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THE FABRICATION OF U233/ALUMINIUM FUEL PLATES FOR A SUB-CRITICAL ASSEMBLY - A SECOND REPORT

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ABSTRACT

Uranium-233 (as 938.66 g uranium dioxide) was fabricated into 176 full-size, 20 'half-size' and 2 'quarter-size' aluminium clad fuel plates. Each full-size plate contained 5 compacts and 'half-size' and 'quarter-size' plates contained 2 compacts and 1 compact respectively; a compact consisted of a cold-pressed dispersion of about 1.5 g of uranium dioxide in aluminium.

The technique involved mixing uranium dioxide and aluminium powders, pressing the compacts in a steel die, end-loading them into a prefabricated aluminium can and end-welding the can; following this the can underwent radiographic inspection for uniformity, decontamination, leak detection and acceptance tests, and final decontamination.

Maximum permissible radiation doses to hands and bodies of the operators were not exceeded.

CONTENTS

1. INTRODUCTION

In an earlier report (Bardsley and Ridal 1966) the authors described the fabrication of one kilogram of uranium-233 (as uranium oxides) into 205 aluminium-clad fuel plates. These fuel plates were required by the Physics Division of the A.A.E.C. Research Establishment as components of beryllia-based sub-critical assemblies. Such assemblies, with a range of fissile-fertile oxides, are being studied to determine buckling and fission ratios in BeO-moderated systems.

The fuel plates so prepared could only be handled without excessive difficulty for a limited time, because the β-γ dose rate from the plates was increasing at about 1 per cent per day owing to the build-up of radioactive daughter products of the 50 p.p.m. of U-232 in the uranium. The fuel plates were therefore chemically processed (Baillie and Cairns 1967) to produce U-233 dioxide with a much lower U-232 daughter product content and corresponding lower β-γ activity.

This report describes the fabrication of the reprocessed U-233 dioxide. The initial β-γ dose rate of the reprocessed oxide (160 mrem/h at 1 ft from one unshielded kg UO_2) was much lower than during the previous fabrication (700 to 900 mrem/h) and this allowed some simpler handling methods to be used. In addition, modifications were introduced into the process to overcome fabrication problems which had arisen previously.

2. SPECIFICATION AND MATERIALS

The fuel element requirements specified by the Physics Division are shown in Table 1; the U-233 distribution was achieved by cold-pressing mixtures of U-233 dioxide powder and aluminium powder to give compacts of size 2.285 x 1.085 x 0.023 in. The optimum-size assemblies could be produced from 160 full-size plates (5 compacts), 20 'half-size' plates (2 compacts) and 2 'quarter-size' plates (1 compact).

The U-233 was supplied in two batches as uranium dioxide with an oxygen to metal ratio of 2.030:1 and isotopic composition as shown in Table 2. It was a uniform brown non-agglomerated powder, calcined at 1500°C and sieved to 100 per cent -300 mesh. The aluminium powder used for mixing with the oxide was 99.99 per cent pure and -300 mesh in size. The aluminium used for the fuel plate can was 99.5 per cent pure (1-S grade) and typical impurity levels were as listed in Table 3.

3. NON-ACTIVE DEVELOPMENT

The major problem encountered during the earlier fabrication technique was the poor quality of the all-round welding (approximately 24 in. per plate). This was caused by U-233 oxide contamination of the weld zone during assembly, resulting

from the friable nature of the compacts.

To reduce this problem, the authors commenced non-active development of a partially fabricated aluminium can, with only one end open for loading and subsequent welding. Concurrently, the optimum powder characteristics and pressing techniques required to produce a high strength, non-friable compact capable of end-loading into the can were determined. The type of can developed is shown in Figure 1 and the results of the compact optimisation investigations are given in Table 4.

Strong, non-friable compacts for end-loading into suitable aluminium cans could be produced by either of the following techniques:

(a) UO_2 calcined 1 hr at 1500°C was sieved to 100 per cent -300 mesh. It was then added to aluminium powder (also 100 per cent -300 mesh) in the ratio 1.2 g UO_2 to 1.75 g Al, mixed by hand shaking, and cold-pressed at 25 t.s.i. into compacts 2.285 x 1.085 x 0.023 in.

(b) UO_2 was isopressed at 20 t.s.i., crushed and sieved to 100 per cent -300 mesh, sintered 1 hr at 1500°C, and sieved again. These particles were mixed with aluminium powder (100 per cent -300 mesh) in the ratio 1.2 g UO_2 to 1.75 g aluminium, and pressed at 25 t.s.i. to a size 2.285 x 1.085 x 0.023 in.

Although Method (b) gave marginally better compacts, Technique (a) was adopted because it involved fewer steps in the UO_2 preparation.

4. ACTIVE FABRICATION

The steps in the fabrication of the fuel plates are shown in Figure 2 and discussed below.

4.1 Mixing and Pressing of the Compacts

The U-233 dioxide was weighed into 1.2 ± 0.01 g lots in glass containers, and transferred to the pressing box where 1.75 ± 0.01 g of aluminium powder was added to each lot. Mixing was carried out by shaking the powders together until no segregated UO_2 regions could be seen on visual inspection; in addition radiographic examination of the pressed compacts produced from the material by this technique showed that the U-233 distribution was satisfactory (see Section 4.4).

