DEVELOPMENT STUDIES FOR ARGONAUT REACTOR FUEL PLATES FABRICATION

T.D. Souza Santos, H.M. Haydt and C. Trench de Freitas
Instituto de Energia Atomica, Sao Paulo, Brasil.

1. INTRODUCTION

The actual fabrication of the fuel elements for the Argonaut reactor of the Instituto de Engenharia Nuclear, Rio de Janeiro, Gb, has been ascribed to the Division of Nuclear Metallurgy of the Instituto de Energia Atómica. The Division was established early in 1962 with the scope of carrying out experimental studies in nuclear metallurgy in general and fuel element fabrication particularly under the program of the Comissão Nacional de Energia Nuclear.

The Division took full responsability of the complete fabrication of the Argonaut fuel elements, starting with the required materials (20% enriched U_3O_8 powder, obtained from the U.S. Atomic Energy Commission, under the present agreements; aluminum powder imported from France; and aluminum bars and plates fabricated under specifications by a local Brazilian integrated plant).

Many difficulties had to be overcome in the earlier stages of the development work, chiefly those related to irregular and erratic behaviour in the roll bonding of the picture frame sets.

The paper describes the experimental work carried out with cermets of U_3O_8 of natural isotope content and aluminum powder. The development work was aimed directly towards the determination of the parameters from which the details of the manufacturing procedure could finally be established, before the actual fuel plates were to be produced. A complete mock-up fuel element with natural isotope content was produced, both to check every important step in the fabrication process and to supply the essential experience for accountability problems.

2. SELECTION OF THE FABRICATION PROCEDURE

The process originally adopted by the Argonne National Laboratory (1) for the production of the Argonaut fuel plates was the extrusion of a mixture of U_3O_8 and aluminum powder, in aluminum containers, with proper devices to prevent the cladding from flowing into the core material at the trailing end. A long strip with the desired cross section was accordingly obtained, from which the actual plates were cut to proper sizes. It has been found (3) that when oxide was substituted by UO_2 there was an appreciable rate of reaction between UO_2 and aluminum.

Such technique could be easily followed here, in one of the large aluminum processing mills of the São Paulo area.

Although such procedure has been used for the production of acceptable plates, it does present an inherent inconvenience, namely the absence of cladding at the ends of the cut plates, as the core is necessarily exposed at the places where the long strip was cut. Although this defect was not found to be particularly important under low power operation, the authors decided that a complete cladding would be a highly desirable feature for the IEN Argonaut fuel elements since 10 KW (thermal) power was contemplated. Consequently, this easy approach to the fabrication procedure was dropped and two other alternatives were studied, viz.: 1) roll bonding of picture frame sets with a thick cermet inside; and 2) cast cladding the cermet with a suitable aluminum alloy into a specially designed ingot mold, following the successful idea developed by Bergua et al ( )(3) at Argonne National Laboratory.

Many difficulties were found during the earlier stages of the experimental work with picture frame sets, chiefly those related to improper bonding of the components during hot rolling operations. These difficulties were later proven to be associated with faulty filling of the insert space, inadequate side welding of the assembled set and to retained humidity in the core, which was responsible for large blisters in the final rolling operations.

The feasibility of the cast cladding alternative was also extensively studied. Although some excellent plates were obtained, including several ones with only about the half of the desired core thickness, the rather strict specification for face cladding thickness do not tolerate even small casting defects. These defects, due to small dross inclusions trapped in the solidifying mass, were probably sufficiently severe to cause either local exposures of the core or abnormal rates of corrosion in reactor operation.

Metallographic examination of cut samples has shown (4) that perfect bonding between core and cladding existed and that very uniform dispersions were produced. Since however no effective way was found for controlling those casting defects, this alternative was put aside.

Insisting in the roll bonding of picture frame sets, a considerable amount of development work has been done, and finally, the process has been established as a safe and reliable way to produce plates which fully meet the dimensional, physical, structural and corrosion resistance requirements.

