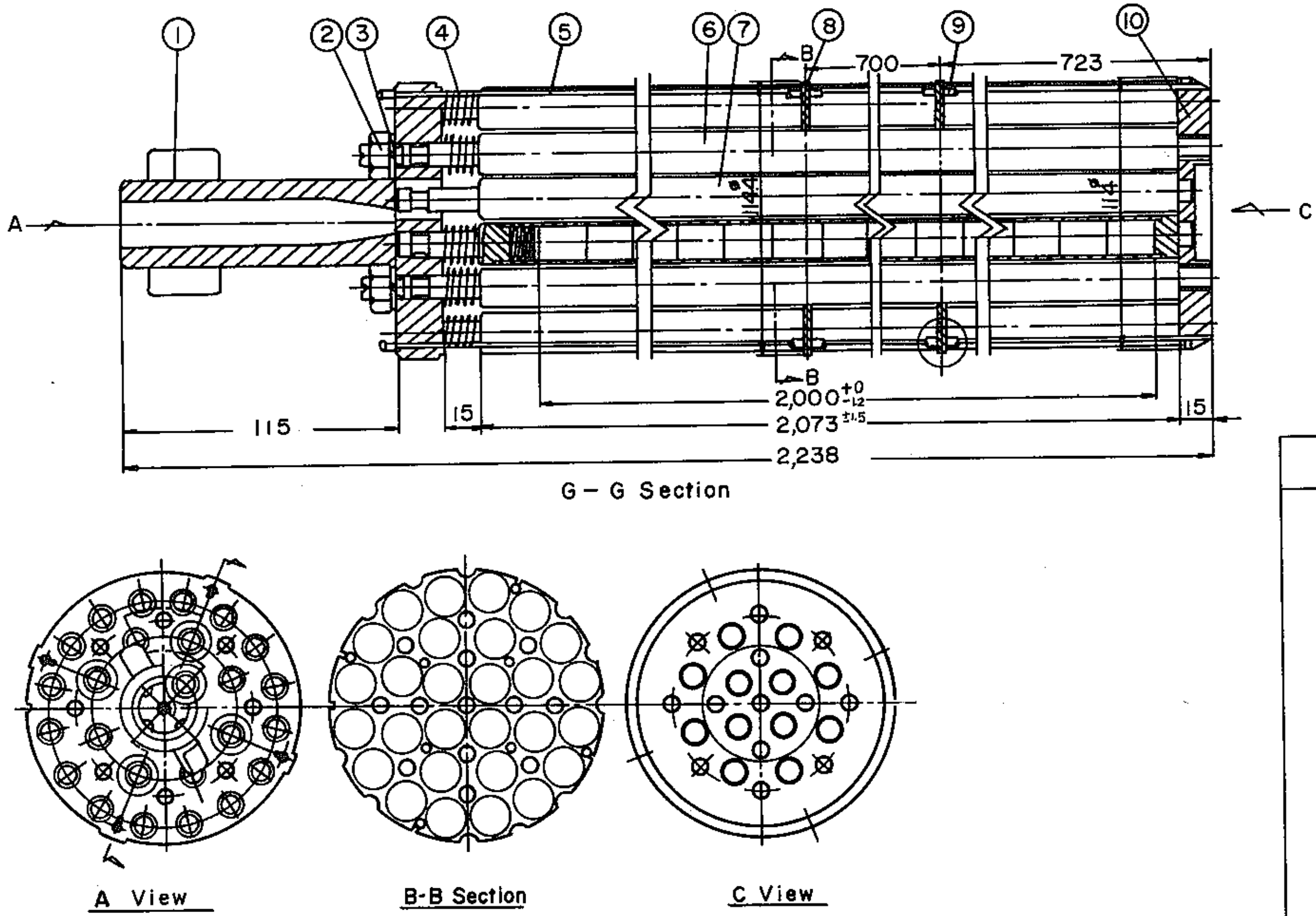
2. Configuration and Specification of DCA Fuel

Fig. 1 shows the design of the fuel assembly. Twenty-eight fuel rods are assembled in cluster to form the inner layer of four rods, the middle layer of eight rods and the outer layer of sixteen-rods, and are secured with both the upper and the lower tie-plates. The fuel rod is about 2.1 meter long and employs a zircalloy-2 cladding tube, in which mixed-oxide pellets of 14.8 mm diameter and 20 mm length are loaded to form the stack of 2000 mm length (Fig. 2). Three kinds of fuel rods are required to complete an assembly; twenty-three F-type rods (for Free standing), four T-type rods (for Tying) and one S-type rod (for Shielding). These free kinds of fuel rods are differentiated with the shape of end plugs of rods as shown in Fig. 2. For special assemblies, two D-type rods (Divided rods) were used in place of standard length rods (undivided rods).

The DCA fuel assemblies are grouped into three kinds (5S, 8S and 8R) according to both the isotopic character and the content of plutonium used for mixed-oxide pellets. In total, thirty-eight assemblies of DC 5S type, twenty-seven assemblies of DC 8S type and twenty-seven assemblies of DC 8R type were fabricated as shown in Table 1.

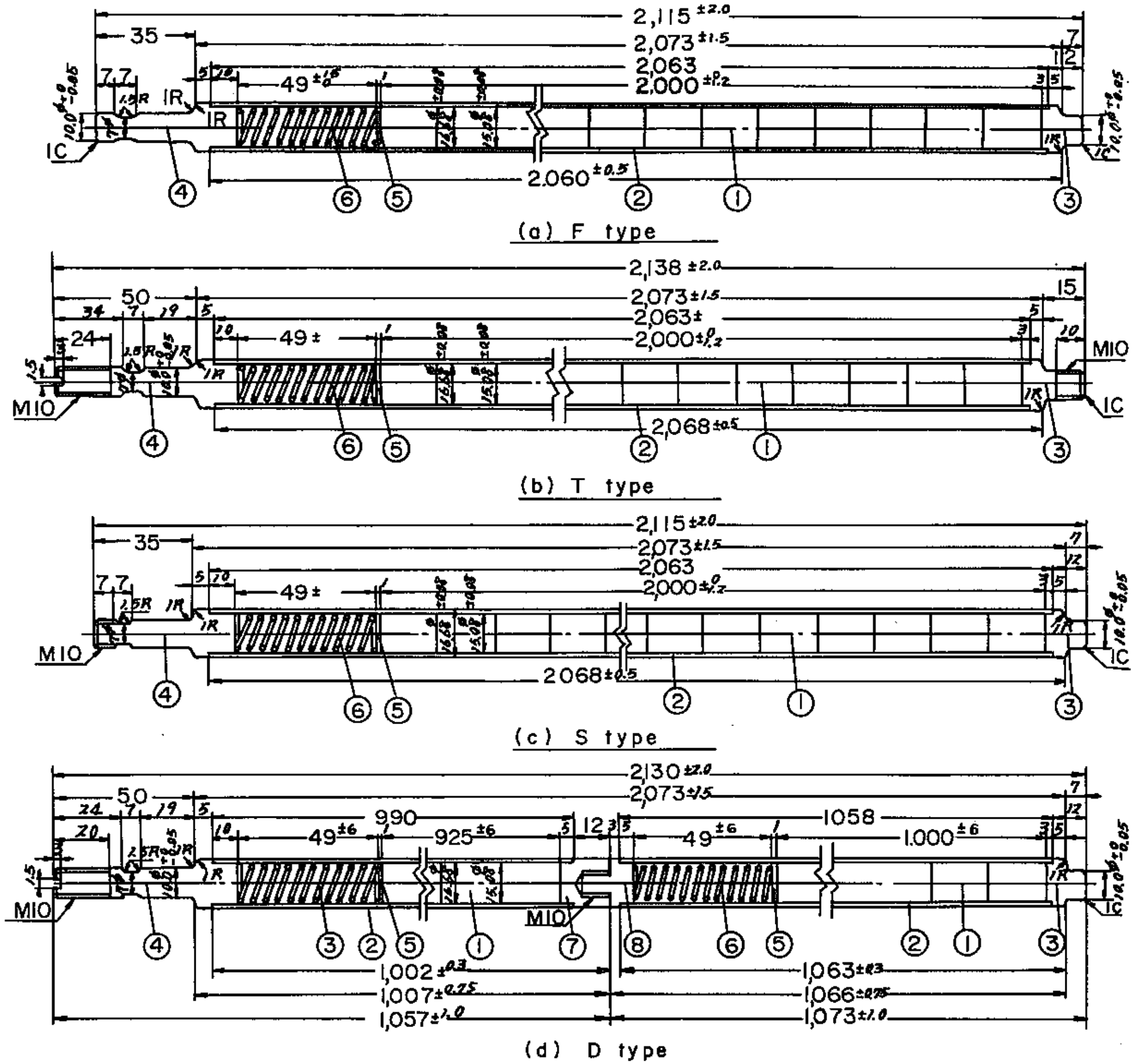
Table 1 also shows the production schedule for the DCA fuel. The characteristics of plutonium used is shown in Table 2.

