

# Experimental studies on the fabrication of thin fuel plates with $U_3O_8$ -Al cermets

By T. D. de Souza Santos, H. M. Haydt and C. T. de Freitas\*

In the course of the experimental studies carried out to establish the fabrication procedure for the fuel elements of the ARGONAUT reactor of the Instituto de Engenharia Nuclear, Rio de Janeiro, Brazil, it has been found necessary to investigate the effect of the main variables on the hot and cold workability of the totally clad fuel plates.

In order to properly evaluate the behaviour of the cermet, which during the rolling operations constitutes the "meat" of each fuel plate, it was found to be desirable to take both the hot and cold rolling operations far beyond the final specified thickness of the mock-up plates. These studies lead then to the actual production, for experimental purposes, of very thin, only 0.70 mm thick, fuel plates, which were totally clad with aluminium alloy. Such thin fuel plates are considered to have some interest for some advanced research reactors.

The paper describes the essential features of the development work done on the production of such thin fuel plates, containing a "meat" which is a plastically deformed cermet of a mixture of  $U_3O_8$  (natural  $^{235}U$  content) and aluminium. Plates of different widths and lengths were produced, including some which were widened to allow further shaping into corrugated sheets, giving a better geometry and higher fuel volumetric densities in the final fuel element assemblies.

## SUMMARY OF THE EXPERIMENTAL STUDIES

Two different procedures were investigated for the production of the thin clad fuel plates. Namely, roll bonding of picture frame sets with a cermet inside the frame component and cast cladding of the cermet with a suitable aluminium alloy into a specially designed ingot mould, following the successful idea developed by Bergua *et al.* [1] which was used for fuel plates in a materials testing reactor [2].

Considerable attention was paid to the cermet preparation, which has required the study of the following variables: (a) the production of  $U_3O_8$  from ammonium diuranate with properties suitable for dispersion; (b) the selection of the proper aluminium powder to be used in the charge; (c) the conditioning of the charge

mixture; (d) the pressing of the compacts into dies under hydraulic pressure; (e) sintering the compacts; (f) mounting the compacts for further processing.

The cermets obtained were to be used further, either in picture-frame sets for roll bonding and final rolling to the desired thickness, or for the production of a cored ingot by a cast cladding technique which was subsequently rolled into the finished thin plates.

The plates produced by both processes were evaluated by metallographic examination of cut specimens, by auto-radiography, by blister testing and by accelerated corrosion tests in de-ionized water at 70°C for 100 hours.

## CERMET PREPARATION

Figure 1 depicts the main steps of the cermet preparation as evolved from the experimental development work, carried out at the Institute's laboratories [3, 4]. The details of the most important operations are presented in the following sections.

The compacting behaviour of the charge mixtures made from various proportions of  $U_3O_8$  and aluminium powder has been investigated by varying: (a) the time, temperature and mass of the ammonium diuranate in the calcining operation; (b) the fineness of the produced  $U_3O_8$ ; (c) the fineness of the aluminium powder; (d) the conditioning operation, which was done in large rubber-lined jars with hard rubber balls; (e) the charge and die lubrication; (f) the die geometry and compacting pressure of the hydraulic presses.

The results have shown that strict control of the calcining variables was essential to obtain reproducibility of powder properties for the further fabrication steps. It was found that the best set of conditions was 875°C for two hours with 2 kg loads in the charging trays in the electric muffle furnace. Once cooled the charge was unloaded and classified with a 325 mesh sieve, the oversize fraction was retained for other purposes. Experience has shown that a lower calcining temperature and shorter times lead to an undesirable formation of blisters in the final annealing of the cold rolled plates. The aluminium powder used was imported from France and only the minus 100 mesh fraction was used. The aluminium powder had the following composition: 0.36% Fe, 0.07% Si, 0.12% Cu, 0.22% Zn and 0.098% Mg. Metallographic

\* Instituto de Energia Atômica, São Paulo.

