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THE MANUFACTURE OF FUEL ELEMENTS OF THE ARGONAUT TYPE

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1. This paper reviews the work performed in order to produce fuel elements of the Argonaut type by the extrusion of stock containing a mixture of powdered metals.

2. The method followed for the preparation of the powdered metals and their handling in charging the stock to be extruded is described.

3. Some conclusions are given as regards the significance of the geometry of the stock as a dominant factor in obtaining a lining of acceptable quality and of a thickness which can be varied at will. The extrusion conditions are determined, with reference to the temperature limits, maximum uranium oxide concentration, etc.

4. The method used to eliminate the uranium from the cut ends is described, and an analysis is presented of the process utilized for the determination of the amount of uranium in each plate.

5. Other possibilities are suggested, to be tried out in the future, in the light of these findings.

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MANUFACTURING OF FUEL ELEMENTS OF THE ARGONAUT TYPE

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THE MANUFACTURE OF FUEL ELEMENTS OF THE ARGONAUT TYPE

by

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This work investigates the conditions required to manufacture fuel elements of the RA .1 Argonaut type.

The fuel elements to be used are in the form of a plate (figure 4) 2.75 mm in thickness by 7.3 mm wide, and 61 cm long, which is manufactured by the extrusion of a presintered mass of U_3O_8 (enriched to 20%) in an aluminum matrix.

The investigation consists of the following steps:

1. Obtention and specification of U_3O_8 and aluminum in a powder for the tests.
2. Filling with slugs for extrusion tests.
3. Extrusion tests.
4. Finishing of fuel elements.
5. Computation of the U_3O_8 content.

Observations are added based on the examination of some specimens.

1. Obtention and specification of U_3O_8 and Al for the Tests:

For the tests, use was made of U_3O_8, obtained by calcination during two hours at 900°C of $UO_3 \times H_2O$ supplied by the Ezeiza plant of the C.N.E.A. Calcination was carried out in porcelain containers and there was no granulometric variation observed during the process (figure 2). The final tests were carried out with Mallinckrodt U_3O_8 finer than the 200 mesh.

Aluminum powder obtained by grinding was used in the preliminary tests. This had the disadvantage of including the organic matter used for the covering. The aluminum powder was obtained by grinding under the size 70 mesh supplied by the Purimex firm. The powder responds to the characteristics of 2 S aluminum.

2. Filling of the Cartridges for the Extrusion Tests:

The cartridges for extrusion were filled with a mixture of U_3O_8 and Al. The mixing was effected by two different methods:

a) Ball-type mill
b) Double-cone mixer.

In both methods it was observed that a mechanical agglomerate of the U_3O_8 was produced. This was ascribed to the U_3O_8 grains which had sharp edges, in contrast with the round shape of the aluminum grains. No advantage

in the use of the ball-type mill to eliminate this effect was observed. The double-cone mixer was used for all subsequent tests with allowances made for the advantages of cleaning and the facility in loading and unloading. Once the mixture was made, it was sifted on the 70 mesh and broken with a rubber spatula. The remnants of U_3O_8 agglomerated during this mixture.

Additional precautions were taken while working with enriched U_3O_8 in order to avoid losses or contaminations. The operations were carried out in a dry box. The work schematic is shown in figure 1.

The filling tests were carried out in a press. Pressing was carried out in stages with a maximum pressure of 0.3 to 0.4 ton per cm^2 to try to obtain even pressing. This gave values of ρ=2.1 Gm/cm^3 for 40% U_3O_8 in the mixture. The method was difficult, requiring complicated handling with the attendant risk of losses and contamination.

Filling tests were carried out on the cartridges by a vibration. A device built on Piazza's specifications (1) was used, which gave good results. Values of ρ = 1.91 Gm/cc for a U_3O_8 content of 45 % in the mixture were obtained.

Closing of the elements was accomplished in the filling dry box, so they were ready for subsequent handling without risk of losing any powder.

3. Extrusion Tests - Several tests were carried out, allowing for the following variables:

1) Geometery of the container cylinders.
2) Material in the cylinders
3) Shape of the matrix.
4) Lubrication.
5) Extrusion temperature and rate
6) Content of uranium oxide.

The press used was a Schloemann 750-ton machine with a bushing diameter of 110 mm and electrical heating.

1) Geometery of the container cylinders: The first design was a cylinder having 5-mm thick parallel sides. This cylinder (figures 3 and 5) has a plug of the same material in its front part, 60 mm in height, which, in this case, provides the material needed for covering. At the end, it carries a drag disc made of brass 30 mm thick, which was used to avoid the formation of an extrusion defect by inversion of the flux in the final stage of pressing. The manner in which this disc is adjusted is important to the process and is discussed below. After closing the element another disc of the same material as the cylinder was put in place. (The dimensions are of no consequence but, in this work a height of 20 mm was used) under pressure, thus, having the unit ready for the process without any risk of a loss of material, which can be manipulated without any special requirements regarding its toxicity.

The use of the disc is necessary to eliminate the defect in extrusion noted in figure 7 and 8, which makes for defects in the extruded plates shown in figure 9. In addition, its function is to propel the powder load, sweeping the wall laterally. However, if this is appreciably reduced in order to improve the effect, fine-load streams are dragged out in the rear part, so that material is lost which is afterwards, difficult to recover chemically. If the seating is very large, the effect of entrainment is reduced, and when the thickness of section x (figure 5) is decreased under a certain limit, approximately s mm, an extrusion defect ahead of the dragging disc is found.