Specifications for both pellet and rods are shown in Table 3 and Table 4 respectively. The shape of pellets is cylindrical, and the specifications for some factors are not so strict as that of fuel rods for a power reactor.



No.	Article
①	upper tie plate
②	hexagonal nut
③	Spring washer
④	Spring
⑤	Hunger wire
⑥	T type rod
⑦	S type rod
⑧	Spacer
⑨	Spacer rig
⑩	lower tie plate

Fig. 1 Design of DCA Fuel Assembly



No	Article
①	fuel Pellet
②	cladding
③	lower end plug
④	upper end plug
⑤	s. s. plate
⑥	plenum spring
⑦	Connective end plug (1)
⑧	〃 (2)

Fig. 2 Design of DCA Fuel Rods

Table 1 Mixed Oxide Fuels Shipped to DCA

	PuO_2 Content	Charactor of Pu	Number of assemblies	Number of fuelrods	Amount of pellets(kg)	Production schedule
DC 5S	0.54 w/o	Standard Grade	38	1094	3745	1972 Mar.~ 1973 Feb.
DC 8S	0.87 w/o	Standard Grade	27	792	2700	1973 Jan.~ 1973 July
DC 8R	0.87 w/o	Reactor Grade	27	786	2665	1973 Aug.~ 1974 Feb.
Total			92	2672	9110	

Table 2 Isotopic Ratio and Surface Dose Rate of Plutonium

Character of Pu	Lot No.	Pu238	Pu239	Pu240	Pu241	Pu242	Surface Dose Rate*	
							γ -ray	Neutron
Standard G.	8202	0.02	90.21	8.76	0.94	0.07	740 mrad/hr.	1.7 mrem/hr.
Reactor G.	8256	0.84	65.01	21.71	9.44	3.00	3500 mrad/hr.	5.4 mrem/hr.

* Average calculated value for the period of production.

Table 3 Specification of Mixed-oxide Pellets for DCA

1. Pu content	0.54 ± 0.03 w/o 0.87 ± 0.03 w/o as $\frac{PuO_2}{PuO_2 + UO_2} \times 100$
2. Content of impurities	E.B.C.; less than 2.35 ppm Al < 500 ppm Ni < 300 ppm Cr < 100 V < 10 Mo < 200 Cd < 1.0 Sn < 10 Mn < 20 B < 1.5 Si < 500 Fe < 500 Zn < 50
3. O/M ratio	2.00 ± 0.03
4. (Pu+U) content	Measured value
5. Density	10.25 ± 0.20 g/cc
6. Dimension	Diameter : $14.8^{+0.1}_{-0.2}$ mm Height : 20 mm
7. Defect	As regulated in ASTM B353-64T Grade RA-1

Table 4 Specifications of Fuel Rod for DCA

1) Appearance of welded part	① under cutting; less than 0.06 mm ② weld diameter; less than 0.08 mm increment from clad diameter ③ colouring; faint red and brown is allowed
2) X-ray radiograph of welded part	① porosity, inclusion; less than 0.5 mm ϕ ② penetration; more than wall thickness ③ thinning of wall; less than 0.16mm
3) Dimension	As indicated in Fig. 2
4) He leakage	less than 10^{-8} STD cc/sec.
5) Surface contamination	loose contamination; less than 10 dpm/dm^2 fixed contamination; less than 600 dpm/dm^2
6) Surface condition of rod	no harmful defects

3. Production of Pellets

Fig. 3 shows the production process of mixed-oxide pellet. The production process features two step blending of plutonium dioxide powder and natural uranium powder, which ensures the macroscopic homogeneity of plutonium distribution in the mixed-oxide powder of low plutonium content. At the first step, 15 kg of 15w/o PuO_2-85w/o UO_2 mixed-oxide powder was compounded by ball-milling, and then this powder lot was diluted into the mixed-oxide powder of low plutonium content by addition of natural uranium dioxide powder. The diluted mixed-oxide powder (27 kg in weight) was agitated for 15 mm in "VI-blender" and then ball-milled for 4 hours in the large rubber-lined pot (60 liter in capacity) to attain uniform plutonium distribution.

Granulation of the mixed-oxide powder was mostly done by a wet process, in which the powder was granulated with the HPC (Hydoxy-Propyl Cellulose) organic binder added as water solution into the powder being agitated in a kneading machine. Size and strength of the granules were some-what controlled by varying both agitating time and the amount of HPC binder. In the later period of the production, the ordinary dry granulation process and even the non-granulation process were successfully applied in 3 lots and 18 lots production respectively. The granulated powder was screened to minus 14 mesh and dried under nitrogen gas stream. Then an organic lubricant ("Stereotex") by 0.3 to 0.5 percent in weight was mixed with the dried granular powder by dry-blending. Pelletizing of the powder was conducted with a mechanical press at the pressure of 2.5 to 3.5 ton/cm^2. Dewaxing and sintering of the pellets were done in 27 kg batch size with the respective

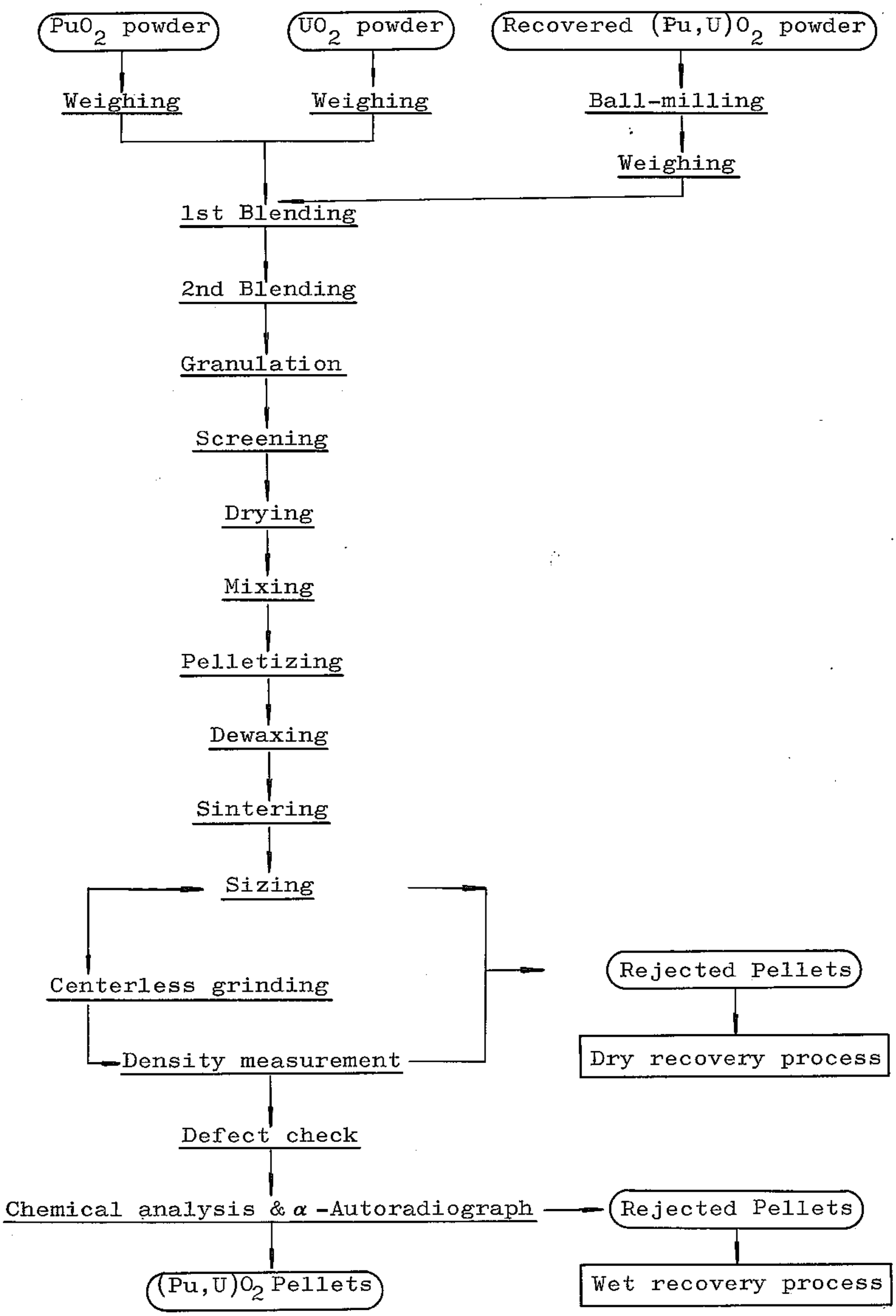


Fig. 3 Flowsheet of Pellet Preparation Process

furnaces of cylindrical muffle type under the flowing mixed gas ($5\%H_2$-$95\%N_2$). While dewaxing was done by heating pellets at 800°C for two hours, sintering condition was varied between 1600°C and 1700°C in the highest sintering temperature, and also between 2 hrs and 3 hrs in keeping time at the highest sintering temperature. Then, every sintered pellets went through a pellet size selector, and the pellets with large diameter were proceeded to a centerless grinder in order to adjust their diameter. However, in the later period of the production, we could omit the centerless grinding step since technical improvement made possible the control of the diameter in as-sintered pellets. Density of sintered pellets was measured by geometric method. Distribution of the densities for every production lot was evaluated following the sampling inspection designated in JIS-Z-9003. By this inspection, 60 pellets were taken at random from about 800 pellets of one sintering lot. Inspection of all pellet of one sintering lot was applied to the lots rejected by sampling inspection.