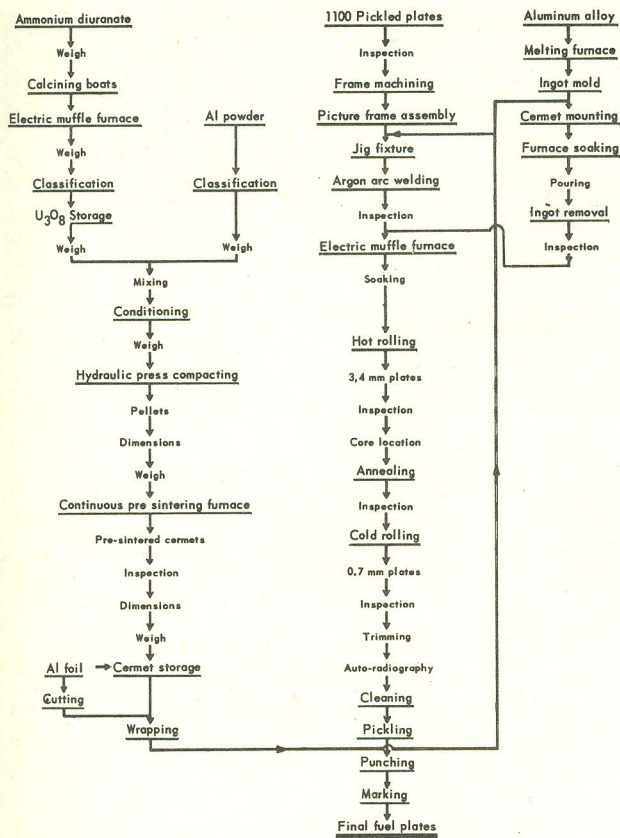


Figure 1. Flowsheet of the fabrication procedure

examination shows that coarser powder tends to produce an uneven distribution of the constituents.

The conditioning step is also of prime importance. The first tests were done in polyvinyl jars with hardened steel balls and gave erratic behaviour in the final rolling of the fuel plates, resulting in irregular bonding of the cermet with the cladding. This was particularly severe with the picture-frame technique. The best results were obtained by preparing the charge in a 3-l polyvinyl jar, tightly closed, with a constant load of hard rubber balls. Wilkinson [5] has shown the superiority of rubber balls over steel balls in the charge preparation of  $U_3O_8$  and aluminium.

The pressing step was done in dies fully lubricated with a solution of stearine in carbon tetrachloride.

The dies used allowed the production of compacts of variable heights (from 6 to 12 mm) and of rectangular cross sections,  $32 \times 64$  mm,  $65 \times 50$  mm or  $65 \times 100$  mm.

The influence of the compacting pressure on the apparent densities of the compacts has been determined for various proportions of minus 325 mesh  $U_3O_8$  and/or minus 100 mesh aluminium powder. The results are shown in Fig. 2. There was no measurable influence of the cross section of the die on the density for the same pressure and same charge composition, for the range of thicknesses indicated above.

The experimental results for the mixtures from 40%  $U_3O_8$ -60% Al to 60%  $U_3O_8$ -40% Al can be represented by the equation  $\log d = \log a + 0.141 \log p$ ,

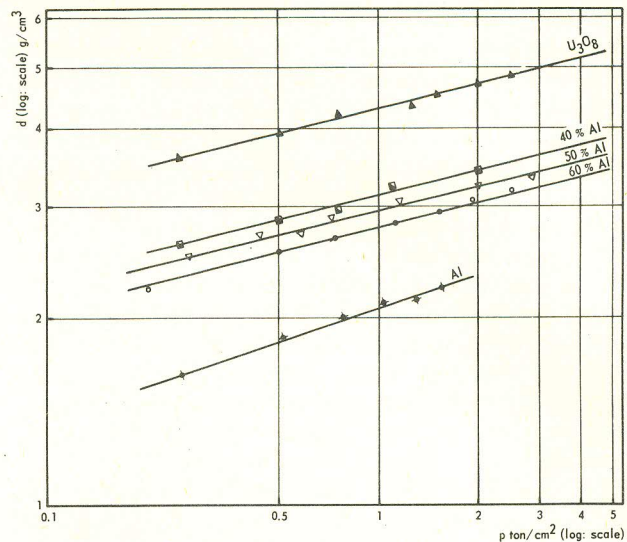


Figure 2. Variation of the density of compacts with composition and compacting pressure

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

It has been found that the presence of even small fissures in the compact impaired the plastic behaviour of the pre-sintered cermet during hot rolling. Due attention was therefore paid at the conditioning and pressing steps to prevent the cermet losing its inherent hot plasticity while still reaching the required density in the final rolled plate.