In the case of the straight-wall tubes, it was observed, after giving a maximum offset of 1 mm to the dragging disc, that the major part of the front plug is extruded in the form of fuel aluminum until a suitable dead angle is formed. When this happens an oxide mix covered with aluminum begins to be extruded. However, in this situation, the amount of aluminum is not enough to cover pans more than 3 to 4 meters long, with yields expressed in U_3O_8, oscillating between 30 and 50% and never exceeding 60%. On figure 10 a partially extruded cylinder can be noted in which there is no additional material left. Unable to use a conical entrance to the matrix, it was thought convenient to vary the geometry of the containers.

This means, in order to increase the yields by changing the geometry, there are two possibilities: to vary the front plug in order to avoid a progressive variation of the oxide in the extruded plate, thus using between 1.5 and 2 fuel elements of 60 cm which otherwise would have had to be rejected due to having too little outside; or the second solution, which consists of counting with more added material for covering, in which it is necessary to creep toward the walls, which practically does not occur in the straight-wall pipes, since the material in the walls passes to the rear part of the dragging disc.

To this end cylinders were machined of variable wall thickness in a continuous fashion giving an adequate value to tan α (figure 5), as to count with a component normal to the matrix for the expulsion of covering materials. It should be pointed out that the cross section of the cylinders is symmetrical to the bushing and not with respect to the matrix, as would be desirable in order to simplify machining.

The following values of tan α were experimented with: 0.125, 0.312, 0.375.

Figure 17 shows the influence of tan α on the oxide content.

As we noted that in the front part of the plate, where the covering supply is carried out essentially with material of the front plug, the influence of tan α is practically zero, but starting from approximately 3 meters it begins to bring material from the walls which, in this fashion, maintains a constant dead angle until practically the end of the extrusion (figure 10, 11, and 12).

For small values of tan α, there is a decrease in the oxide content in an intermediate zone. This effect, which is also a function of the oxide concentration, as will be observed further ahead, is accounted for the fact that when α is lodged for a given length of cartridge and oxide concentration, the decrease in the dead angle begins at three-quarters of the length. This phenomena has not been systematically investigated, so it is not possible to present tables for the extreme values.

In view of the symmetry of the cartridge, as the tangent increases, the amount of material not active in the margins of the plate also increases.

In the following table is presented data of tan α covering thickness and width of the inactive material on the edges for positions of 4 meters in the length of the plate.

Thickness of the Margin	Covering Thickness	Tangent α :
3 mm	0.1-0.2 mm	0.125
5 mm	0.2-0.3 mm	0.312
7 mm	0.4-0.5 mm	0.375

For the case of tan α 0.312, yields were obtained in some specific cases of more than 90%. This value of tan α has been set because it give good overlap without markedly increasing the aluminum content. The variation in the covering along the extruded plate can be seen in Figure 13.

The other solution, which has been proposed to increase the covering, is to avoid a continuous variation of the oxide in the first 2 meters. Use was made of the front plugs having a parabolic section to compensate for the greater velocity of the central flux until the time of extrusion when the dead angle is formed for aluminum and the covering falls within the normal values. Tests were carried out with symmetrical plugs with respect to the bushing with some positive results, but there is the danger of a flux of streams which contain uranium oxide between the covering material and the parabolic plug. Therefore this particular method was abandoned without any final conclusion.

2) Cylinder materials: To improve the patch of cross section and resistance to corrosion, tests were carried out with aluminum cylinders of 99.8% purity, but adhesion of the aluminum in the face of the matrix and in the friction planes was damaging the thin cover. This disadvantage could not be resolved by polishing the matrix or by lubrication on the basis of the emulsified graphite so that 61 S alloy was used which, with a slight loss of the capture cross section, gives very good results. Its corrosion resistance is known.

3) Shape of matrix: A straight-edged matrix was used and following a series of tests from a friction plane of 3 mm height. The final design was reached with a height for the friction planes of 0.7 mm, with a progressive decrease to the edges, from 7.5 mm from those until it was zero at the extremities. The discharge is 4 to 6º and the step 0.4 mm. This design was used for over 40 extruded cartridges without any drawback. The surface of the friction planes is polished with No. 2 alumina and colloidal graphite lubrication (figure 6) was used. In figure 14 we shall note the defects caused by the unduly large friction plane.

4) Lubrication: The tests carried out with lubricants were not conclusive, and it was dangerous to use them in the equipment available because of the possibility of the formation of double layers.

5) Extrusion temperature and extrusion rates: The cartridges are preheated for 4 hours at extrusion temperature, after which they are extruded.

For the alloy used (61 S), a temperature was found at which the quality of the covering was optimum, and this gave a utilization range from 360-420°C, with a bushing temperature of about 370°C. A lower temperature value makes the pressure (750 tons) inadequate, and higher temperatures cause sticking of the fine covering of the matrix.

We experimented with temperatures up to 550°C, with a bushing temperature up to 420°C, and observed a defect due to temperatures around 500°C (figure 16). The extrusion rates were low, some 1 to 2 meters per minute. At higher speeds it is easy to scratch the cover.

It was observed that the covering thickness, as expected, is a function of the temperature difference between the bushing and the cartridge. As the cartridge temperature rises, the covering thickness decreases due to greater central flux.

For values greater than 100°C, the quality of the covering layer is poor.