Followings are primary information regarding the properties of mixed-oxide pellets for DCA.

(i) Plutonium content was controlled within 0.02% in absolute value. Fig. 4 shows PuO_2 content in some production lots of DC 8R series.

(ii) Sizes of plutonium spots observed by α-autoradiograph were less than 300 μm and distributed mostly between 20 μm and 200 μm.

(iii) To the total 9660 kg pellet produced in 432 lots, sintered pellet densities were controlled with the standard deviation of 0.06 g/cm^3 in each sintering lot and of 0.07 g/cm^3 in whole sintered pellets. Fig. 5-1 and Fig. 5-2 show respectively the distribution of densities and diameter of as-sintered pellets in DCA 8R series pellets, which is expressed by number of lots vis average pellet density calculated for 60 sample pellets. Fig. 6 shows the density variation for some production lots in DC 8R series, in which dry granulation or nongranulation processes were applied. Rather high pellet densities were obtained by these processes.

(iv) Concerning the impurity content of pellets, metallic impurities are as described below except two rejected sintering lots which showed high Mo content due to oxidation of molybdenum boat by air contaminated N_2-H_2 gas.

	Al	B	Cd	Cr	Fe	Mn	Ni	Si	V	Zn	Mo	Sn
Spec.(ppm)	500	1.5	1.0	100	500	20	300	500	10	50	200	10
Observed (ppm)	<25	<0.3	<1.0	<10	<140	<6	<10	<400	<10	<50	<75	<10

Relatively high silicon content was caused by the silicon-rubber linning of the ball mill vessel. Fig. 7 shows the impurity content by equivalent boron content of DC 8R series pellets, in which the equivalent boron content was calculated to the metallic impurities mentioned above.