The last important step in the fabrication procedure was the pre-sintering of the compacts in an argon atmosphere at  $580^\circ C$  in a continuous multi-zone tubular electric furnace. This furnace was designed and built by a local manufacturer.

Slow heating to the sintering temperature and slow cooling to room temperature, at the rates of  $200^\circ C/h$  and  $150^\circ C/h$  respectively, have been provided to avoid cracks due to thermal gradient stresses. The operation was done in large rib-bottomed graphite boats of special design to avoid distortions. The pre-sintering did not alter substantially the cermet density, but does improve appreciably its high temperature plasticity during the hot rolling operations. It is also effective in driving off residual moisture and volatile constituents present in the conditioned charge.

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

#### PICTURE-FRAME CLADDING

The pre-sintered cermets were wrapped with dead annealed 0.10 mm thick 1100 aluminium foil before they were hand set into the frame component, machined from 1100 pickled aluminium plate, 11.5 mm thick. Special precautions were adopted to ensure complete freedom of surface defects both in the frame and in the two cover sheets, cut from 2.0 mm thick 1100

aluminium plates. The composition of the aluminium plates is: 0.37 % Fe, 0.092 % Zn, 0.034 % Si, 0.032 % Mn; traces of Mg and Cu were not found. The frame was machined carefully to assure a tight fit of the wrapped cermet in the frame opening. At first, residual air was evacuated from the inner space through a hole drilled at the trailing end of the frame. It was found later that this step was unnecessary.

A special jig was designed and built to assemble the frame with the cermet inside and the two cover plates in the proper position before welding under an argon arc with a tungsten tip. The welding was satisfactory and the metallographic examinations of the seam welds did not disclose cracks or clusters of entrapped oxide.

### CAST CLADDING

To avoid the erosion of the cermet by the impingement of the metal stream during pouring into the ingot mould, the cermet was wrapped with a 0.25 mm thick aluminium foil.

The alloyed cast iron ingot mould comprised three parts, an inner U-shaped part, which determined the dimensions of the ingot, and two outer plates. These parts were assembled together with C-clamps. To avoid sticking, all the surfaces which were to be in contact with the molten alloy were thoroughly coated with a thin suspension of calcined bone ash. The inner part had holes, through which positioning stainless steel screws could slide ensuring the proper position of the cermet during the pouring operation. Some auxiliary devices were provided to set the cermet at its precise location. Any off-centring of the cermet caused difficulties in the hot rolling operations and unevenness in the cladding thickness of the final plates.

After the cermet was set in the ingot mould, it was transferred to an electric oven furnace for soaking at 500 °C for 30 minutes.

Most of the experimental heats were produced with aluminium of the following composition: 0.23 % Fe, 0.024 % Si, 0.084 % Zn, 0.014 % Mn; traces of Mg and Cu were not found. Further work is under way with other aluminium alloys. The metal charge was melted in a graphite crucible in an electric furnace kept at 790–800 °C; this was found to be the most convenient temperature for the experimental set-up utilized. Pouring was done carefully to assure a fast and uniform rate of filling the ingot mould up to the top. After filling, more hot metal was poured into the shrinkage cavity to fill it properly and to avoid casting defects in the upper zone of the ingot.

Soon after the pouring was completed and the upper zone had solidified, the positioning screws were removed, the C-clamps unfastened and the ingot removed from the inner part. The holes left by the removal of the positioning screws were immediately welded electrically with aluminium of the same composition, in an argon arc. After cooling to room temperature, the ingots were inspected for residual minor surface and casting defects, which, if localized (small

cross inclusions or small punctures), were removed by light hand filing.