We observed in figure 18, a whole series of cartridges with constant alpha tangents, a bushing temperature of 340°C, giving, along the plate, a variation of density as a function of the temperature difference between the cartridge and the bushing.

The density variation is about 0.1 Gm/cc for the central area, the density difference in the first section is insignificant. The dispersion in the final section is caused by slight length differences in cartridges of the order of the centimeter.

6) Uranium oxide contents: We made a study of 40%, 45% and 48% contents. It was observed that for high contents, 48%, the covering must break, even with high tan α values.

Where tan α is constant (figure 19) it was observed that as the U_3O_8 content increases, the effect caused by a decrease in the U_3O_8 content in an intermediate zone goes down, as a consequence of an increase in the covering thickness. This effect is attributed to the fact that as the U_3O_8 content increases, there is a greater addition of material to the walls, which begin to creep from the first stage of extrusion (figure 11). It would seem that this effect is negligible for a 45% content and practically zero for 48%. This does not apply to a 40% content that may cause rejection of fuel elements in a 400 cm area, due to low content.

The elements that contain 48% uranium, on the other hand, are only slightly ductile, since subsequent forming, straightening out, for instance, is somewhat risky. Accordingly, the last elements in the series were made with a charge of 45% uranium oxide.

4. Finishing of fuel elements. - The plates obtained from extrusion are then cut and straightened out in agreement with the design sought by means of a special machine, first indicating the line of cut and allowing for the lowest possible activity level in connection with the percentage of dioxide used in earlier tests.

Each section of the cut plate is so marked that it is possible to determine its position in the extruded plate and to determine from which slug it comes.

To avoid contamination of the reactor's cooling water, we must eliminate any uranium on the surface. This can be done by either painting with the right paint or by chemically eliminating the exposed uranium.

For this project, both methods were used. In good quality fuel elements, each was washed with concentrated nitric acid, which eliminated the superficial uranium and small defects from the ends. The amount of uranium that passes to solution is about 0.007 Gm expressed in U for each plate. Thereafter, we made two washes with water.

To avoid a substantial dissolution of the uranium and penetration of acid into possible cracks for elements with large defects, capable of being used in a reactor, aluminum paint was used as a covering. They are currently in operation.

Metallographic observations: In figure 16, we see a uniform distribution of the U_3O_8 particles in the Al matrix, without any appreciable agglomeration of the U_3O_8 particles.

The extrusion conditions are adequate for good sintering of powdered aluminum and U_3O_8.

A correct adhesion between the covering and the base material was observed.

5. Computation of U_3O_8 content: We determined the U_3O_8 content by measuring the density of each plate, finding an empirical correlation between the total density and the U_3O_8 content in small samples, 1 cm long, taken to the width of the extruded plate.

Starting from the theoretical formula $\frac{1}{d} = \sum \frac{xi}{di}$

in which d is the density of the sample and xi the components with a density di.

To obtain the computation, an evaluation is made of the components of the mixture U_3O_8 of unknown density, aluminum, UO_2, and Al_2O_3, formed in the sintered product as a consequence of a possible reaction between U_3O_8 and aluminum.

In this way we obtain:

$$\frac{1}{D} = \frac{x_{U308}}{x_{U_3O_8}} + \frac{x_{UO2}}{d_{UO_2}} + \frac{x_{Al_2O_3}}{d_{Al_2O_3}} + \frac{x_{Al}}{d_{Al}}$$

It is assumed that the amount of UO_2 formed is proportional to the amount of U_3O_8 and, therefore, the quantity of Al_2O_3 is proportional to that of UO_2 and thus, to that of U_3O_8.

In other words:

$$\frac{1}{D} = \frac{x_{U_3O_8}}{d_{U_3O_8}} + \frac{a\,x_{U_3O_8}}{d_{UO_2}} + \frac{b\,x_{U_3O_8}}{d_{Al_2O_3}} + \frac{x_{Al}}{d_{Al}}$$

and: $$x_{Al} = 1 - x_{U_3O_8} - x_{UO_2} - x_{Al_2O_3}$$

Thus: $$\frac{1}{D} = x_{U_3O_8} \quad A + B$$

Values A and B are obtained experimentally by statistical analysis of one population of 34 samples.

The correlation coefficient of the samples is:

$$r = 0.91$$

and the typical error of the evaluations is

$$Se = \pm\ 0.043$$

which corresponds for a confidence limit of 95%, to a degree of correlation for a population of $P = \begin{cases} 0.82 \\ 0.95 \end{cases}$ giving $A = -4.17099$, $B = 1.5786$

There is a graphic expression of this equation in figure 21; from the value of B, we deduce the density of aluminum which is : d_{Al} 2.6495/cm^3

The apparent density of U_3O_8, considering only aluminum and U_3O_8 is

obtained from A and gives d_{U3O8} 7.20 Gm/cm^3

Yields

The U_3O_8 yields in the fuel elements, which may be used as a function of the temperature difference between the cartridge and the bushing are the following:

Temperature Difference	Yields %	Number of Cartridges Consicered
0 - 40°C	71.3	16
40- 60°C	65	5
60-130°C	61	6

The maximum yields obtained are the following:

Element No.	Yields %
63	94.62
67	95.46
68	98.02
76	92.60
50	97.89
75	94.20

SUMMARY

For loading the R.A.1 reactor, Argonaut model, we made plates of fuel elements by extruding the mixture of aluminum powder with 45% of U_3O_8 (20% enriched) from aluminum cartridges, 61S, with sloping walls of tan α 0.312 through a matrix of 0.7 mm having a step at 380°C. This is the total yield of the reactor load as a percentage of U_3O_8, taking into consideration inadequate fuel elements and rejected plates amounting to 65%.