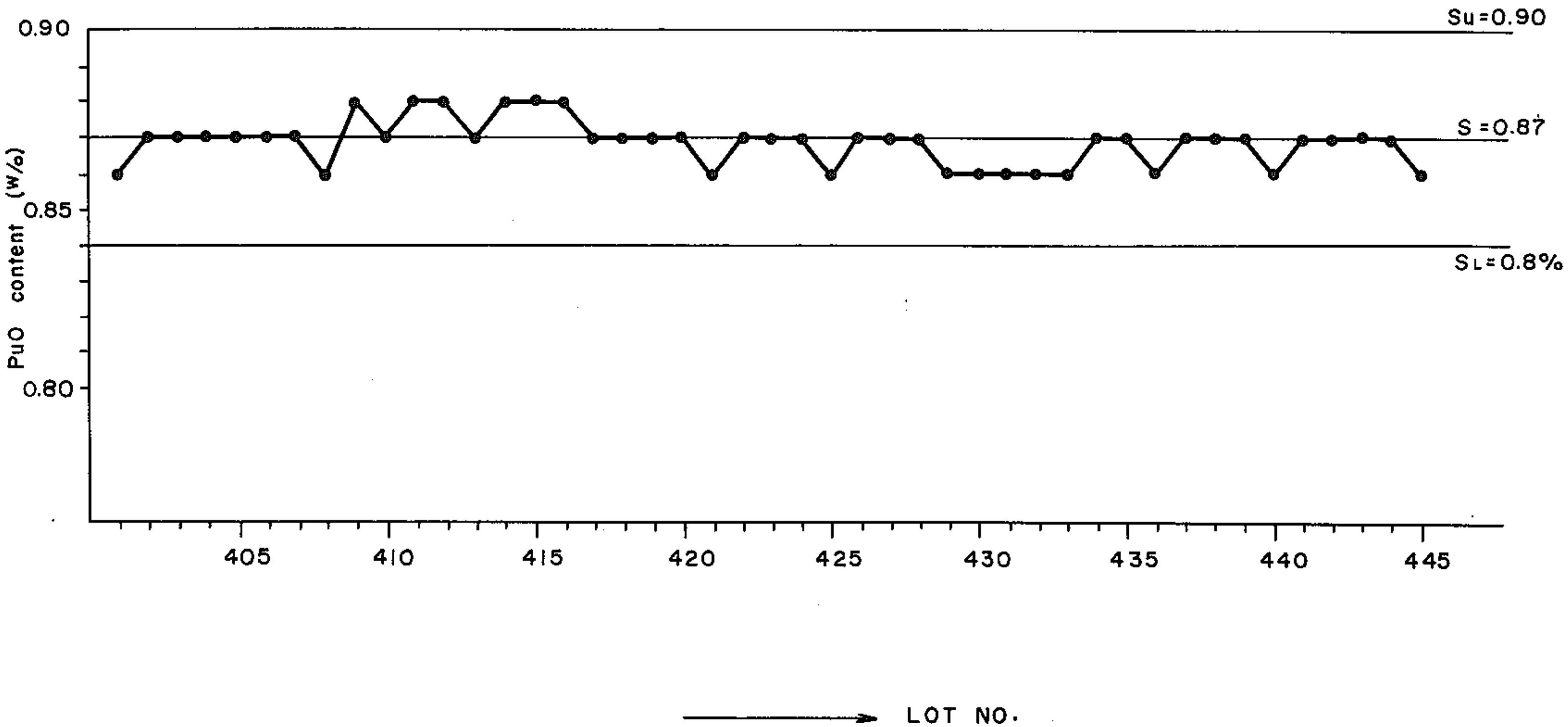


Fig. 4 PuO_2 Content to Some Production Lots in DC8R Series

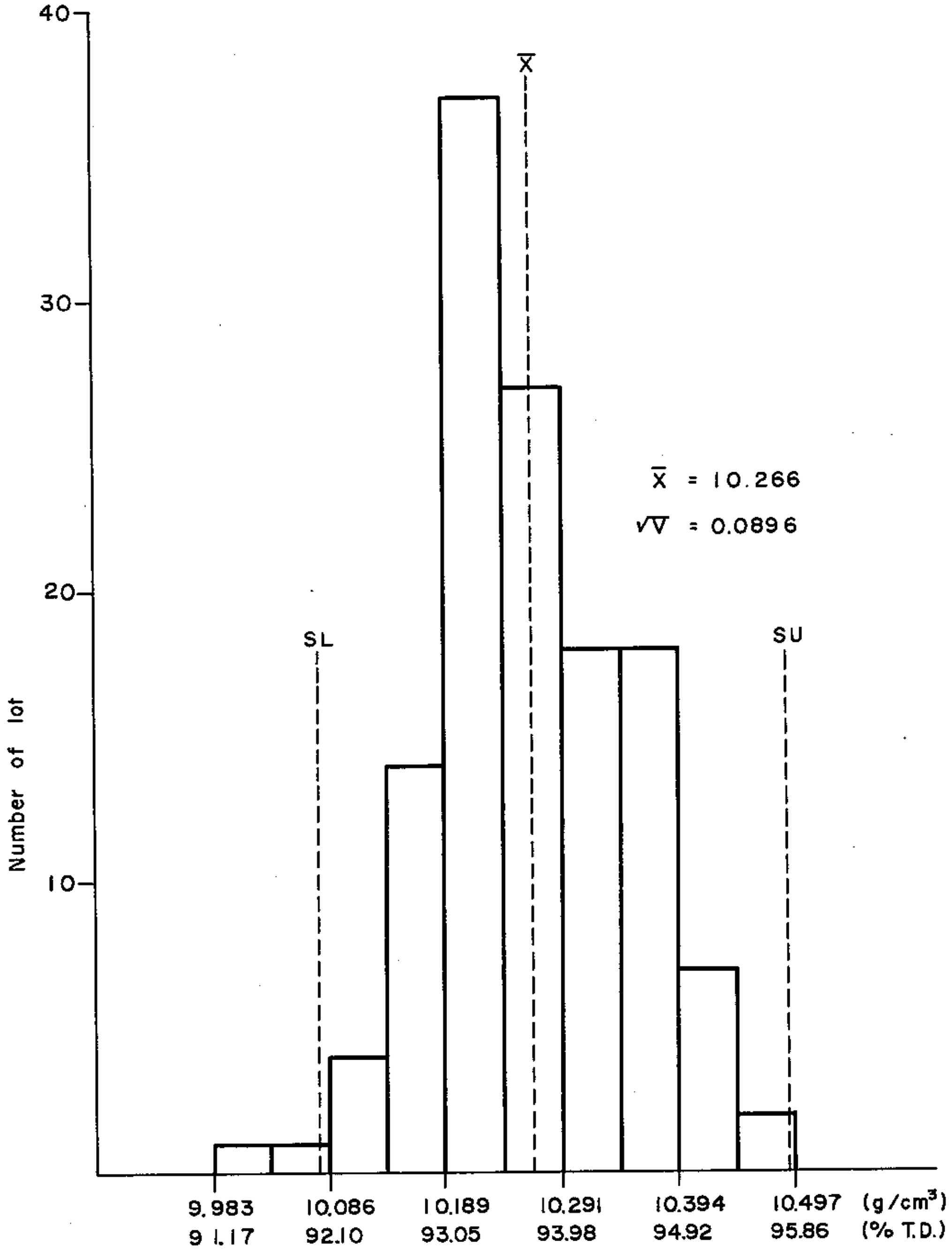


Fig. 5-1 Distribution of Densities in DC8R Series Pellets.