The mixture was pressed in a hardened steel die at 25 t.s.i. to give a compact of size 2.285 x 1.085 x 0.023 in; accurate levelling of the powder was necessary before pressing to give a uniform compact. The resulting compacts were of high strength, non-friable, and could be handled without excessive care; they

were therefore stacked five at a time in metal trays, ready for assembly.

4.2 Assembly

Prefabricated cans were made in the open laboratory by edge-welding on two sides and at one end. They were then partially enclosed in a polythene bag to facilitate decontamination and were introduced into the welding box. Five compacts were end-loaded into each can using a metal plate as a ram; this plate was also used to 'size' the cavity in the can before loading commenced. Finally a $\frac{1}{4}$ in. wide aluminium spacer piece was introduced to form an end closure and then cropped with a guillotine, to give a smooth parallel end for welding.

4.3 Welding

This can was edge-welded using a jig (Figure 3) in which the plate was clamped between two copper heat rings, a 0.050 in. projection being allowed for welding.

The gas used for welding outside the glove box was pure argon (3.5 litres min^{-1}). A higher gas flow was necessary to achieve good quality welds inside the glove boxes probably because of the low pressure (- 1.5 in. w.g.) in the glove box. Helium gas mixtures, originally used for leak detection, were retained for glove box conditions because they have a more stable welding arc than pure argon.

Welding details were as follows:

Equipment:	T.A.D.-1-E.M.F. Alternating current, 200 amp, resistance type		
Current:	30 amp maximum, manually controlled through a variable resistance		
Process:	T.I.G.	Electrode:	$^1/_{16}$ in dia.
Nozzle:	in.	Glove Box Gas Flow:	10 ft^3 min^{-1} of argon 200 ft^3 min^{-1} of helium

The only significant problem encountered was caused during welding of the end by the abrupt change in cross section of the can; a rapid change in the welding current at this point was required to avoid 'burning off' the edges. Although this problem was not a major one, the can design should be optimised in any future fabrication.

All welds were visually inspected for large pin holes, or other obvious welding defects. Repairs, where necessary, were carried out before decontamination.

4.4 Radiography

The first 16 elements produced were radiographed to show that a uniform U-233 distribution was obtained; a typical radiograph is shown in Figure 4. This confirmed results obtained during non-active development which showed that no significant

variations in U-233 distribution occurred.

Welded plates were transferred to the decontamination box and wiped with dry paper tissues before removing the protective polythene bag. The plates were then transferred to a fume hood for final decontamination and leak detection; smear tests and α - counting results showed that at this stage the total U-233 surface contamination was less than 6×10^{-3} μCi cm^{-2}. After removing most of this contamination by abrading the surface with steel wool wetted with a water and Teepol solution, every element had a total U-233 surface contamination of less than 1×10^{-3} μCi cm^{-2}; this was well within the previously accepted limit of 3×10^{-3} μCi cm^{-2}.

Only one completed element had to be rejected because of an apparent leak; this however, was caused by a crack in the side of the aluminium can at the change of section and not by a welding fault.

4.6 Inspection and Acceptance

Final acceptance of the plates was based on an α-probe survey of the surface to determine any point sources of U-233 contamination in excess of 3×10^{-3} μCi cm^{-2} and a smear check to ensure that the maximum loose surface contamination did not exceed 1×10^{-3} μCi cm^{-2}; this technique ensured that all welds were sound.

The end-weld bead was then forged between flat steel plates to remove weld projections and to ensure that the plate would pass through a 0.050 in acceptance gauge.

The plates were enclosed in polythene bags and placed sixteen at a time in racks in the transport containers.

4.7 Accounting of Fissile Material

4.7.1 Initial fabrication and reprocessing

The original amount of U-233 supplied was 999.32 g and the amount allocated to the 205 fuel plates previously fabricated (Bardsley and Ridal 1966) was 994.59 g, this being the sum of the individual oxide weights used to manufacture the compacts. The estimated accuracy was ± 2 per cent because waste generated during pressing operations was recycled; when the fuel plates were chemically reprocessed a check on this was obtained.

After dissolution of the fuel plates and unused U-233, chemical analysis indicated 988.62 g of U-233 in solution instead of the theoretical content of

999.32 g of U-233. The indicated loss of 10.67 g of U-233 was within the accuracy of the analytical technique used; this was estimated to be ± 1.5 per cent, giving a possible physical error of ± 15 g. For convenience in accounting the indicated loss was allocated to the waste generated during the initial fabrication work and 'written off'.

After chemical reprocessing to produce U-233 dioxide the total U-233 contained was 967.22 ± 14.5 g.

4.7.2 Refabrication

Owing to the high strength of the compacted powders waste generated during the pressing of each compact in the refabrication process had to be chemically reprocessed before recycling. Chemical analysis showed that the waste produced in each pressing operation was 36 ± 1.5 per cent U-233 dioxide. During the accounting the U-233 dioxide allocated to each compact was adjusted on this basis. The overall accuracy of the accounting procedure was estimated to be ± 2 per cent and a check on this estimate will be carried out during subsequent chemical reprocessing.

4.7.3 Material allocation

The 967.22 g U-233 supplied for refabrication was accounted for as follows:

938.66 g	used in the fabrication of 176 full-size, 20 half-size, 2 quarter-size fuel plates.
23.11 g	unused
4.364 g	collected as recoverable waste and returned to store
1.09 g	collected as contaminated waste and buried.