The conclusions of this study suggest the possibility of extruding small plates of a much greater thickness to bring them to size by lamination. In this way breaks of the aluminum cover by loss of adhesion since are avoided, since it is known that the extruded plates can have appreciable reductions.

Our thanks go to the Argonne National Laboratory for the important data evolved during this study.

Further, the authors wish to express their thanks to "Camea House" for the equipment made available, particularly to Mr. Doler, the engineer, for his advice on the extrusion of fuel elements.

We also wish to thank the Department of Nuclear Reactors, particularly the Metallurgical Division of the Analytical Chemistry Section and the Electro-Mechanical Division, which in many ways have made this project possible.

References

1. The Argonaut Reactor - Comunicación privada del Argonne National Laboratory.

2. Elementos combustibles de tipo disperso - C.E. Weber y H.H. Hirsch. Acta de la Conferencia Internacional sobre la utilización de la energía atómica con fines pacíficos - Naciones Unidas, Ginebra 1956 V 9 P:1-217.

3. The Chemistry of Uranium - J.K. Katz y E.I. Rabinowitch - Mc Graw Hill Book Co., New York 1951.

4. J. Piazza, Revista de la F. I. Q. Santa Fé, V:XXIV, 1955 Pg. 10.

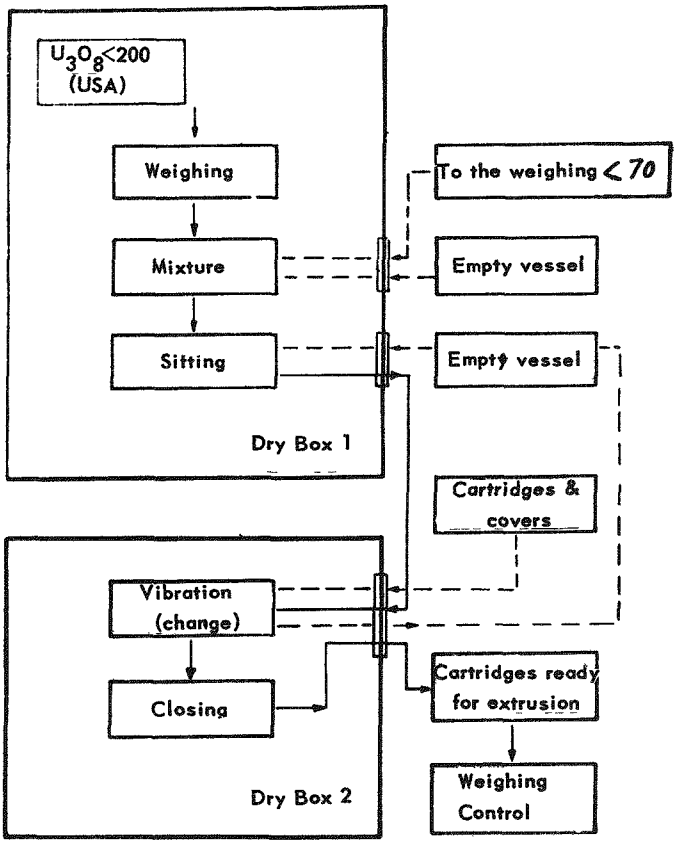

Fig. 1. - Schematic of loading process.

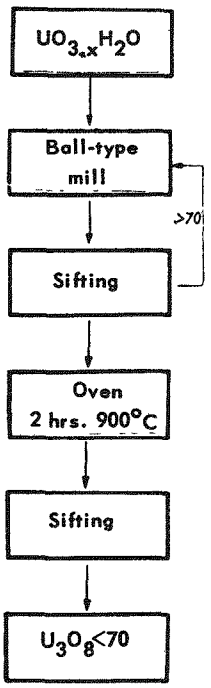

Fig.2. - Preparation of U_3O_8 for the test.

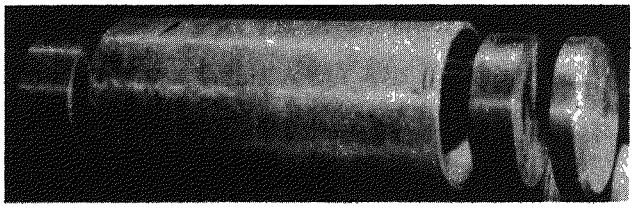

Fig. 3. - View of the extrusion cartridge

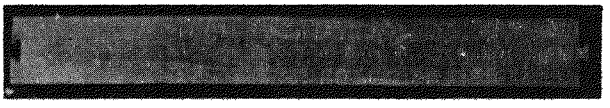

Fig. 4. - Finished fuel elements (M: x 0.15).