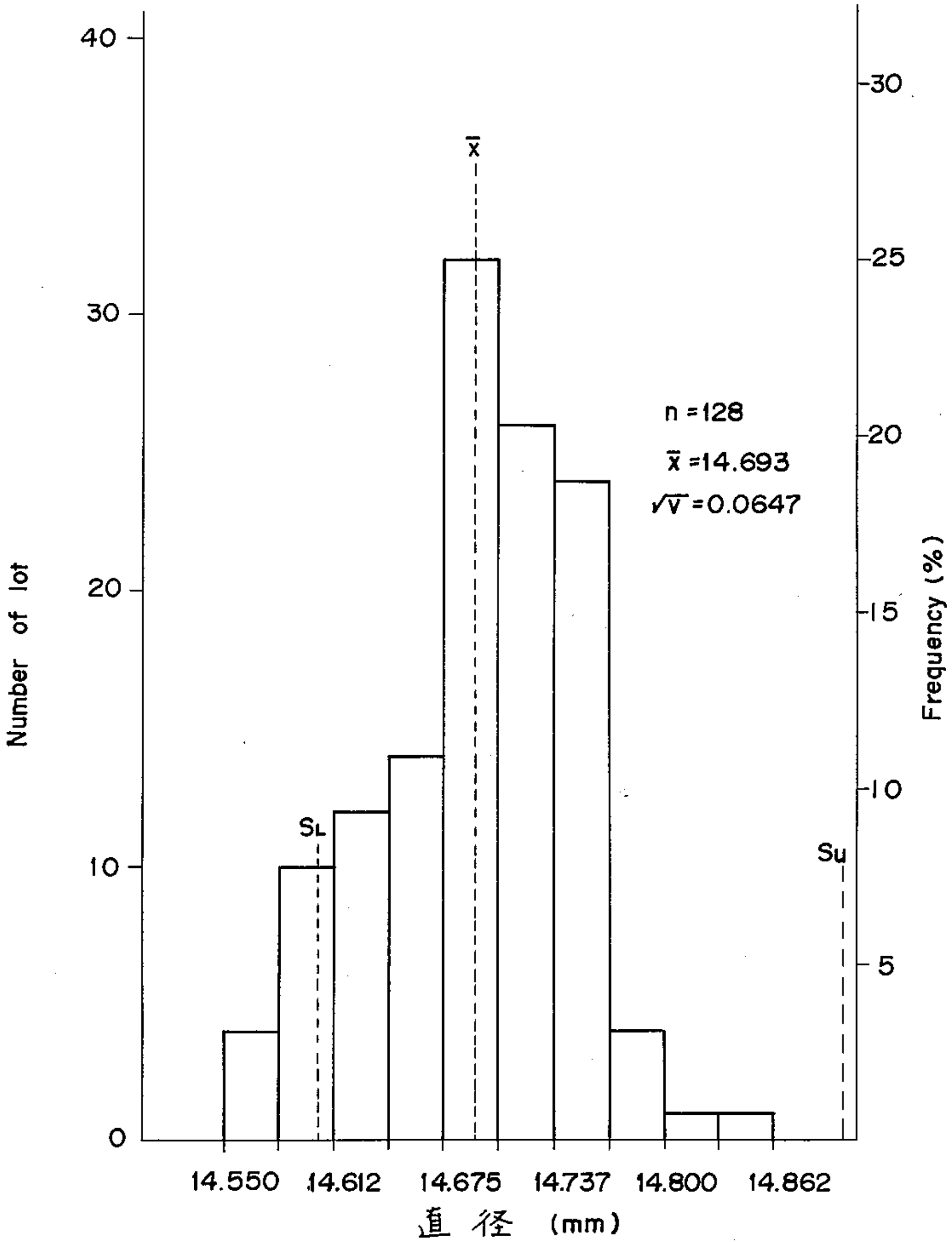


Fig. 5-2 Distribution of Diameter in DC8R Series Pellets

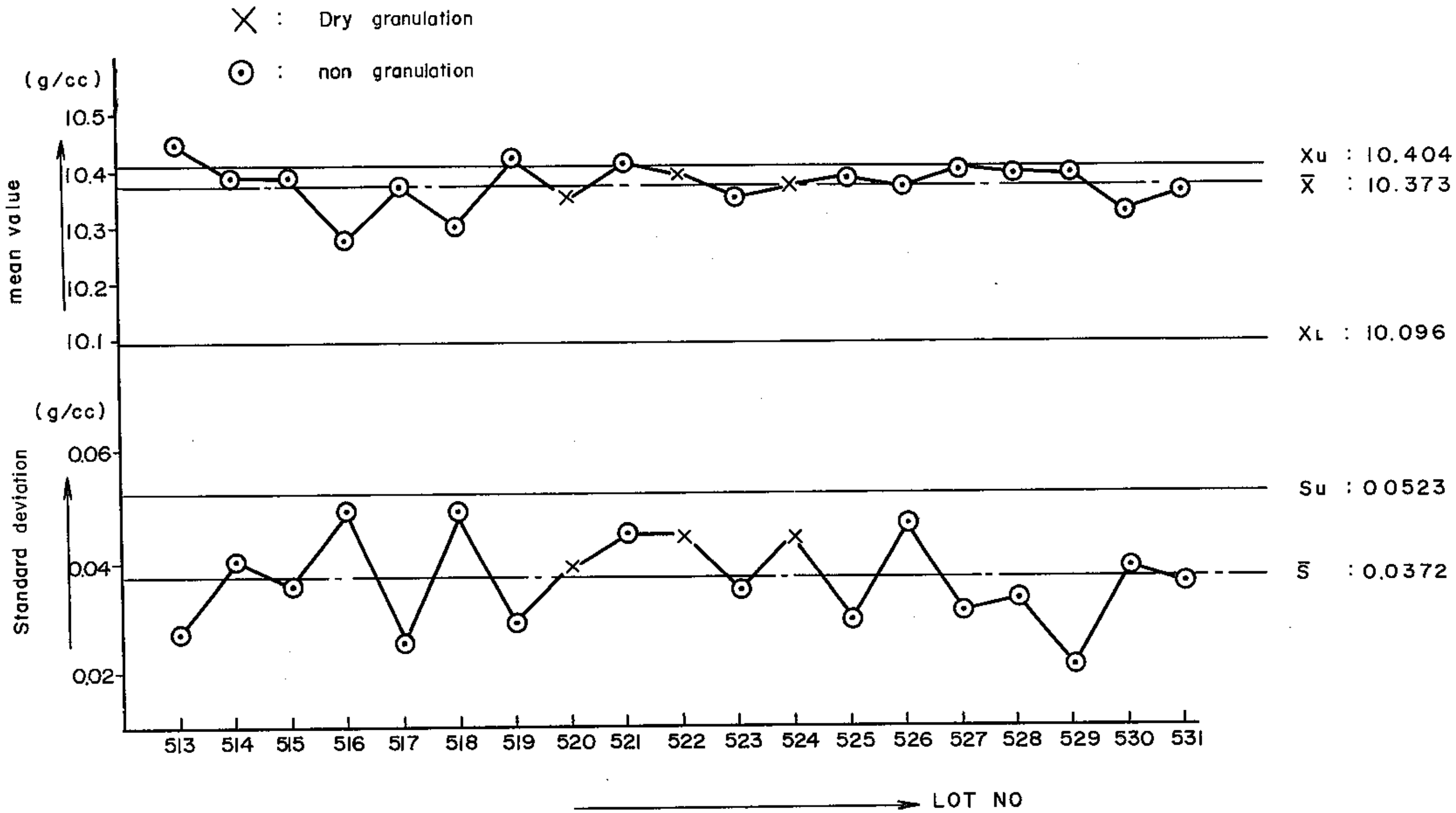


Fig. 6 Density of Sintered Pellets to Some Production Lots in DC8R Series

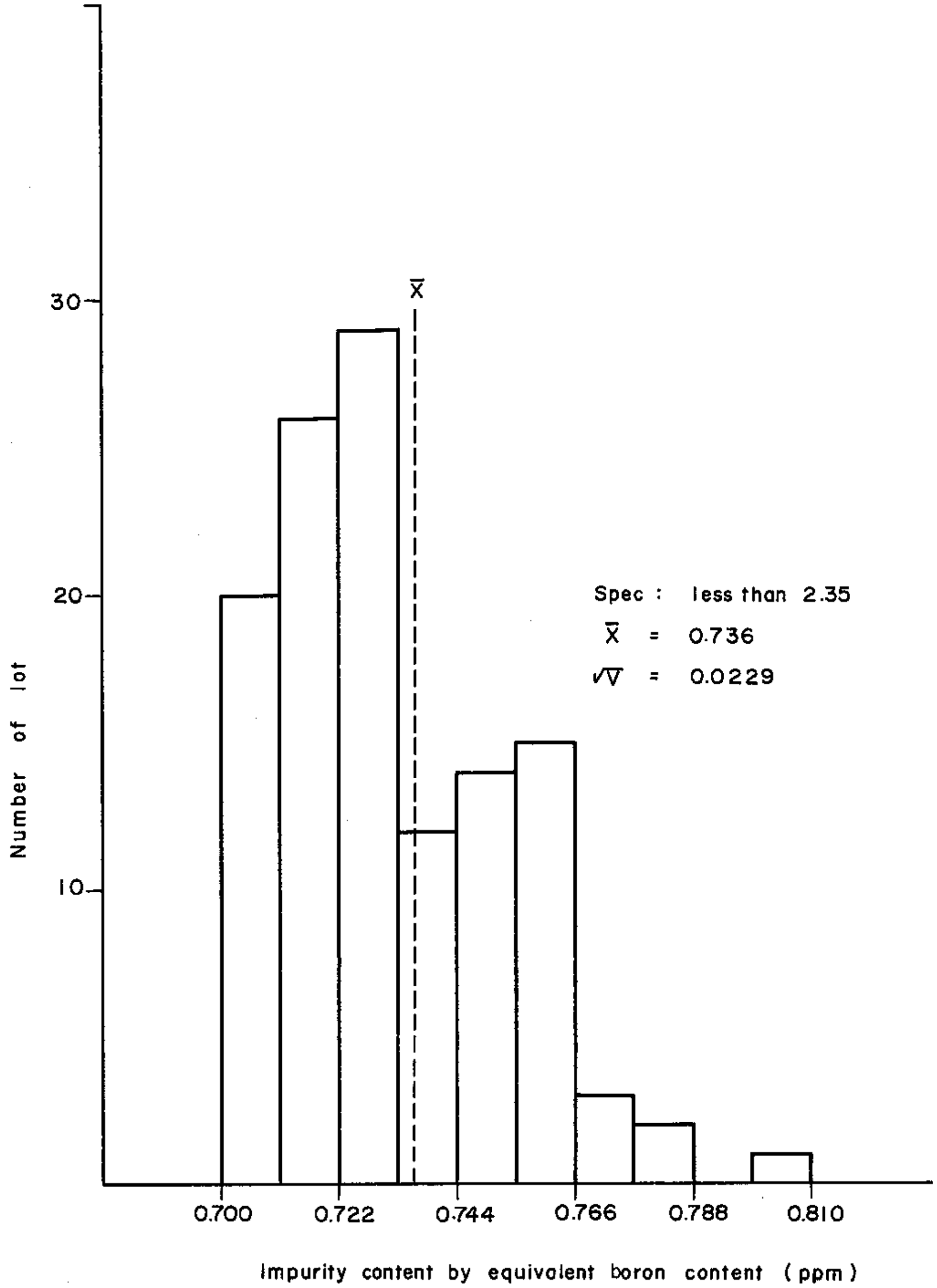


Fig.7 Impurity Content by Equivalent Boron Content of DCSR Series Pellets.

4. Reprocessing of Reject Pellets

In the course of the production, the pellets rejected due to some defects other than metallic impurity content were reprocessed through a continuous oxidation-reduction process, and the recovered powder was recycled to the pellet production process. Fig. 8 shows schematically the structure of the equipment for the process. This equipment consists of two rotary kilns for both oxidation and reduction, a pellet feeding mechanism, and a powder recycling and out-let mechanism. Rotation speed of these kilns is controllable at one to seven minutes per one rotation, which corresponds to thirty minutes to four hours holding of the powder in the kiln. The powder outlet mechanism is preventive to air-oxidation of the powder by cooling it on a water cooled screw conveyer. The material out of the oxidation kiln flows on an eight mesh screen, and unpulverized pellets of plus eight mesh are removed to recycle as feed material to the oxidation kiln. The counter currents of air and mixed gas ($5\%H_2$-$95\%N_2$) are forced through the respective kiln for oxidation and reduction. Usually, the rejected pellets were treated at 650°C for two hours for oxidation and at 850°C for two hours for reduction, and this oxidation-reduction cycle was repeated twice. The characteristics of the recovered mixed-oxide powder were as described below, and the recovered powder was blended with virgin PuO_2 and UO_2 powder upto 15w/o of whole powder in a starting mixed-oxide powder of the pellet production.

O/M ratio	:	2.10 ~ 2.05
Specific surface area (m^2/g)	:	0.7 ~ 0.9
Apparent density (g/cc)	:	1.9 ~ 2.4 (tapped) 3.2 ~ 3.7 (not tapped)

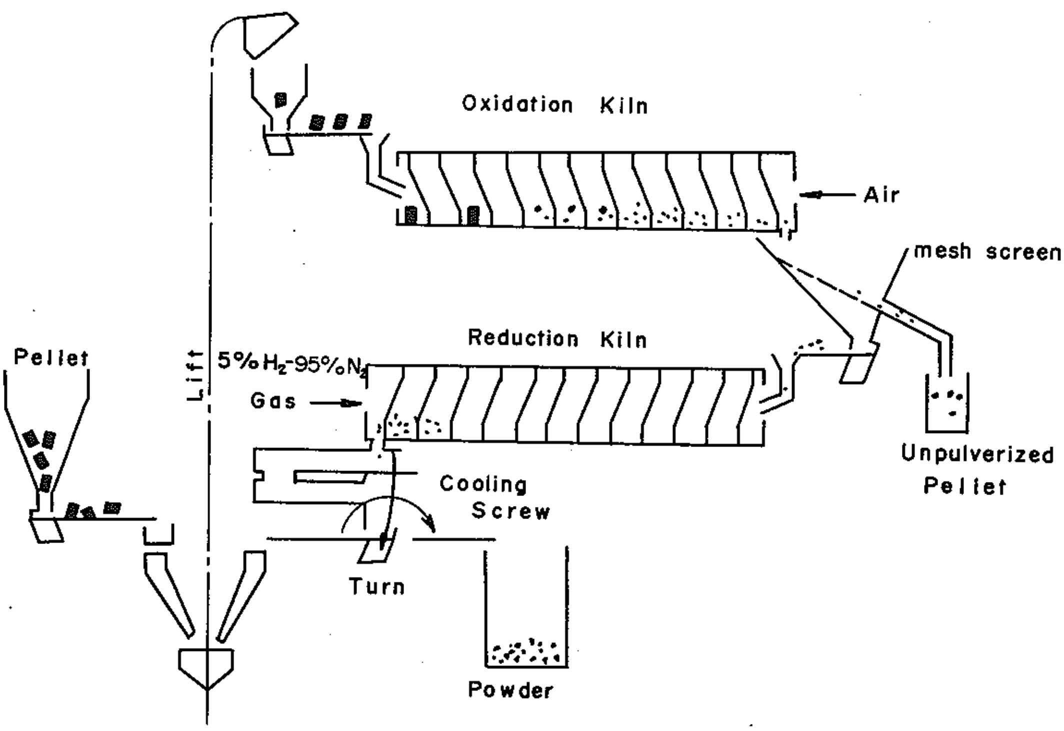


Fig. 8 Schamatic Diagram of Dry Recovery Process Equipment

5. Fabrication of Fuel Rod

The fabrication process of fuel rods is shown in Fig. 9. The processes marked with asterisk in this flow sheet were performed in the glove box. Cladding tubes with the lower endplugs welded, upper endplugs and plenum springs were supplied by a subcontractor. Tools and equipment were designed and installed in order to improve both efficiency and safety of the work, and to prevent high radiation exposure of the operator.