3. SUMMARY OF THE FABRICATION PROCEDURE

Fig, 1 depicts the main steps of the fabrication procedure as evolved from the experimental development work, carried out at the Division's laboratories.

The details of the most important operations will be presented in the following chapters. The flowsheet indicates some of the extensive controlling operations carried out during mock-up plates fabrication.

These steps were critically scrutinized in the course of a complete experimental run which allowed the production of 17 complete plates, to check the final fabrication details as well as the accountability problems involved. Those plates were subsequently assembled in a mock-up fuel element, as it will be shown.

4. PRODUCTION OF THE COMPACTS

The compacting behaviour of the charge mixtures made out of various proportions of natural isotope content U_3O_8, produced by the Division from nuclear grade purity ammonium diuranate, and aluminum powder, has been thoroughly investigated, by varying: a) the time, temperature and mass of the ammonium diuranate charge in the calcining operation for the production of U_3O_8; b) the fineness of the produced U_3O_8; c) the fineness of the aluminum powder; d) the conditioning operation, done in large rubber lined jars with hard rubber balls; e)charge and die lubrification; and f) the die geometry and compacting pressure.

The first experimental determinations were made with a die for the production of 10 mm diameter cylindrical pellets. Once the essential data were obtained, larger rectangular based dies were used for the experimental production of cermets whose largest dimension was close to the desired width of the core; such pellets were assembled edgewise into the frame and have thus enabled the experimental determination of the spreading coefficient of the core in the hot rolling operations. These data were of importance for the design of the final die, which is to be used for the actual fuel plate production.

The behaviour of the cermets in the final mock-up plates, as well as during the further fabrication steps, was the criterion for determining the set of conditions in cermet preparation which should be better used.

The results have shown that strict control of the calcining step, both of temperature and of time, was essential to afford reproducibility of the compacting step results. It has been found that calcining under 750°C during 2 hours produced a U_3O_8 powder which accounted for rather severe blister formation in the final annealing operations of the cold rolled plates. An electric muffle furnace was used for the calcining of the ammonium diuranate, loaded into two dishes, each of which with the capacity of 2,5 kg of ammonium diuranate. Once cooled the unloaded charge, it was sieved through 200 mesh, the oversized fraction being diverted to other studies.

Aluminum powder has also been classified in a 100 mesh screen. Coarser aluminum powder has shown the trend of producing non uniform dispersion in the final rolled thin core.

The conditioning step is also of prime importance. The first tests done in a polyvinyl jar with hardened steel balls gave erratic behaviour in the final roll bonding with the face plates. The best results were obtained preparing the charge in a 3,5 dm^3 rubber lined jar, tightly closed, into which a constant load of rubber balls were put together. Wilkinson (5) has shown the superiority of the results obtained with rubber stops instead of steel balls in the charge preparation of U_3O_8 and aluminum.

The pressing step has been previously done in fully lubricated dies, through a solution of parafine or stearine in dehydrated alcohol. As some blisters found in the preliminary steps of roll bonding were possibly due to the evolution of water vapor absorbed by residual lubricant in the vicinity of the compact walls, it has been thought better to avoid entirely this step.

The influence of the pressure on the apparent densities of the compacts has been determined: a) for cylindrical pellets of 10 mm of diameter; b) for prismatic pellets of 10 x 60 mm of basis; and c) for prismatic pellets with basis of 32 x 60 mm. The following charge mixtures of minus 325 mesh U_3O_8 and/or 100 mesh aluminum powder were studied: a) U_3O_8 alone; b) 60% U_3O_8 and 40% Al; c) 50% of each; d) 40% U_3O_8 and 60% Al; and e) Al alone.

The obtained results were plotted in Fig. 2. In this graph have been also plotted the results obtained with 50% mixtures of enriched (20% U-235) U_3O_8 and aluminum.