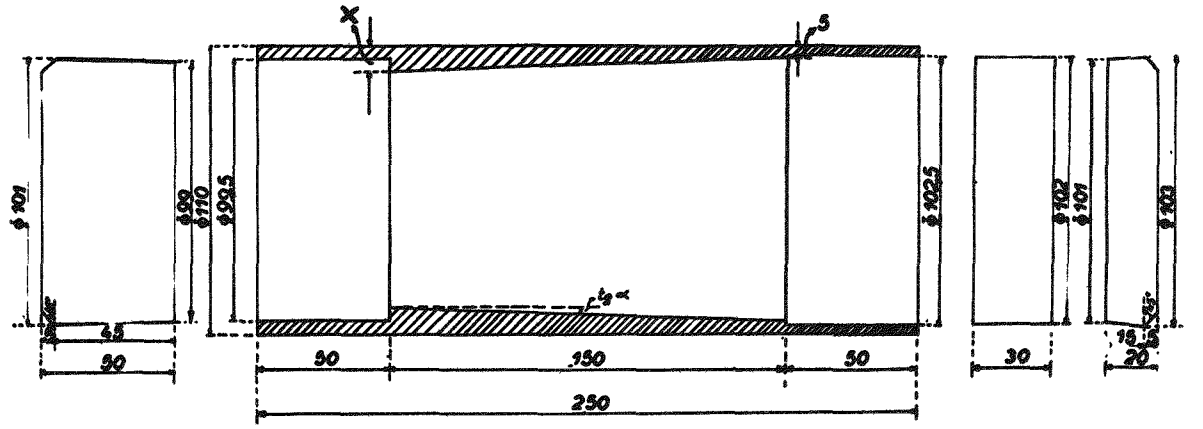

Fig. 5. - Longitudinal cross section of extrusion cartridge (dimensions in mm).

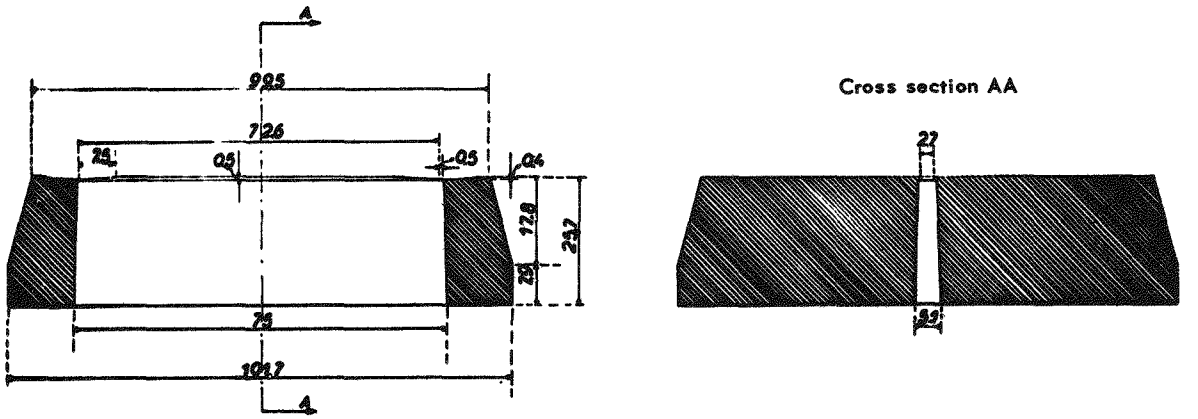

Figure 6. Extrusion matrix (dimensions in mm).

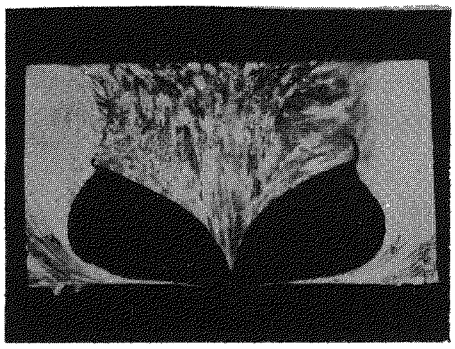

Fig. 7.- Front view of the extrusion tail showing inversion of flux (M: x 0.5)

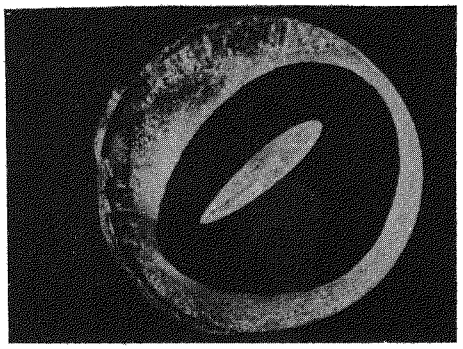

Fig. 8.- Front view of the extrusion tail with an inversion of the flux (M: x 0.5)

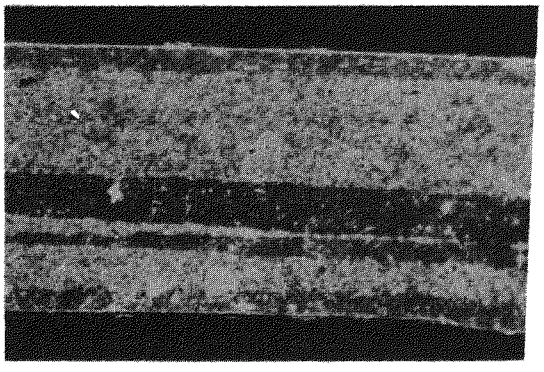

Fig. 9- Transverse cross section of a fuel element with central aluminum for flux inversion. (M: x 20).