The equipment for pellet loading into the cladding tube consists of pellets arrangement mechanism, pellets stack measuring mechanism and pellets loading mechanism. These mechanisms are performed automatically and the operation is proceeded in sequence. Fig. 10 shows the equipment installed in the glove-box. Fig. 11 shows the end of a cladding tube attached with an "open end mask" fixed by "tubulous polyethylene film". This mask was devised to protect the end surface of the cladding tube from considerable plutonium contamination due to touching the end surface with pellets. After pellet loading, the loaded fuel tubes (fuel rods) were transported to the open hood box to do decontamination of the open tube end. In the decontamination work, the open end mask above mentioned is removed out and a substitute endplug is inserted after decontamination as shown in Fig. 12. This substitute endplug prevents the pellets and small plutonium particles from falling out of the fuel rod and from re-contaminating the open tube end. Thus, by using both the open end mask and the substitute endplugs, fixed contamination at the welded part of the fuel rods was controlled at less than 30 dpm. For the welding of the upper end plug, fifteen

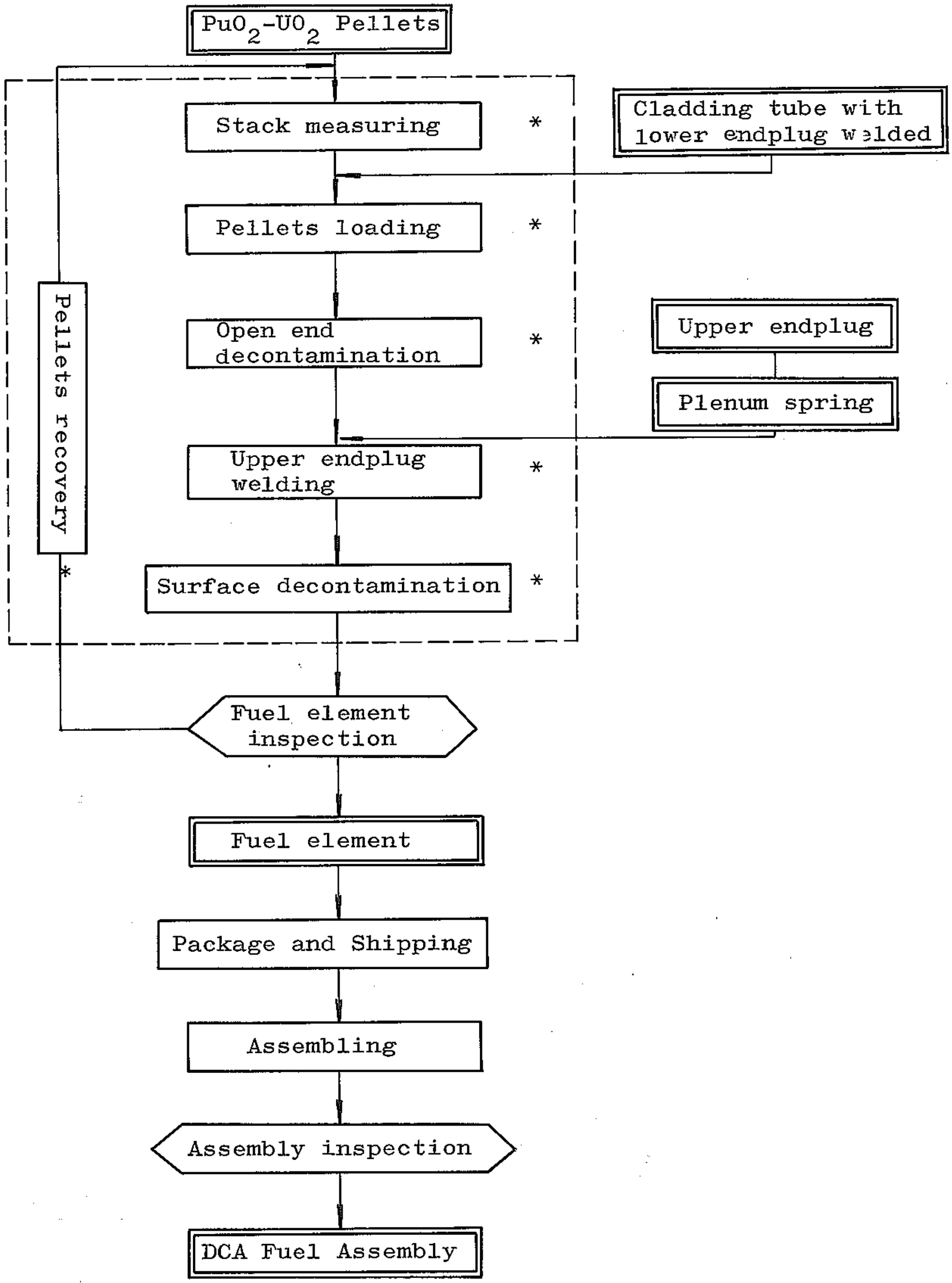


Fig. 9 The Flow Sheet of the Fabrication of DCA Fuel Assembly.

fuel rods attached with substitute end plugs were loaded into the welding chamber at the same time as a batch operation, and the chamber atmosphere was refilled by helium gas after vacuum degassing. The endplug was welded one by one to fuel rod by TIG method after removal of the sabstitute endplug, insertion of a plenum spring and a upper end plug. Welding conditions such as rotation speed of rod, welding time and electric current were controlled by the mechanism of the welder. The typical welding conditions applied are as follows.

Rotation speed : 6 r.p.m.,

Electric current : 45 ampere (15 sec.) - 70 ampere (20 sec.)

Concerning the defects occurred in the weld part such as porosity and cracks, following informations were obtained.

(1) Occurrence of defects were affected by the grade of contact between endplug and tube end, heat generation rate, and setting of chill block.

(2) Purity of helium atmosphere* is of little significance as a cause of defects. (*alittle contamination by oxygen)

After welding of upper endplug, the fuel rods were inspected for contamination, helium leakage rate, defects in welded part by X-ray radiograph, weight, length, straightness and surface defects of cladding by vision.

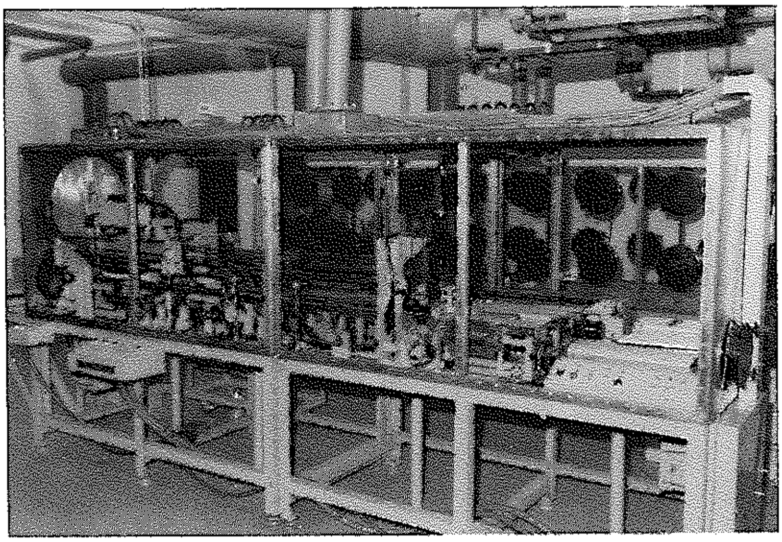


Fig.10 The Equipment and Glove-box for Loading Pellets into Cladding Tube.