### HOT AND COLD ROLLING

It must be stressed that the actual welding by diffusion of the components of picture-frame assemblies is done by the hot roll bonding, which requires proper heating and soaking and an adequate rate of deformation. For the cored ingots obtained by cast cladding, the hot rolling is also a very important step, as it determines the properties for further mechanical processing.

Using the conventional picture frame cermet, the assembled set was 150 mm × 115 mm × 15.4 mm thick, with an enclosed cermet of 100 mm × 64 mm × 11.4 mm. The cast ingot, when the cermet was 64 mm × 32 mm × 12.5 mm, was 130 mm × 62 mm × 29.5 mm, with a taper of 4° along its thickness. Larger cast clad ingots were also used, with a cross section of 64 mm × 50 mm and of the same thickness, and still larger ones are to be used in further work.

Ingot and picture-frame assemblies were soaked in a muffle electric furnace at temperatures of 590–600 °C for 20 and 30 minutes respectively, before removal to the rolling mill. Intermediate anneals and blister tests after completion of the rolling schedule were also done in this temperature range, for 10 to 20 minutes, depending upon the thickness of the plate being processed.

The hot and cold rolling operations were carried out in a Stannat-Mann high precision rolling mill. It has been found experimentally that, in the breaking down phase, passes giving more than a 12 % reduction tended to warp the stock unduly and that much lighter reductions between passes impaired the roll bonding or the initial breaking down of the cast structure of the clad metal.

For picture-frame assemblies the treatment schedule was as follows: 59 % reduction by hot rolling, a furnace anneal for 20 minutes, 61 % reduction by hot rolling, a second furnace anneal, 61 % reduction by hot rolling, followed by cooling to room temperature for edge and end cutting. After a 10-minute furnace anneal, a 30 % reduction by cold rolling was followed by a 10-minute anneal and a final 72 % reduction by cold rolling to the required 0.7 mm thickness. This was followed by a blister test for 45 minutes at 590 °C. The thickness of the processed plate is 3.4 mm at the end of the hot rolling stage and the trimming is done when the plate reaches 2.5 mm. Final trimming gives the thin plate with the final dimensions of 0.5 mm side cladding and an end cladding of 5 to 8 mm.

The reduction schedule required for the cast clad ingots was heavier so as to break down the cast structure, cross rolling was used to achieve the final specified width of the "meat". After furnace soaking for 30 minutes, the ingot was hot rolled to a 66 % reduction, annealed for 20 minutes, hot rolled again to a 64 % reduction and reheated for 10 minutes, hot rolled once more to a 56 % reduction, annealed for 10 minutes and finally hot rolled down to 3.2 mm.

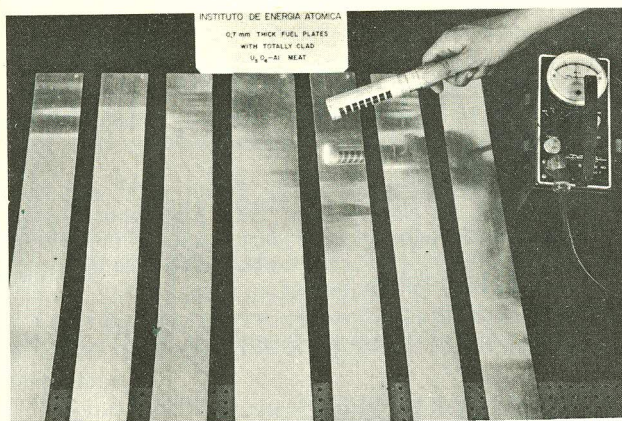


Figure 3. 0.7 mm thick fuel plates (ends of the plates)

The central wider plate was obtained by the picture-frame technique and has a 0.5 mm thick and 103 mm wide "meat", with 0.1 mm cladding. The others were obtained by cast cladding and have a 0.4 mm thick and 34 mm wide "meat", with 0.2 mm cladding