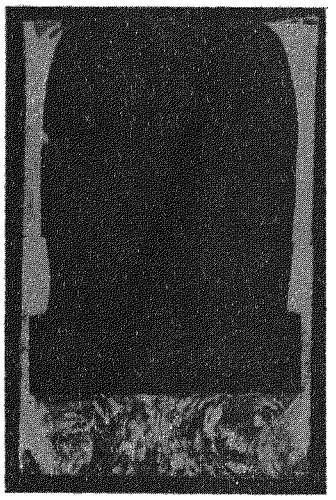

Fig. 10. - Fuel element with parallel walls partially extruded (M: x 0.10)

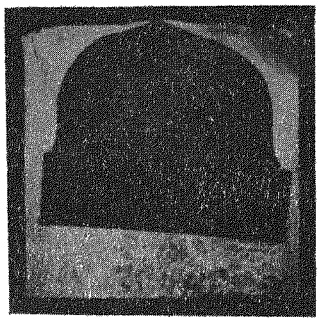

Fig. 11. - Half-extruded cartridge (M: x 0.3)

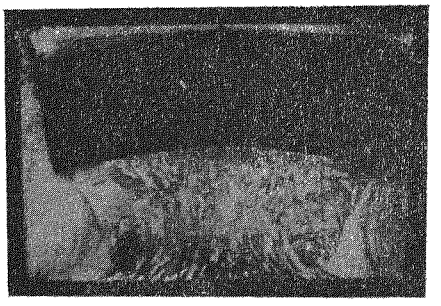

Fig. 12. - Extrusion end completely extruded (M: x 0.5)

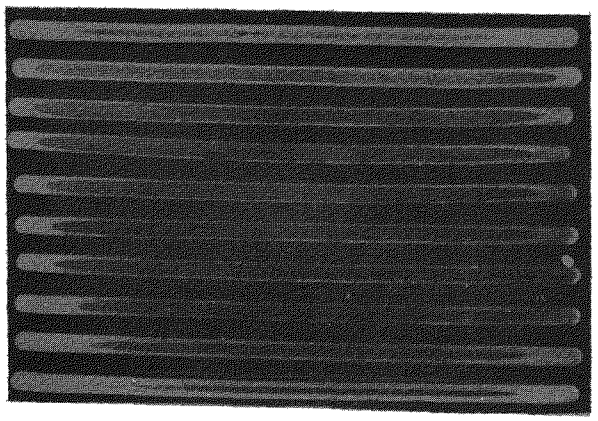

Fig. 13. - Transverse cross sections every 60 cm of extruded plate. (Actual size).

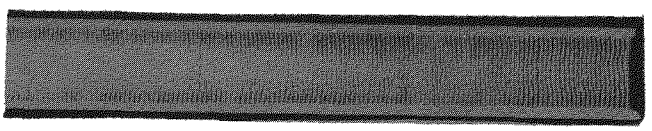

Fig. 14. - Plate with effect due to a very large friction plane

Fig. 15. - Defect due to temperature

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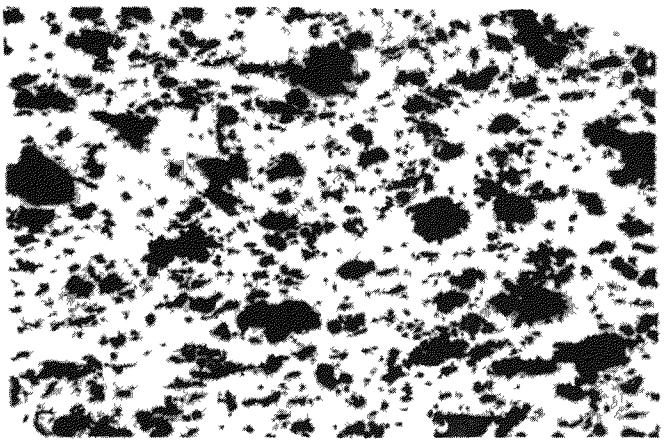

Fig. 16. - Microphotograph of a transverse cross section of a fuel element with 45% of natural U_3O_8.

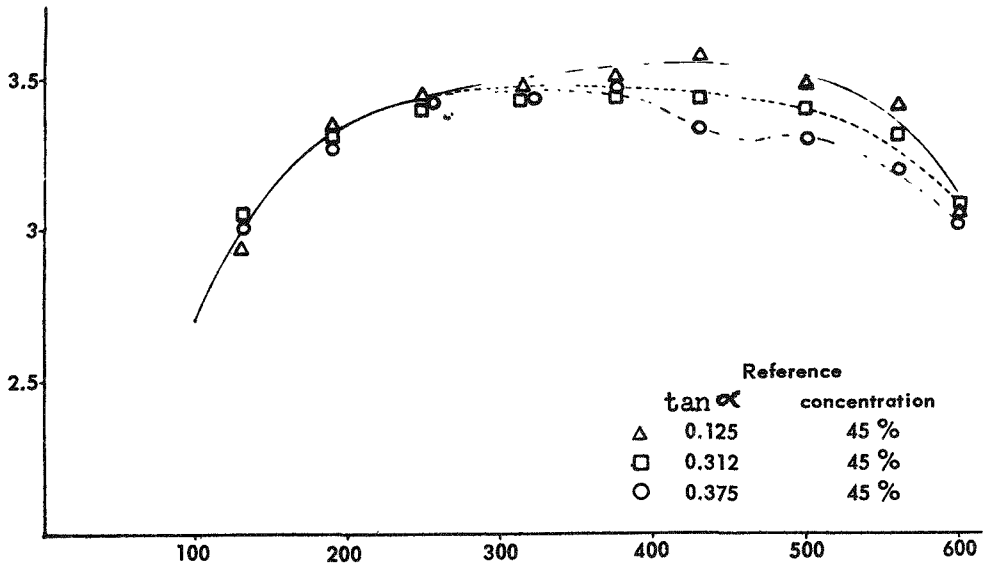

Fig. 17. - Density of the plate against length for a load of U_3O_8 of 45% and variable tan α