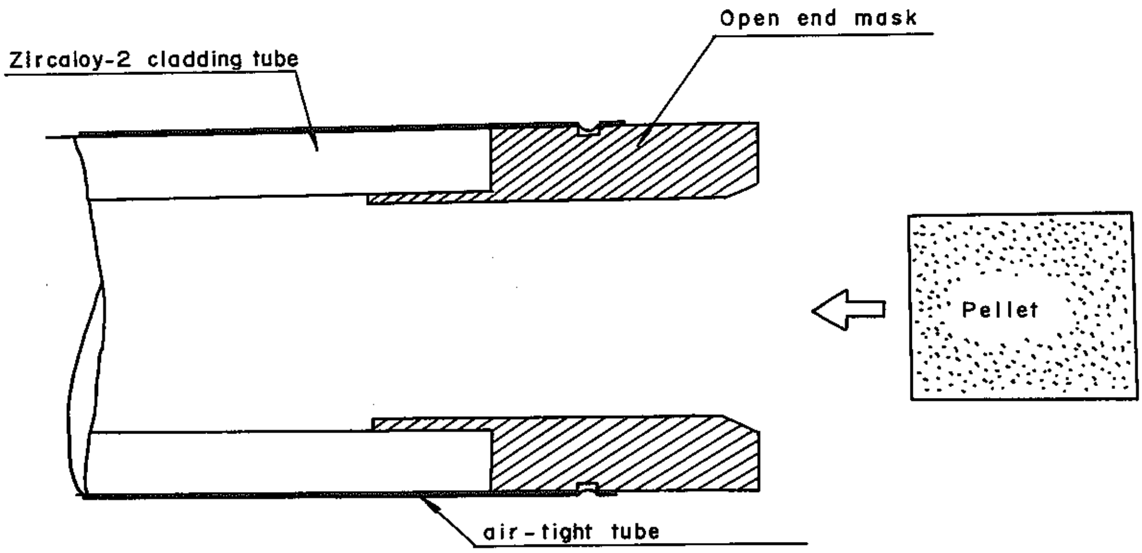
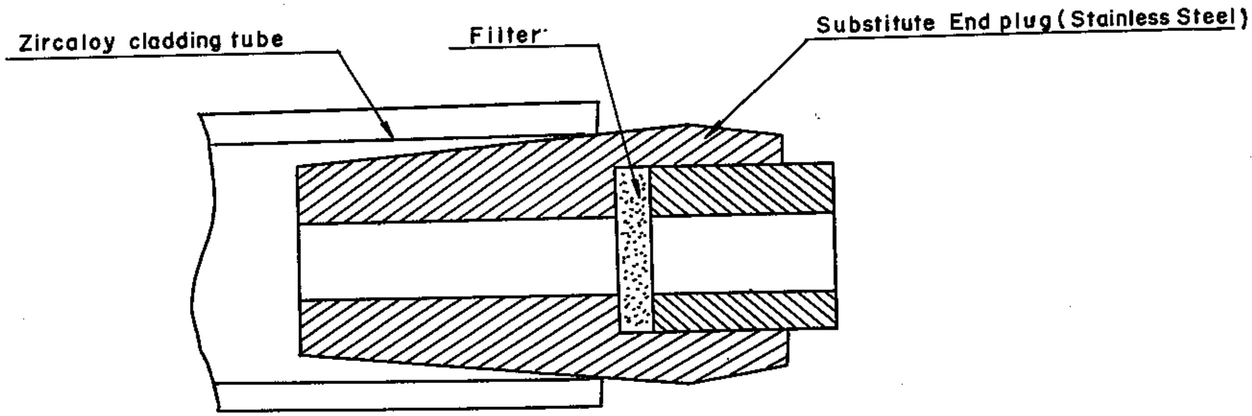


Fig. 11 Open End Mask Fixing



A Section

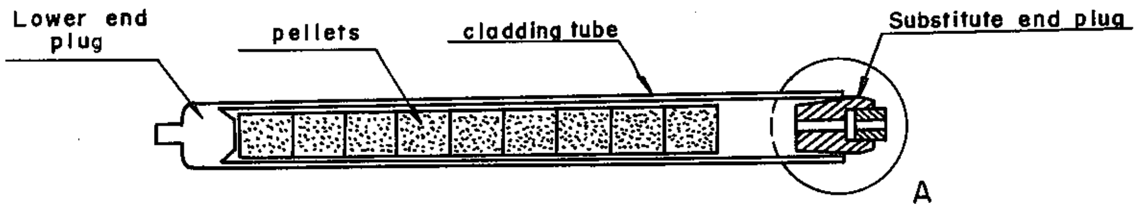
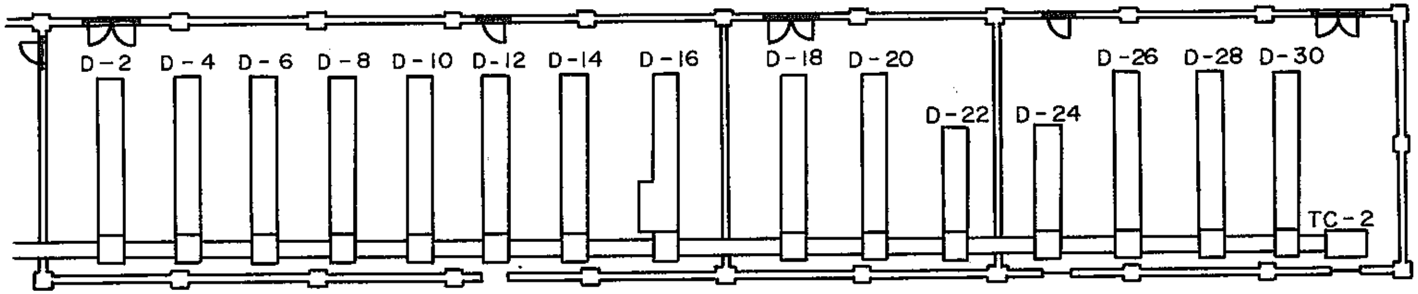


Fig. 12 Substitute End Plug

6. Production Facility

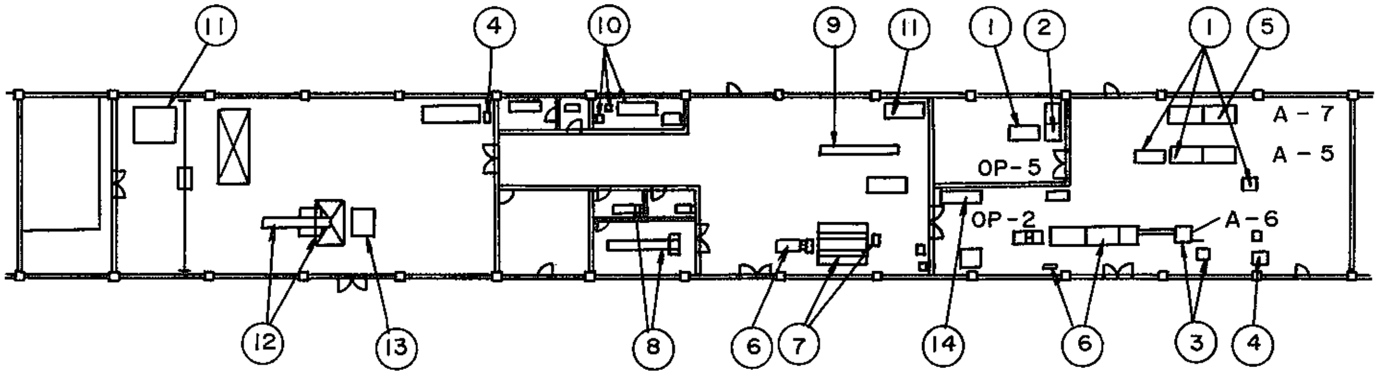
Array of the glove boxes for pellet preparation is shown in Fig. 13. Each box houses the equipment for each production step such as powder blending and sintering. Therefore, the production is conducted in batch wise, and the product material at each production step is transfered from a glove box (step) to another glove box (step) by an electric mini-cart through a tunnel which is attached to the array of glove boxes. Production capacity of the facility is about 5 tons of mixed-oxide per year, which is mainly determined by the capacity of sintering furnace, and batch size for each production step is 27 kg mixed-oxide. Arrangement of the glove boxes and other facilities for fabrication of fuel rod and assembly is shown in Fig. 14. Capacity of fuel rod fabrication is fifteen rods per day for 2 m long DCA rod, which was mainly determined by the capacity of pellet loading process. The fuel rods is carried by specially designed cart from a glove box (step) to another glove box (step).



Box No.	Objective
D-2	Power storage
D-4	Pellet crushing for reprocessing
D-6	Drying and sieving of powder
D-8	Weighing of powder
D-10	Powder blending
D-12	Wet granulation
D-14	Dry granulation
D-16	Pellitizing

Box No.	objective
D-18	Dewaxing & sintering
D-20	" "
D-22	Pellet drying
D-24	Pellet size selection
D-26	Centerless grinding
D-28	Pellet inspection
D-30	Temporary pellet storage

Fig. 13 Glove Boxes for Pellet Production



①	Pellet loading to cladding tube
②	Decontamination of tube end
③	Upper endplug welding
④	Purifier of He gas
⑤	Dismantle of rejected rod
⑥	Decontamination of rod surface
⑦	He-leak detector
⑧	X-ray radiography
⑨	Measurement of rod dimension
⑩	Metallography
⑪	Storage of fuel rod
⑫	Assembling of rods
⑬	Profirometer

Fig.14 Glove Boxes, and Other Facilities for Fuel Rod Fabrication and Fuel Assembly.

7. Safety

Nuclear critical safety is secured by designating "box fissile limit" for each glove box or fabrication step and by the operation of "carts" for fuel material transportation by people of material control group which is independent from production group.

Personal radiation exposure was limited to less than 300 mrem per three menths in this project by the application of precautionary measure such as attachment of a lead plate and a lead glass plate on the glove box pannel of high radiation activity.