4.8 Fabrication Time

The total time required to develop the method and fabricate the fuel plates was 8 weeks. This included 3 weeks for non-active development and 5 weeks for active fabrication of decontamination operations.

Non-active development commenced on July 5th, 1965 and ceased on July 22nd, 1965.

Active fabrication commenced on February 1st, 1966 and ceased on March 10th, 1966.

5. OPERATING HAZARDS AND CONTROL MEASURES

The major hazards involved in the handling of U-233 (in this case containing

50 p.p.m. U-232) are toxicity, criticality and radiation.

U-233 is a Group II radiotoxic substance with a maximum permissible concentration in air of 1.1×10^{-2} $\mu g\ m^{-3}$. This material, whether it contains U-232 or not, must therefore be fabricated under totally enclosed conditions to ensure that contamination is kept to a safe level. The fabrication process was carried out completely in glove boxes connected by a waist-high central tunnel (Figure 5); the pressure in the boxes was maintained at a negative value between 1 and 1.5 in w.g. Under these conditions, contamination of the atmosphere outside the glove boxes was undetectable (< 0.1 m.p.c.); minor incidents producing surface contamination (1 to 5 m.p.l.) occurred, mainly during the removal of partly decontaminated plates from the boxes for further cleaning.

Criticality control during operations was maintained by limiting the amount of material in any box to 100 g, this amount being recorded by a disc system on each box. The only exception to this 100 g limit was the primary storage and weighing box which was cleared to contain up to 500 g of U-233; all liquids and potential moderators were excluded from this box and the 100 g lots of U-233 dioxide in polythene bottles were stacked rigidly 1 foot apart in a simple rack system. Completed elements were stored and transported in a box of the type shown in Figure 6. The box was approved for the carriage of sixteen elements only, the number being limited by the geometry of the internal rack.

The main sources of radiation hazard in processing were:

(a) gamma radiation (up to 2.6 MeV) mainly from the decay of U-232 as shown in Figure 7,

(b) beta radiation (up to 2.7 MeV) from the decay series (particularly from Bi-212 and Tl-208), and

(c) neutron radiation from the U-233 (α, n) reaction with aluminium.

Ten days after separation the β-γ dose rate at 1 ft from an unshielded kilogram of the reprocessed U-233 dioxide was equivalent to 160 mrem/h. Owen (1964) measured the β component as about 65 per cent of the total dose, which indicates that the γ component of the dose rate of this material was 60 mrem/h; this is shown in Figure 7 and compared to that predicted by Arnold (1962). In view of this low β-γ dose rate, fabrication of the fuel plates was carried out without any form of lead shielding to protect the operators; the total body dose to each operator was controlled only by minimising the fabrication time of each fuel plate. It should be noted that although the total fabrication time was five weeks a freshly reprocessed batch of UO_2 was supplied every three days, so that the material being fabricated was never more than 5 days old.

The average body doses for various operations shown in Table 5 are similar to those measured during the initial fabrication of U-233 oxide. The earlier fabrication commenced 120 days after chemical reprocessing and used lead-shielded enclosures to reduce the dose rate for some operations.

The maximum β-γ doses received by any operator for the five week fabrication period were:

Wrist and finger 2100 mrem gamma, 1020 mrem beta,

Body dose 190 mrem gamma, 240 mrem beta.

The total fast neutron dose during the whole of this period was 100 mrem.

6. SUMMARY

Direct fabrication of 176 full-size, 20 'half-size', and 2 'quarter-size' U-233 fuel plates using U-233 dioxide containing 50 p.p.m. U-232, was carried out under α-glove box conditions. Fabrication commenced approximately 10 days after chemical separation. Approach distances and handling times were optimised to reduce radiation doses to the operators. The maximum permissible β-γ doses to the hands or body (1500 mrem/week and 100 mrem/week respectively) were not exceeded.

The operation was carried out in 8 weeks, including the 3 weeks required for non-active development.

No major fabrication problems occurred. However, slight changes in can design would facilitate end-welding in the glove box and should be incorporated into any future refabrication.

7. ACKNOWLEDGEMENTS

The authors wish to acknowledge W. Buykx who assisted in much of the fabrication work, and P. Gillespie who carried out the radiographic examination.

8. REFERENCES

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Baillie, M., and Cairns, R. (1967). - The Recovery and Purification of U-233 from Aluminium Clad Uranium Oxide Fuel Elements - paper in preparation.

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Owen, F. E. (1964). - Beta-gamma Dose Rates from U-232 in U-233. H. W. 81964.

TABLE 1

FUEL ELEMENT SPECIFICATION

Element Specification:

Finished dimensions 11.850 in. x 1.350 in. x 0.050 in.

Internal dimensions 11.450 in. x 1.090 in. x 0.025 in.

The element is to contain not less than 4.5 g and not more than 6.0 g U-233 distributed uniformly along the length.

Number of Elements:

Not less than 160 'full-size' elements with 20 'half-size' (2 compacts) and 2 'quarter-size' (one compact).