The results show that there is no appreciable influence of the die geometry (the results for enriched U_3O_8-Al mixtures are for 22mm cylindrical compacts), as the scattering of the values are generally within 2% of the average values. For the range of pressures exerted, a linear correlation exists when log d is plotted against log p, d and p being, respectively, the apparent densities and the effective pressures.

The results for those mixtures can be represented by the equation

$$\log d = \log a + 0{,}141 \log p \qquad (1)$$

where d represents the apparent density, in g/cm^3, a is a constant (apparent density for 1 t/cm^2 compacting pressure) and p the compacting pressure, in t/cm^2.

Fig. 3 represents the calculated densities for constant pressures plotted against the charge composition. This graph affords an easy way to select the charge composition to obtain a given density under specified compacting pressure.

It has been found that the presence of even small cracks in the compact would impair the plastic behaviour of the pre-sintered cermet during roll bonding. Due attention was paid at the conditioning step so that the charge could be pressed under low pressures for not losing its inherent hot plasticity and still reaching the required density, dictaded by mass requirements of the plate.

The experimental results obtained in the course of experimental plate production supplied the spreading coeficient of the core during hot and cold rolling, to meet the design specifications of the final thin core. With those data it has been possible to design the final large prismatic die, through which the actual single pellets for the Argonaut plates are being produced.

5. PRE-SINTERING OF THE COMPACTS

An important step introduced in the fabrication procedure was the pre-sintering of the compacts, under argon atmosphere at 500°C, in a continuous electric tubular furnace. This furnace was designed by the Division and built by a local manufacturer.

Slow heating to the sintering temperature and further slow cooling to room temperature has been provided to avoid cracks due to the thermal gradient stresses. The operation is done in large rib-bottomed graphite boats of special design to avoid distortions during the operation.

The pre-sintering does not alter substantially the cermet density but improves appreciably its high temperature plasticity during the roll bonding operation. It is effective also in driving off the residual moisture and volatile constituents left by the lubricant added, which, if present, will cause blister formation in the rolling operations after the bonding has been achieved.

The rates of heating and of cooling are of about 600°C/hr and 400°C/hr respectively and the charge remains about 60 minutes in the maximum temperature, of 600°C. Final cooling to room temperature, is afforded by the long water-cooled chamber at the discharge end of the furnace.

Metallographic examination of the cermets after pre-sintering did not show any evidence of reaction between U_3O_8, and aluminum having taken place.

6. ASSEMBLY OF PICTURE FRAME COMPONENTS AND ROLL BONDING

The pre-sintered cermets were wrapped around with dead annealed 0,10 mm thick 1100 aluminum foil before hand set into the frame component, machined from 1100 12,3 mm thick pickled plate. Special precautions were adopted to insure complete freedom of surface defects both in the frame and in the cover sheets cut from 2,2 mm 1100 aluminum plates. Machining of the frame was carefully done to assure a tight fitting of the wrapped cermet into the frame opening. For a time evacuation of residual air has been provided by evacuating through cylindrical holes drilled at the trailing end of the frame. It has latter been found that this step was unnecessary.

Before the assembly of the components, the aluminum alloy parts were thoroughly pickled, washed and dried.

A special jig was designed to assemble the frame with its core insert and the two cover plates in the proper position before welding under argon arc with tungsten tip. The welding was satisfactory and metallographic examinations did not disclose cracks or entrapped oxide.

The assemblies once welded were soaked for 45 minutes in a specially designed muffle furnace, electrically heated, at the temperature of 590°C. Strict control of temperature, of soaking time and location of the assembly within the furnace, are necessary to avoid defects during hot roll bonding operations.

As stated before, for the design of the die to be used in the actual Argonaut fuel plates, it was necessary to determine the core spreading coefficient after the hot and cold rolling operations. The first experimental plates had one single 32 x 64 mm, variable height cermet placed lengthwise into the frame. After known its behaviour, new plates were produced with three adjoining cermets, side by side, so that their total length was 96 mm with 64 mm in width, fairly close to the required dimensions of the final core. These three adjoining cermets had the same total mass of U_3O_8 required for the actual Argonaut fuel plates. The final data obtained from these plates, rolled down to the specified final thickness, supplied the required information on the spreading coeficient for the final design and construction of the compacting die which is to be used for the production of the actual fuel plates. Experimental plates produced with this final die have shown that in the rolled plates the core width does not present fluctuations of more than 0,1 mm of the expected.