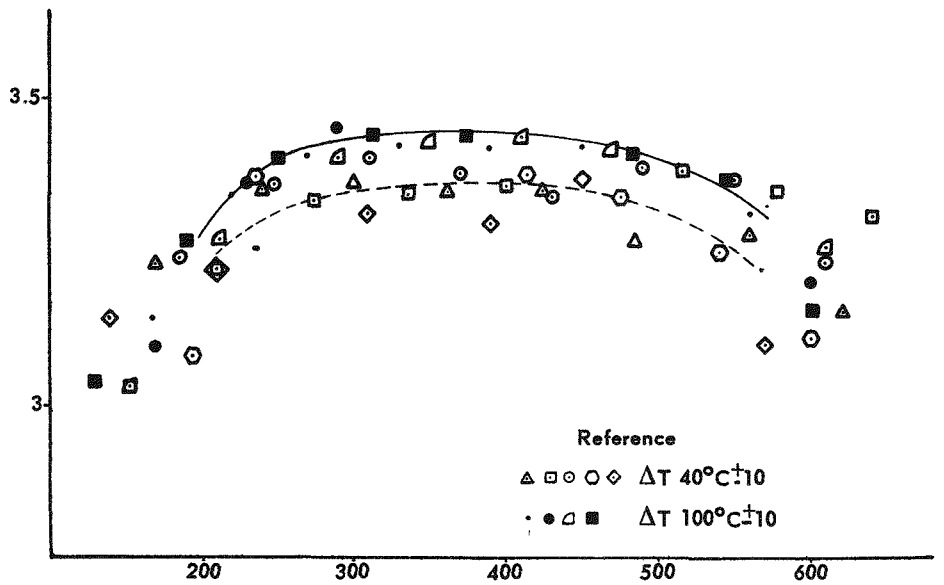

Fig. 18. - Density of the plate against the length for constant tan α, difference of temperature between the element and the bushing being variable.

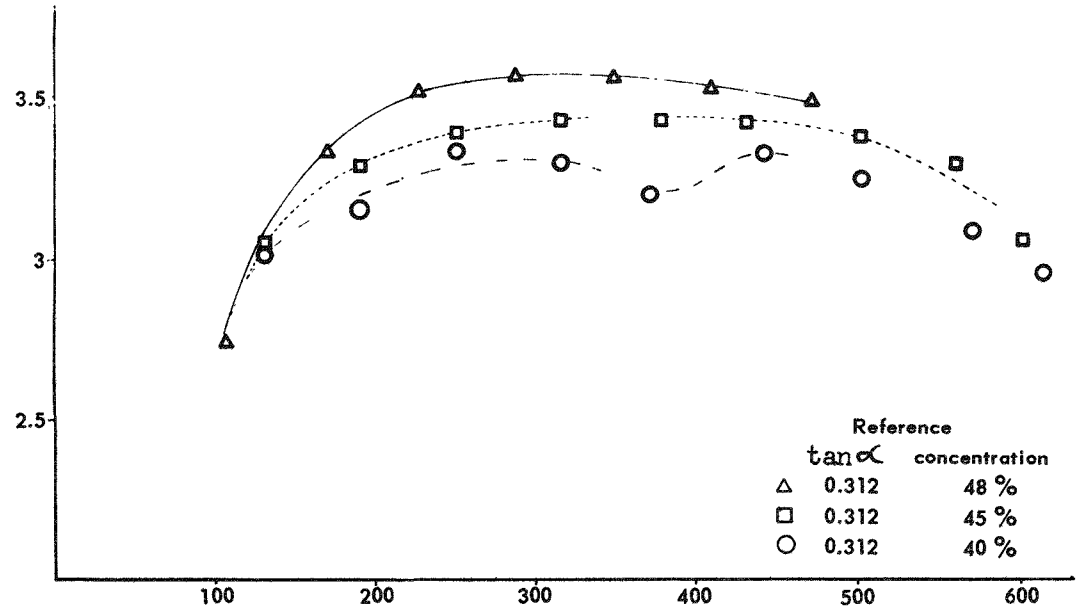

Fig. 19. - Density of the plate against length for U_3O_8 variable content.