TABLE 2

ISOTOPIC COMPOSITION

Isotopic Specification: (all figures given as percentages)

	U-233	U-234	U-235	U-236	U-238
Batch 1	96.99	2.35	0.212	0.028	0.418
Batch 2	96.98	2.35	0.215	0.021	0.434

U-232 content 50 parts per million U-233

TABLE 3

SPECIFICATION OF ALUMINIUM SHEET (1-S GRADE)

USED FOR FUEL CANS

Element	Nominal Order (%)
Si	0.14
Fe	0.43
Cu	0.027
Mn	0.040
Mg	0.005
Zn	0.009
Cr	0.001
Ti	0.011
Ni	0.003
Pb	0.004
Bi	0.001
V	0.007
Ga	0.008
B	0.001
Be	< 0.001
Sn	< 0.001

TABLE 4

COMPACT OPTIMISATION RESULTS

UO_2 Specification	Aluminium (mesh size)	Weights	Mixing Methods	Pressing Pressure	Compact Thickness	Observations
Calcined 1 h at 1500°C 100% -300 mesh	- 120 + 200	1.2 g UO_2 1.75 g Al	Hand Shaking	25 t.s.i.	0.022 ± 0.001	The coarse Al powder caused poor UO_2 distribution, compact weak.
Calcined 1 h at 1500°C -76 + 300	100% - 300	"	"	"	"	Good compact but non-uniform distribution, UO_2 particle size too large.
Calcined 1 h at 1500°C 100% -300 mesh (technique (a) in text)	100% - 300	"	"	"	"	Good compacts strong with acceptable UO_2 distribution
Calcined 1 h at 1000°C 100% -300 mesh	100% - 300	"	"	"	0.02 ± 0.001	Poor compacts, weak with non-uniform distribution, UO_2 probably too fine.
Isopressed 20 t.s.i. crushed 100% -300 mesh	100% - 300	"	"	"	"	Weak compacts not acceptable.
Isopressed 20 t.s.i. crushed-sintered 1 h at 1500°C 100% -300 mesh	100% - 300	"	"	"	0.022 ± 0.001	Excellent compacts, density measured as 94% of theoretical.
As received UO_2 powder (Calcined 1 h at 1000°C)	- 300	"	by grinding	"	0.023 ± 0.001	Very poor compacts, similar to those obtained in earlier U-233 fabrication.

TABLE 5

AVERAGE β-γ DOSE RATES TO THE BODY

FOR INDIVIDUAL OPERATIONS

Weighing	5 mrem/h	Assembly	5 mrem/h
Mixing	3 mrem/h	Welding	2 mrem/h
Pressing	3 mrem/h	Decontamination	7 mrem/h