The hot and cold rolling operations were done in the Stannat Mann four-high rolling mill. It has been found experimentally that too heavy passes tended to warp the plates unduly; that too light passes would difficult the roll bonding and work harden the stock at a fast rate. The hot rolling is better accomplished with one light starting pass followed with fairly heavy passes to assure the complete bonding of the components.

After hot rolling is completed when the plate is 3,5 mm thick, a light anneal is done in an electric muffle furnace at the temperature of 580°C during 20 minutes, after which final cold rolling down to the final specified thickness is done.

7. INSPECTION AND FINISHING OF THE MOCK-UP PLATES

After the final cold rolling, the plates are thoroughly checked for any eventual defects and the actual core position is determined by autoradiography. With the fabrication procedure previously described, there are no end defects which could impair the performance of the fuel plates in reactor operation.

The final cutting operations by edge and end trimming is done carefully in a Do-All type mechanical saw. A special jig fixture has been provided for the finishing of the cut plates into the final specified dimensions. The plates are then cleaned first with solvents for removal of residual lubricating oil, pickled, and then throughly washed. The plates are subsequently punched for the openings, in a special jig fixture to allow complete recovery of the punched out disks containing the core. Finally, each plate is marked with proper final identification marks.

Final inspection is done in view of: a) dimensional tolerances; b) uniformity of core distribution; c) uniformity of side and end claddings; d) warpage. During the development work many final plates were subjected to accelerated corrosion tests in de-ionized water at 65°C during 144 hours. Others were cut for metallographic examination of: a) uniformity of core structure; b) bonding of the core to the side cladding; and c) measurements of the effective cladding thickness.

The fig. 4 shows the microstructure of the core in a longitudinal section of the plate, parallel to the rolling direction. It shows the quality of the bonding achieved and the absence of inclusions which could impair a perfect bond between the core and the cladding.

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8. CONCLUSIONS

1. The fabrication of fuel plates completely clad at their ends for an Argonaut reactor which is expected to operate at the 10 kWt power level, was studied extensively, both by cast cladding and roll bonding of picture frame techniques.

2. The main difficulty found with cast cladding was the control of dross inclusions, which are responsible for localized puncturing of the face cladding, only 0,35 mm thick.

3. Roll bonding of picture frame sets of 1100 aluminum enclosing a pre sintered cermet of high density 50% minus 325 mesh U_3O_8 and 50% minus 100 mesh aluminum powder, assured the production of fully acceptable plates.

4. The experimental studies aimed the quantitative determination of the fabrication variables on the yield, reproducibility of the results and quality of the plates, as well as of the parameters required for the design of the dies and of the picture frame assembly. In those studies, the charges used were prepared in various proportions of natural isotope content U_3O_8 and aluminum powder.

5. The compaction of properly conditioned charges was studied and the obtained results indicate that for the mixtures in the range from 40 to 60% U_3O_8, the density varies with the pressure according to the equation (1) (pg. 6). The density of pellets produced with 50% aluminum and 50% of 20% enriched U_3O_8, to be used in the fabrication of the actual fuel plates for the reactor, agreed fairly well with the results of the experimental series.

6. The pre sintering of the pellets, done in a continuous electric furnace of local construction, under argon, assured freedom from blisters in the fabrication steps. The hot workability of the picture frame sets was found in the plates cold rolled to the final thickness (2,5 mm).

7. The structure of the plates was homogeneous and no major irregularities were found by metallographic examination of cut specimens. Cladding is regular and corrosion tests at 65°C in deionized water have not disclosed unexpected abnormalities.