8. Future Work - "Fabrication of FUGEN Fuel"

With accomplishment of DCA fuel program, Tokai-works PNC is going to undertake the fabrication of FUGEN fuel as a next task. "FUGEN" is a prototype heavy water reactor of 165 MWe electric power generation as mentioned before. A brief description of the FUGEN fuel is given here for the information. Table 4 indicates the main specifications of the fuel. Fig. 15 shows the composition of fuel rods in the fuel assembly. The shape of the pellet is shown in Fig. 16. Fig. 17 gives the structure of the fuel assembly. It is very unique comparing with PWR fuel or BWR fuel that the fuel assembly is the cylindrical cluster of fuel rods, which is composed of 16 high plutonium-enriched fuel rods and 22 low plutonium-enriched fuel rods. Shape of the pellet and length of the fuel rods are very different from those of DCA fuel. Especially that the length of fuel rods is twice that of DCA rod requires considerable modification to the fuel fabrication facility used for DCA fuel. There is a new technical problem that very narrow gap between fuel rods (about 2 mm) is to be assured in the fuel assembly. We are now conducting the pre-production test to overcome new technical problems due to stricter requirement, and to establish the technology. In total, 100 fuel assemblies for initial core loading are to be fabricated until the fall of 1977, which is equivalent to the production of about 17.8 tons of mixed-oxide pellets.

Table 4 Specifications of FUGEN Fuel Assembly

(1) Mixed-oxide pellets

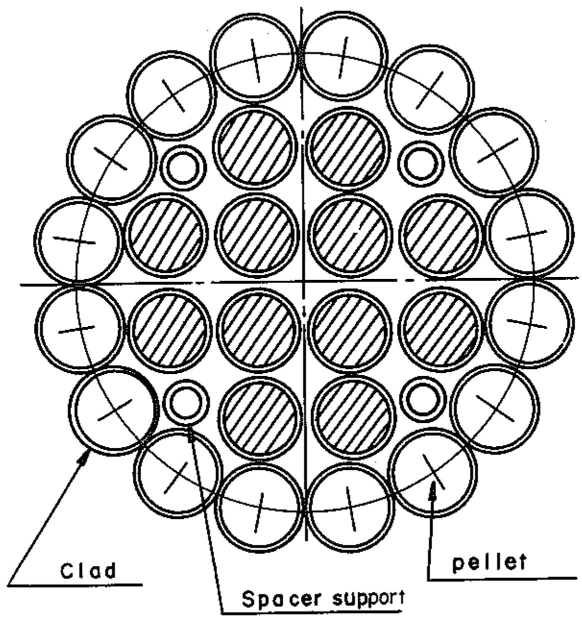
Material	:	PuO_2-UO_2 (Natural)
Pu content	:	(i) 0.55 w/o Pu fissile for the outer layer fuel rods
		(ii) 0.80 w/o Pu fissile for the inner layer fuel rods
Density	:	95.0 % T.D.
Diameter	:	14.40 mm
Hight	:	18 mm
Shape	:	As shown in Fig. 16.

(2) Cladding tube

Material	:	Zircalloy-2
Inner diameter	:	14.70 mm
Outer diameter	:	16.46 mm
Thickness	:	0.80 mm

(3) Fuel rod

Outer diameter	:	16.46 mm
Active length	:	3700 mm
Plenum length	:	275 mm
Plenum gas	:	He 1 atm
Total length	:	4125 mm & 4151 mm



Pellet	Region	Pu-fis. O_2 content	PuO_2 content
(hatched)	High enriched	0.80 %	~ 0.995 %
(open)	Low enriched	0.55 %	~ 0.664%

Fig. 15 Composition of Fuel Rods

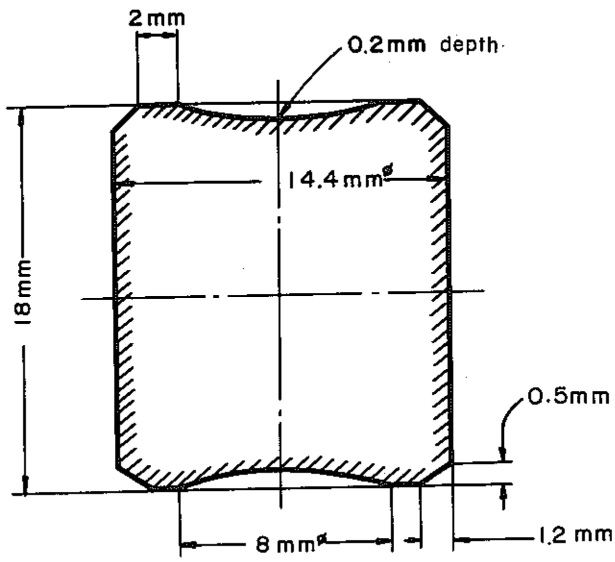


Fig. 16 Shape of Pellet

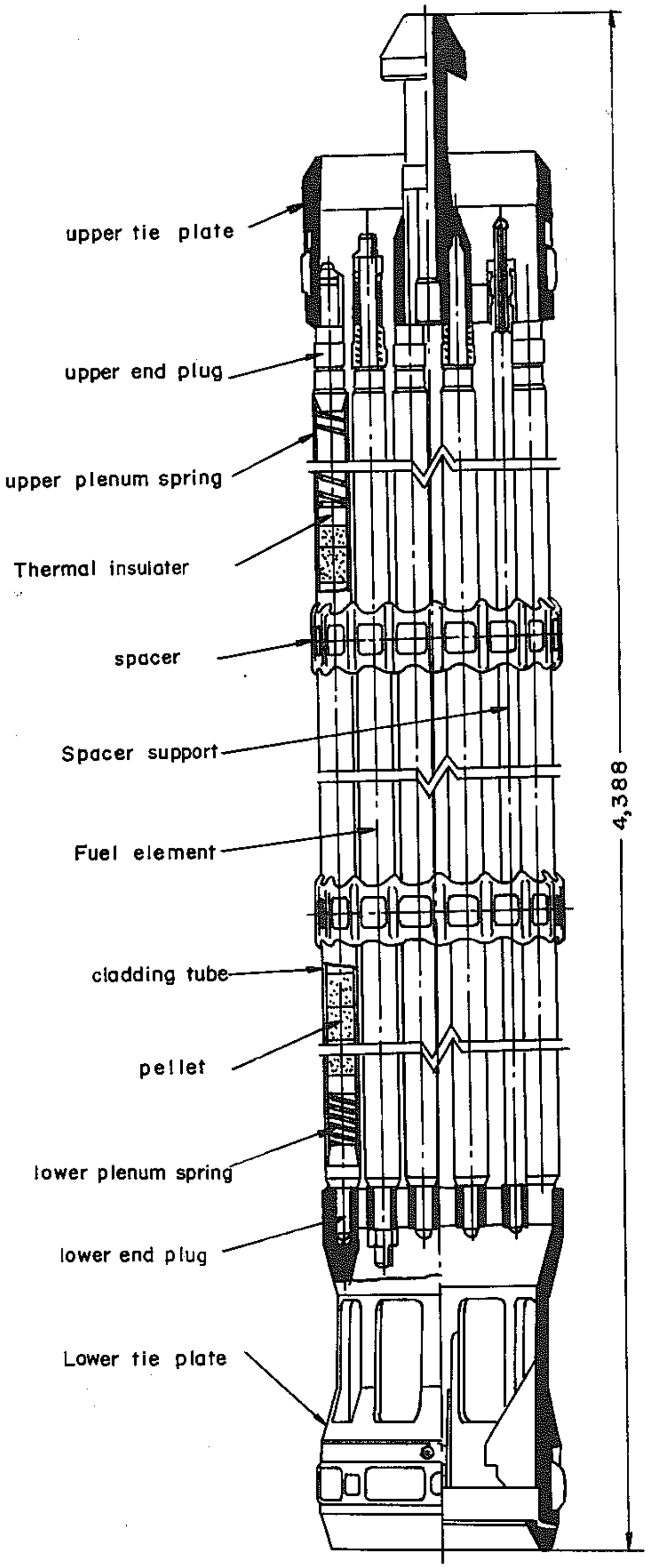


Fig. 17 Structure of The Assembly

CONCLUSION

Nearly 10 tons of mixed-oxide fuel in 92 fuel assemblies for the Deuterium Critical Assembly (DCA) were successfully produced from April 1972 through March 1974. This is the first mass-production of mixed-oxide in Japan. The newly-built Plutonium Fabrication Facility of Tokai-Works PNC demonstrated its capability.

Being based on the experience and informations obtained during DCA fuel production project, the new project for the production of FUGEN (a prototype heavy water reactor of 165 MWe) is designed, and the preproduction test is being conducted after modification of facilities.