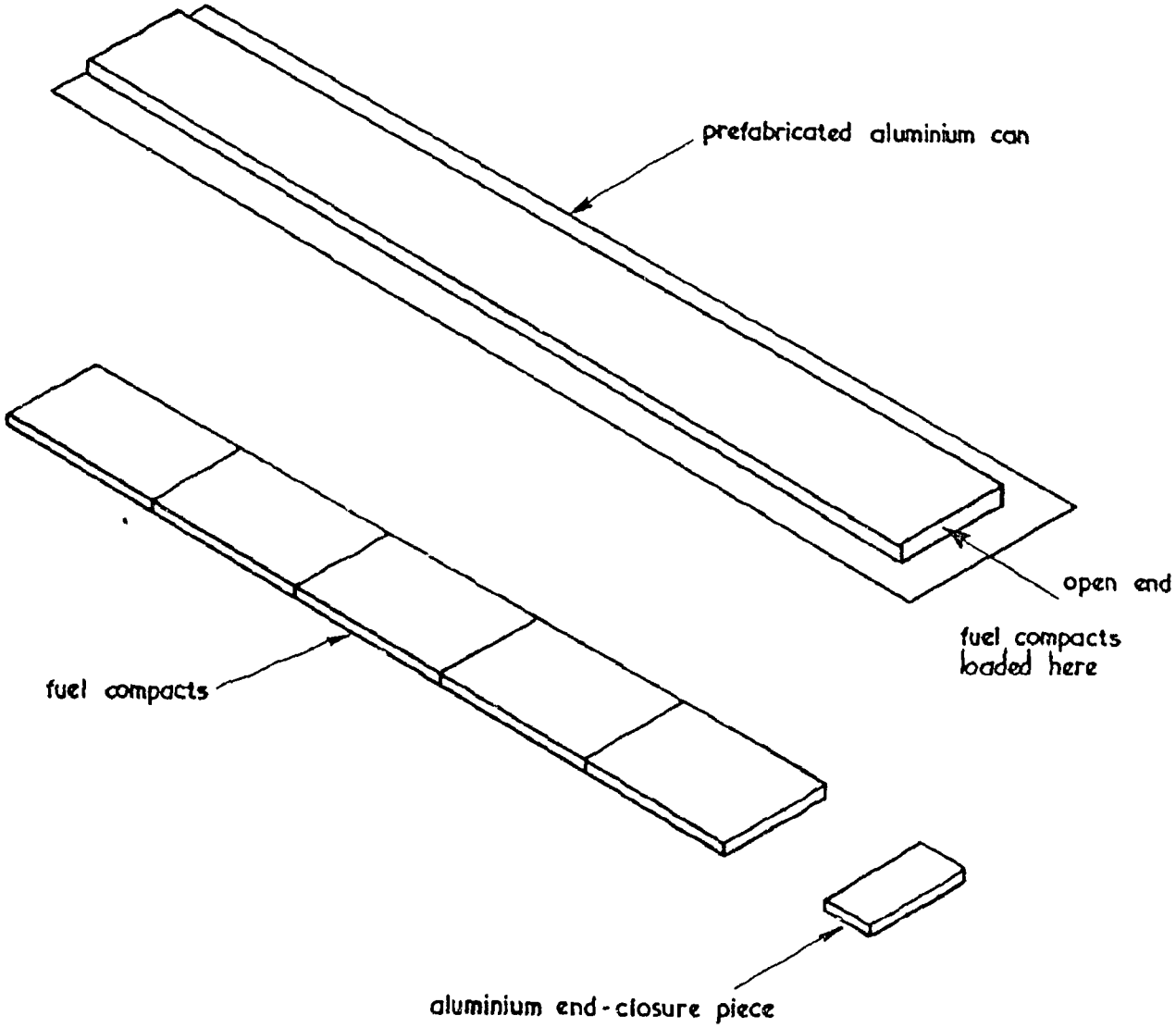

FIGURE 1. METHOD OF ASSEMBLY OF FUEL PLATES

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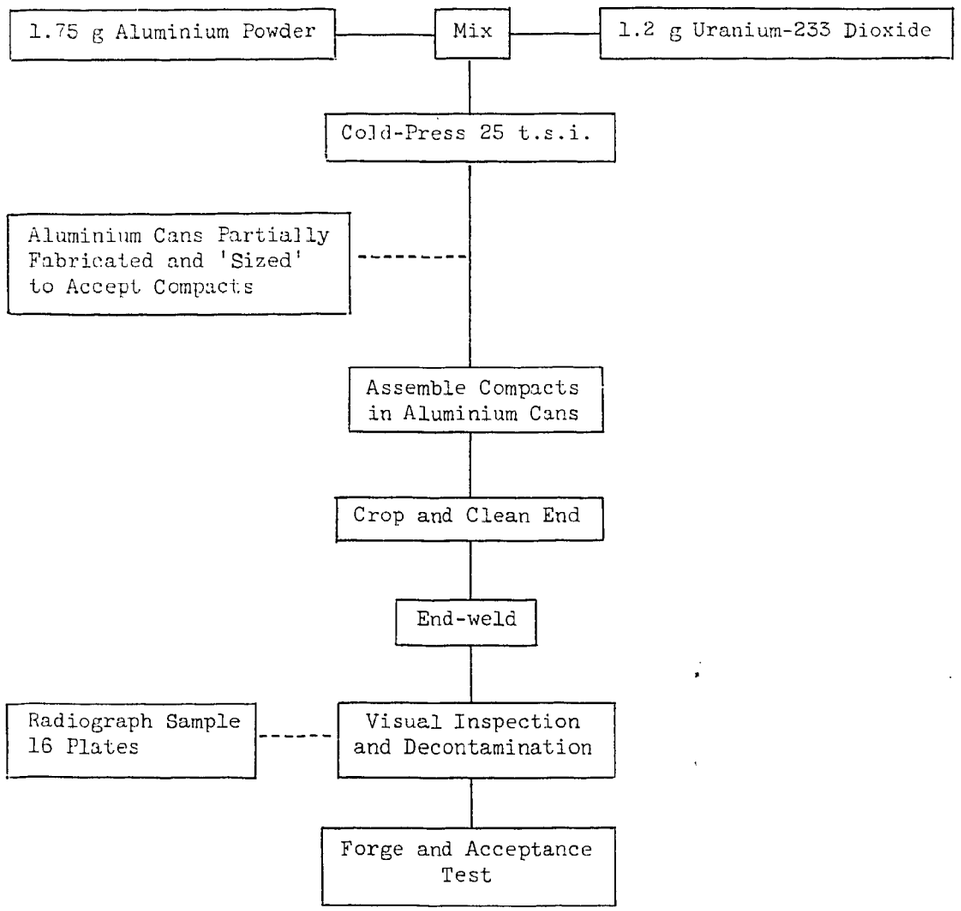