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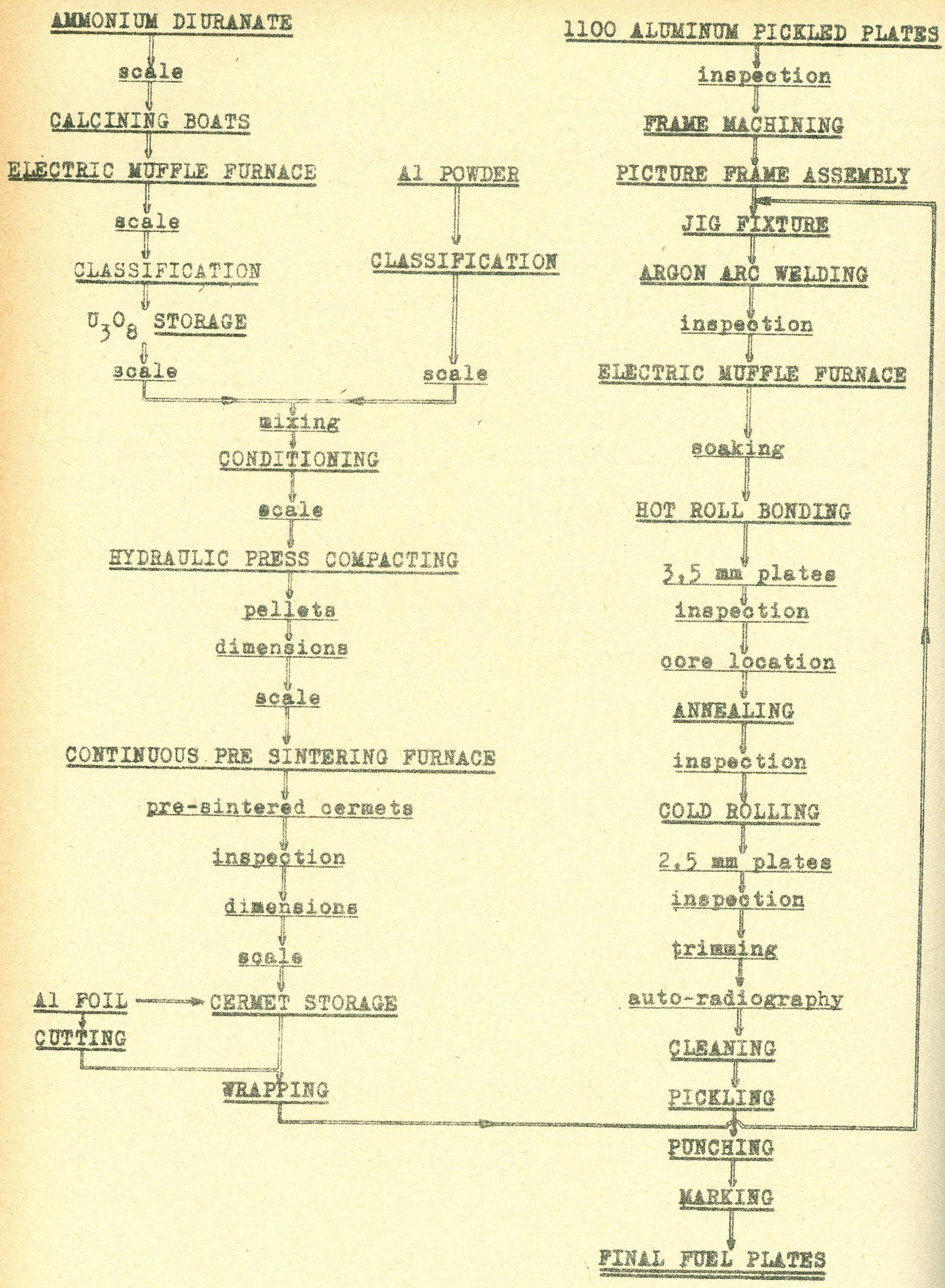


Fig. 1 - Flowsheet of the fabrication procedure.