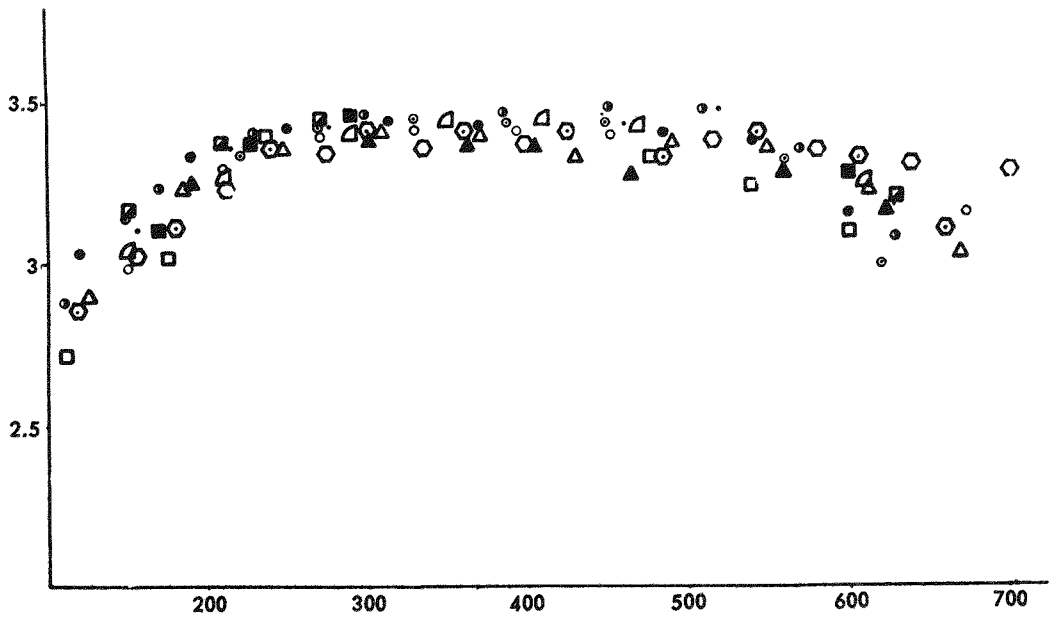

Fig. 20. - Density of plate against length for 3 fuel elements containing 45% U_3O_8 and tan α 0.312

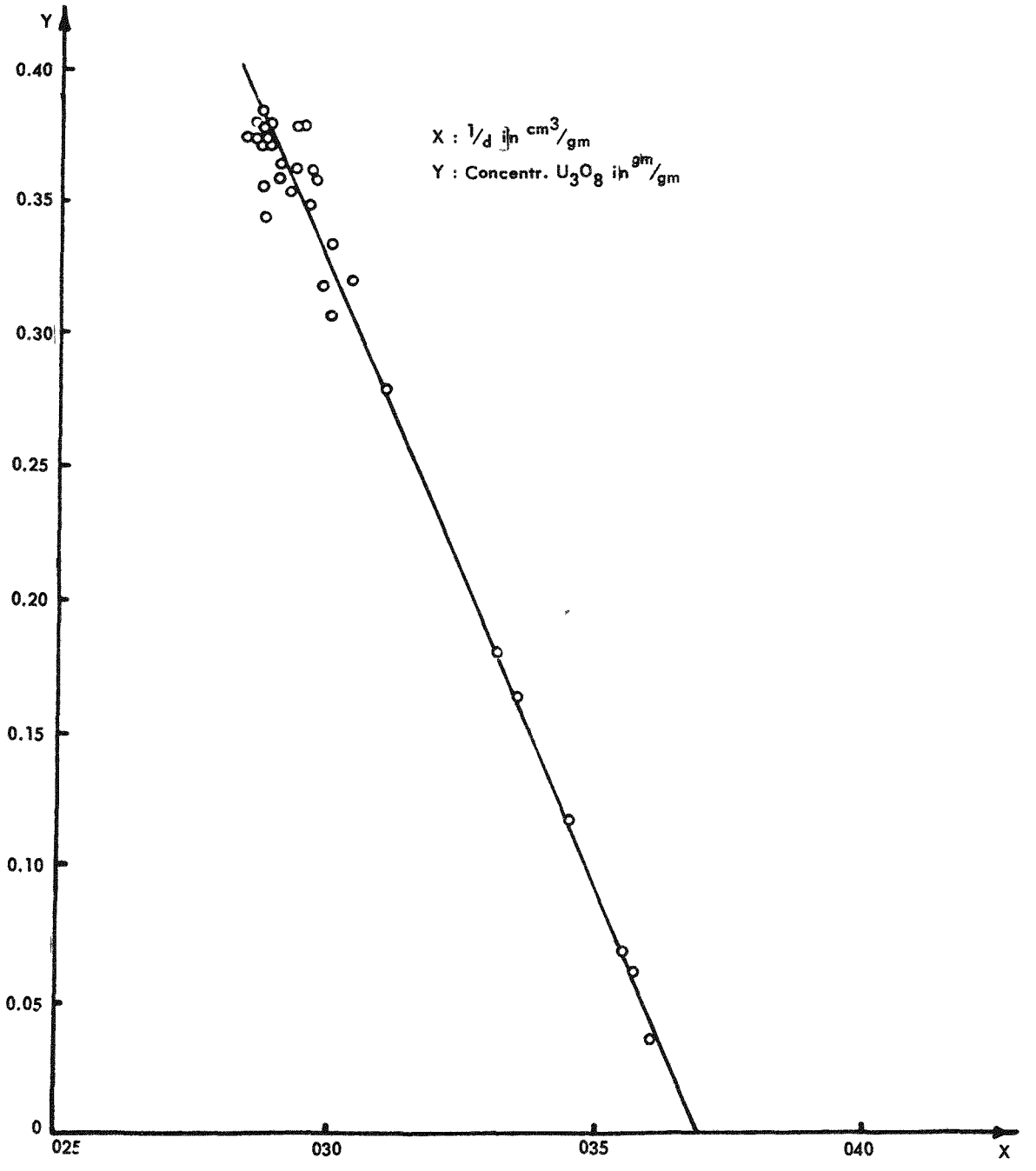

Fig. 21. - Concentration of U_3O_8 in Gm/Gm against the reciprocal of the density in cm^3/Gm.

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