FIGURE 2

FLOW SHEET FOR ACTIVE FUEL PLATE PRODUCTION

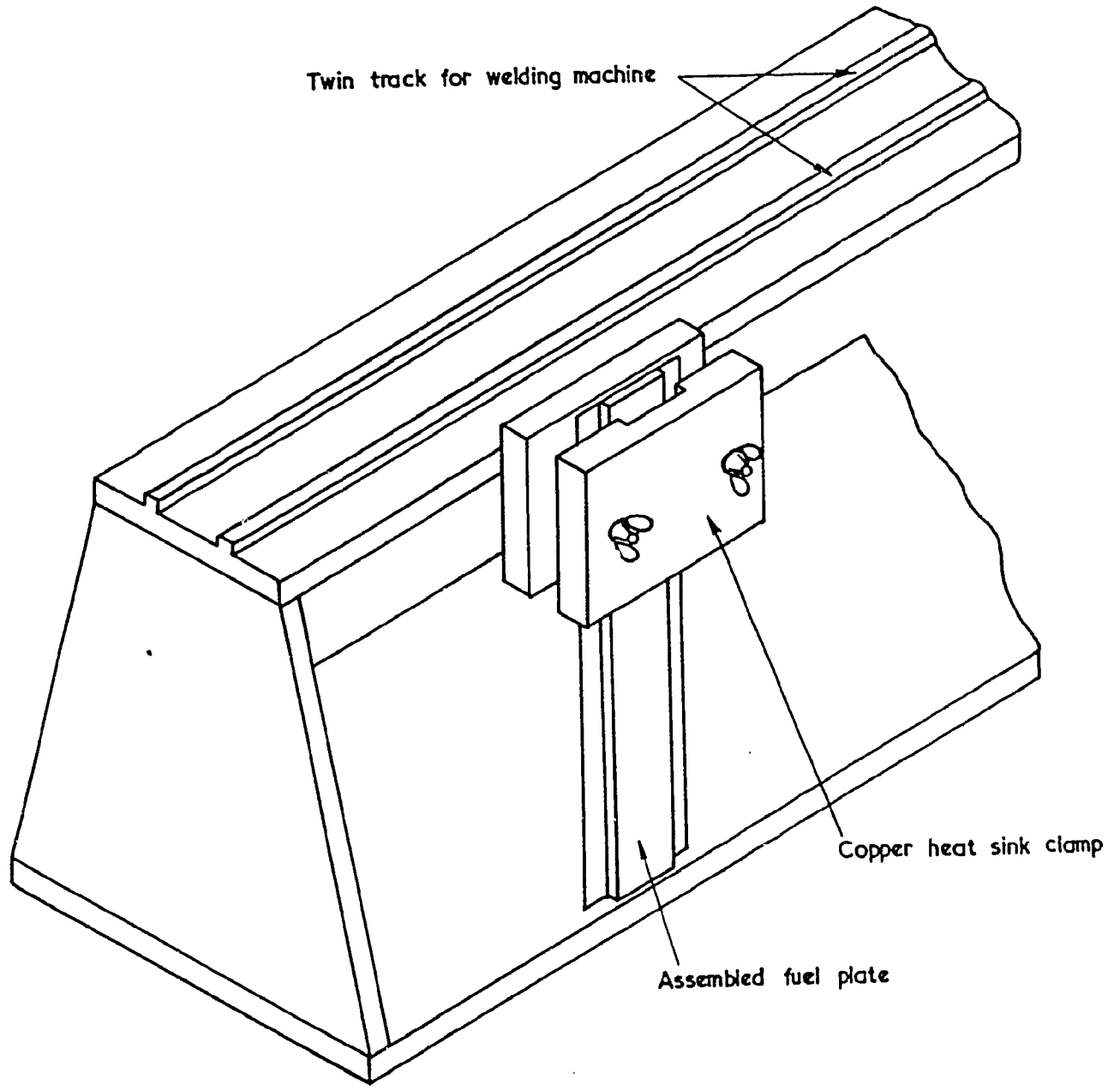

FIGURE 3. JIG FOR END-WELDING OF FUEL PLATE

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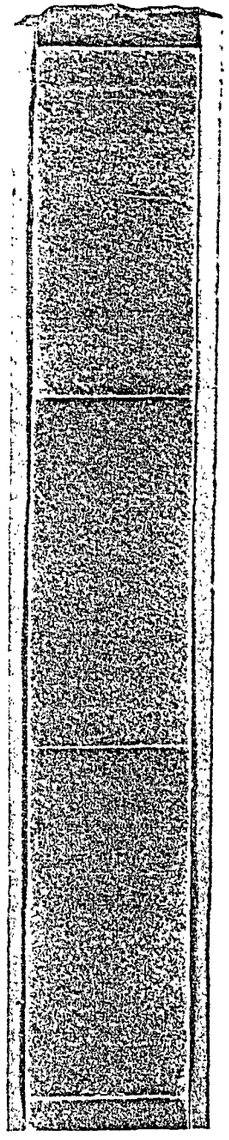