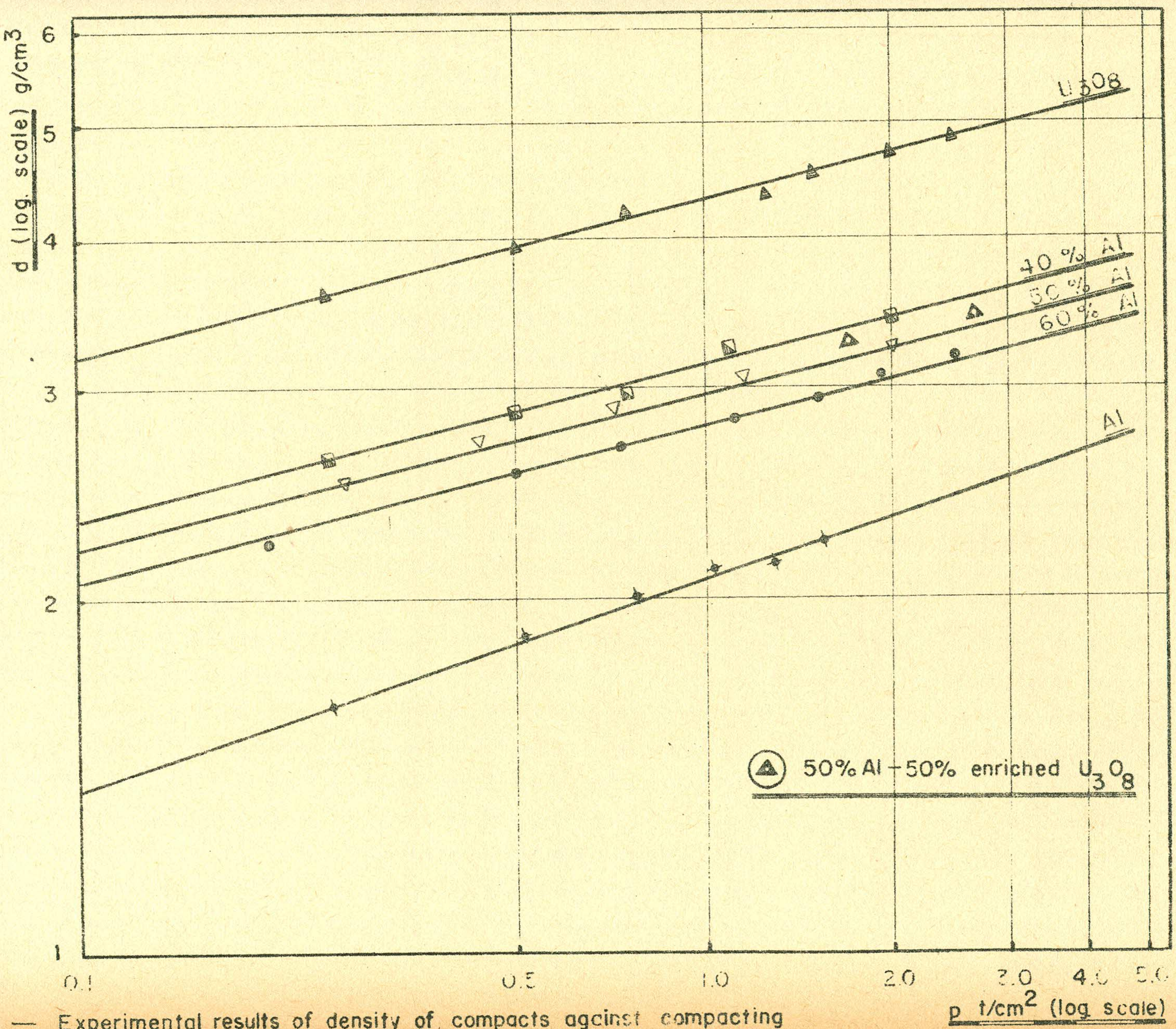


Fig. 2 — Experimental results of density of compacts against compacting pressures for Al and U_3O_8 (natural) mixtures.

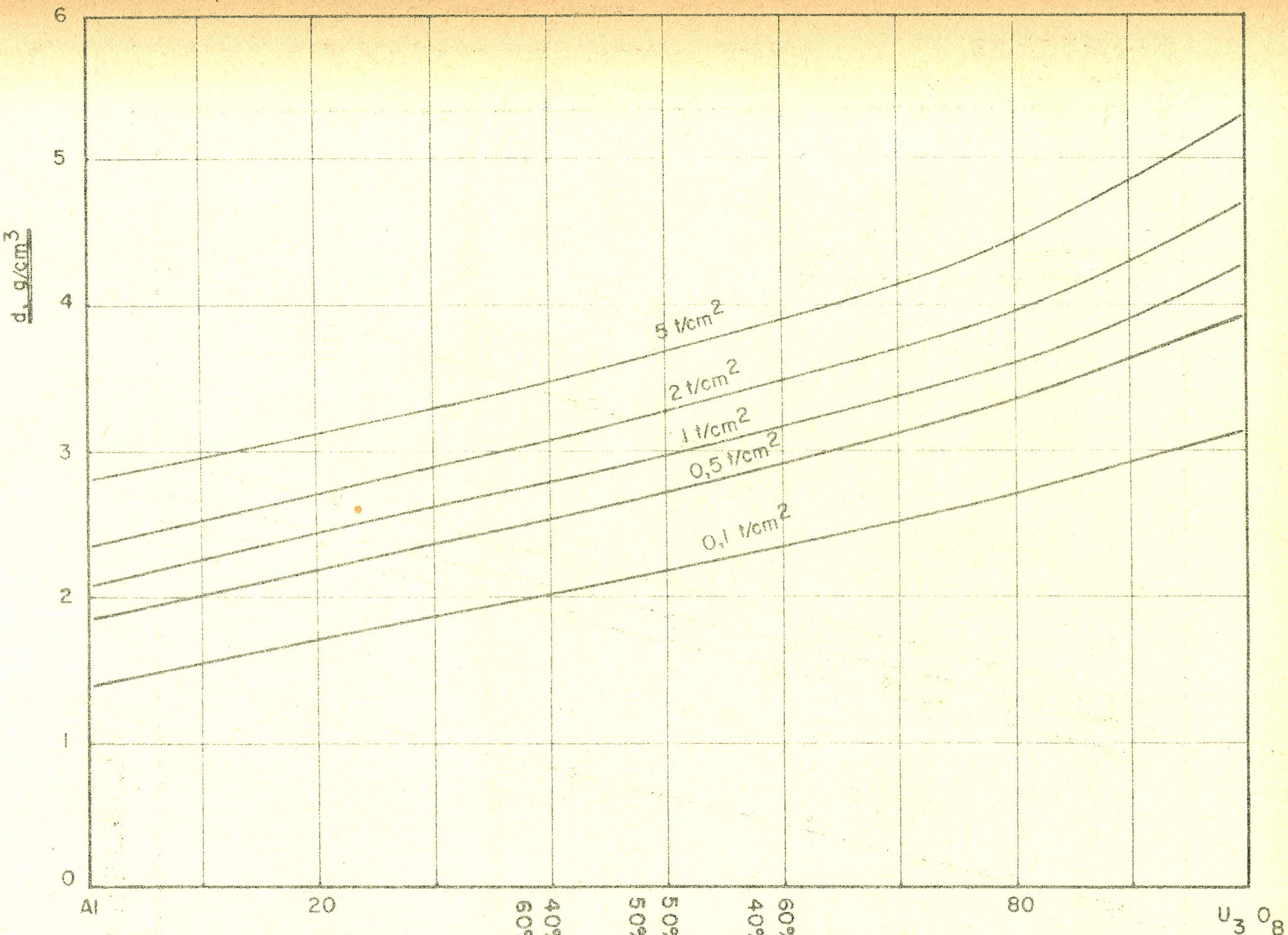
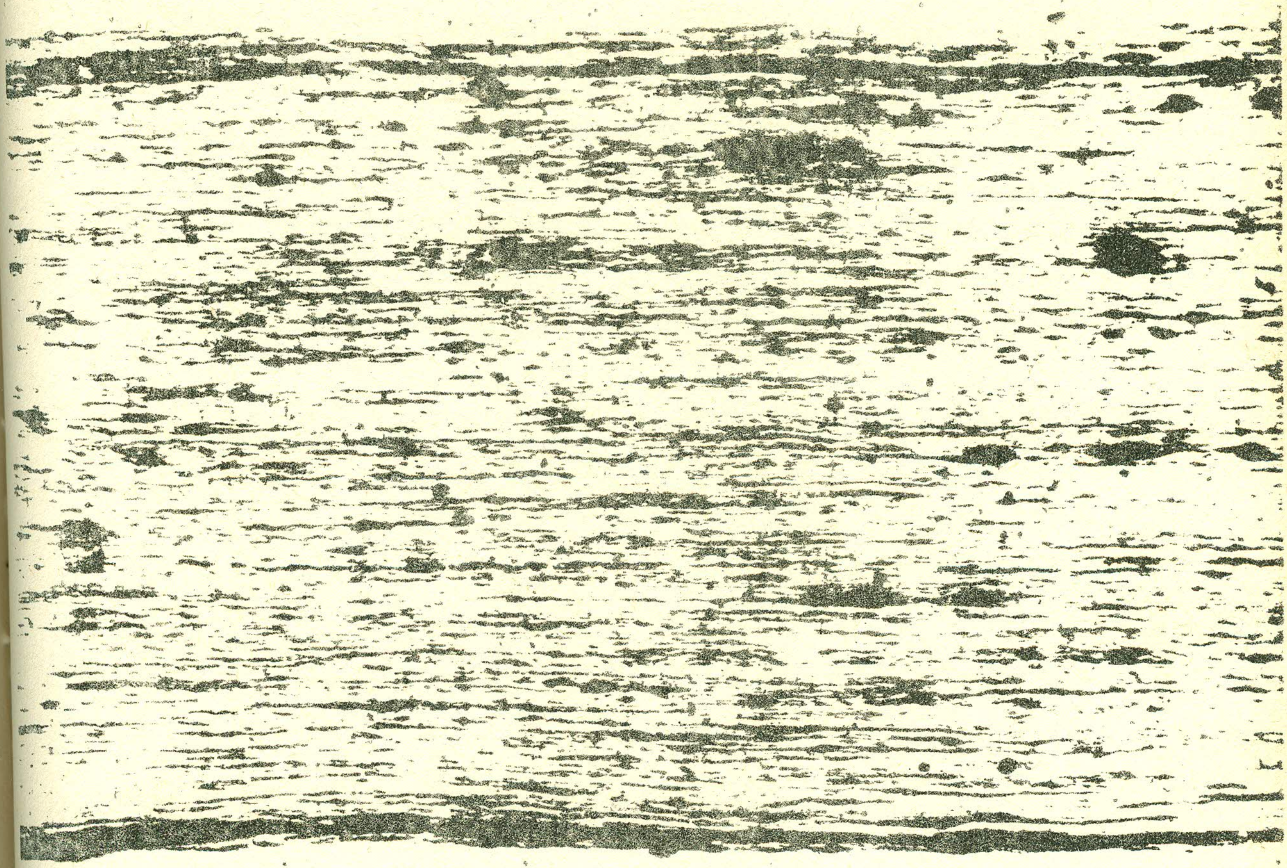


Fig. 3 — Variation of density of compacts with composition and compacting pressure (calculated values).



section of the core in a 2,5 mm roll bonded plate. As-polished. 55 x.