FIGURE 4. PORTION OF A TYPICAL RADIOGRAPH OF A COMPLETED U-233 FUEL PLATE

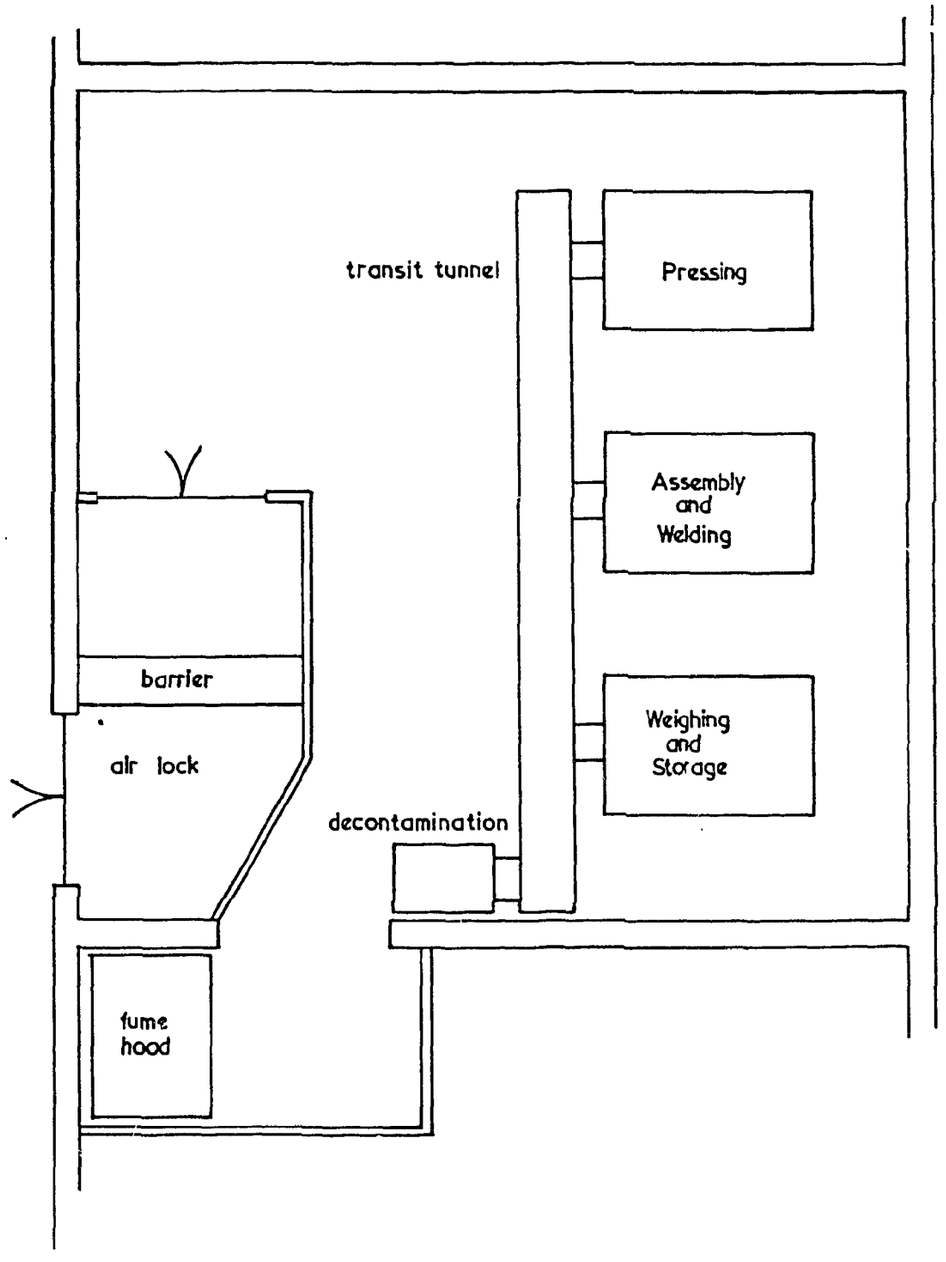

FIGURE 5. SCHEMATIC LAYOUT OF GLOVEBOX SUITE

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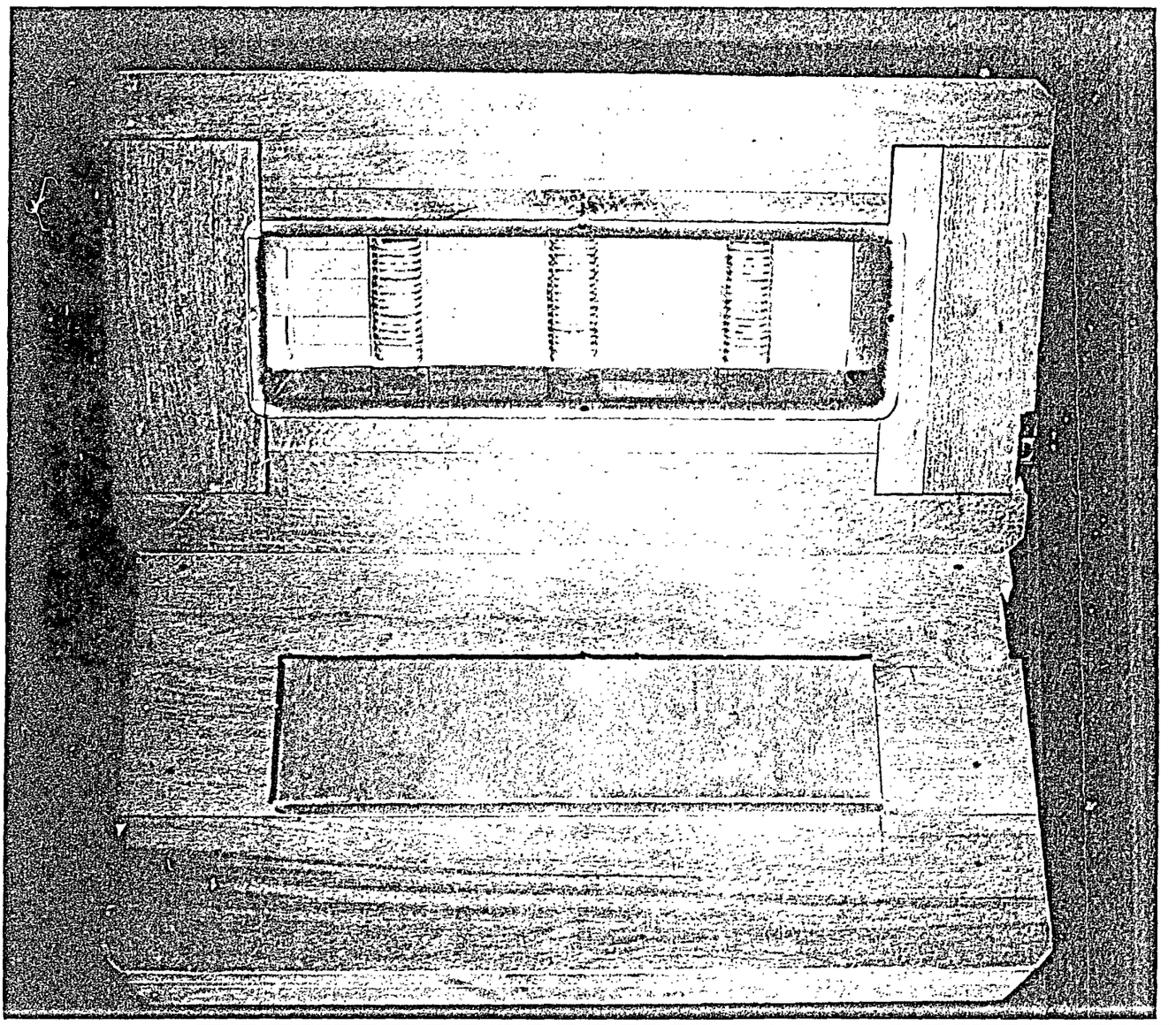

FIGURE 6. TRANSPORT CONTAINER

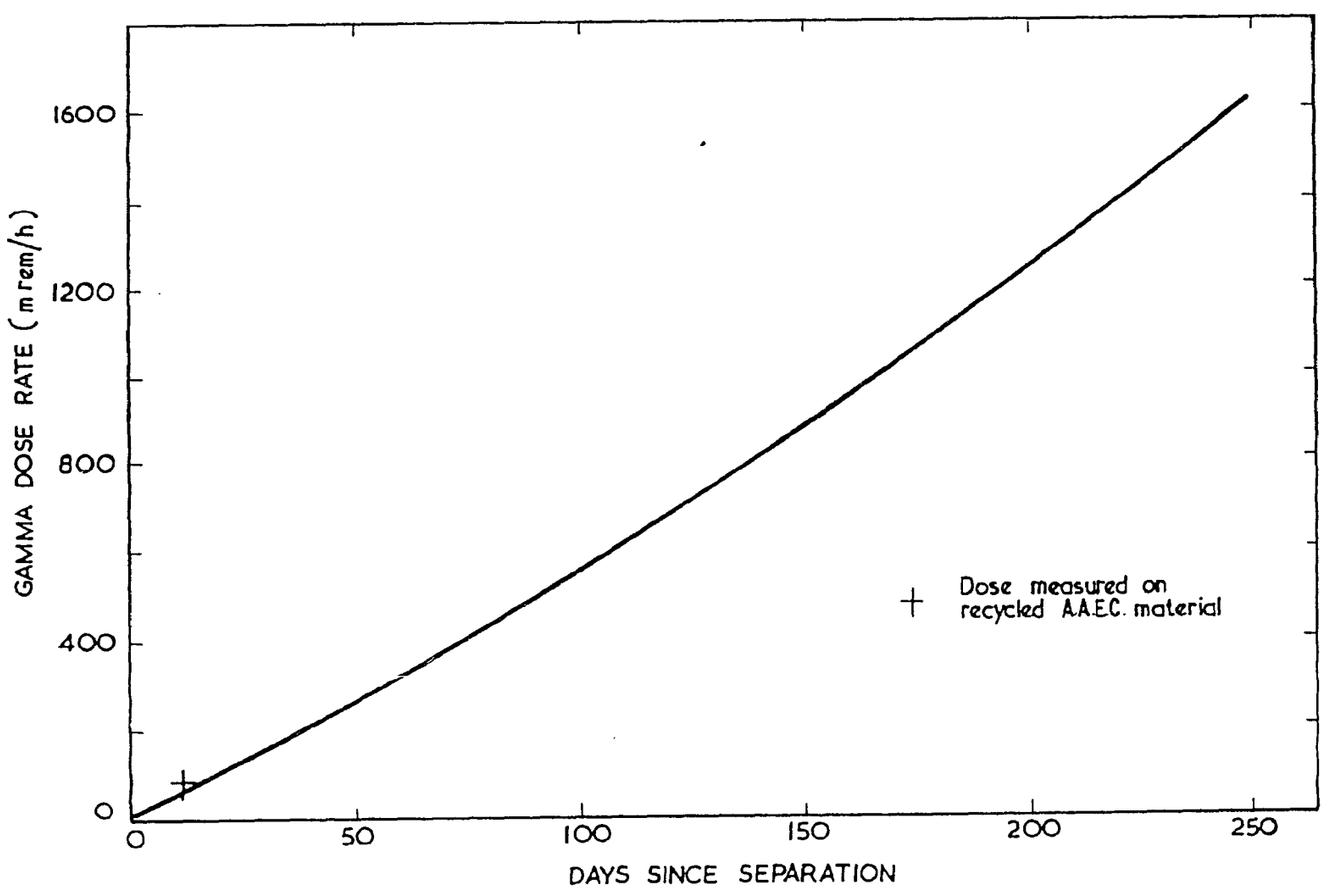

FIGURE 7. GAMMA DOSE RATE v. TIME FOR 1kg U-233 AS UO_2 (50 p.p.m. U-232)
(Based on Arnold 1962)

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