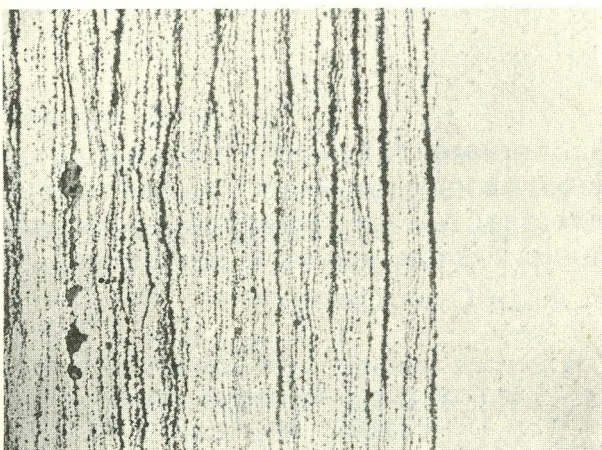


Figure 4. Micrograph of a longitudinal section of a 0.7 mm thick fuel plate with 54.36%  $U_3O_8$ -45.64% Al showing the dispersion of the "meat" and the bonding with cladding ( $\times 260$ )

After cooling to room temperature and edge and end trimming, it was annealed for 10 minutes, cold rolled down to 2.2 mm, once more annealed for 10 minutes and finally rolled down to the final 0.7 mm thickness. A final blister test annealing was then carried out for 45 minutes at 590°C. The final dimensions of the cladding material were substantially the same as that of the thin plates from the picture-frame process.

Figure 3 shows the extremities of some of the long thin plates produced.

Some of the wider plates obtained with cross rolling in the earlier stages of hot working were subsequently corrugated in a die in a hydraulic press, with waves 20 mm long and 6 to 8 mm deep.

#### EVALUATION OF THE PLATES

Many of the thin plates produced were cut and the mounted specimens were examined under a Leitz

MM5 metallographic microscope, after suitable polishing and etching, to disclose the "meat" structure and the bonding with the cladding material. At the beginning of the development work on the picture-frame technique, bonding defects caused considerable difficulty. These defects were removed when the individual operations were examined in detail.

Figure 4 shows one of the micrographs of a 0.7 mm cast clad plate, with a 0.15 mm aluminium alloy cladding and a final deformed "meat" of 0.40 mm thickness. The uniformity of the dispersion and the good bond achieved should be noted. Blister tests did not disclose unwanted defects.

Accelerated corrosion tests were performed on several end sections of the thin plates in de-ionized water at 70°C for 100 hours. The mean rate of mass increase was 0.036 mg/cm<sup>2</sup>/d. A more extensive programme of testing at higher temperatures will be carried out.

Auto-radiographic examinations are carried out as a routine inspection, both for determining the precise location of the "meat" and for checking the uniformity of the dispersion. The density of the "meat" at the edges is, as expected, slightly lower than the mean core value. No major irregularities were found at the ends of the "meat" in the plate, its contour line being rather sharp and not showing the "dog boning" effect.

#### CONCLUSIONS

The fabrication of 0.7 mm thin fuel plates completely clad at their ends was studied extensively, both by roll bonding of picture-frame sets and by cast cladding, using a cermet with 40 to 60%  $U_3O_8$  dispersed in aluminium powder.

The compaction of the properly conditioned charges was studied and the obtained results show that the densities varied with the compacting pressure according to Fig. 2.

The pre-sintering of the compacts, which was done under argon in a continuous electric furnace of local construction, assured freedom from blisters in the fabrication steps and improved the hot plasticity of the cermet during hot rolling.

The structure of the thin clad plates was homogeneous and no major irregularities were found by metallographic examination of cut specimens. The cladding is regular and the corrosion tests done at 70°C during 100 hours in de-ionized water have not disclosed unexpected abnormalities.

#### ACKNOWLEDGEMENT

The authors acknowledge the valuable assistance rendered by Mr. P. S. Carvalho Pereira da Silva, of the Instituto de Pesquisas Tecnológicas, Divisão de Metalurgia, on the metallographic examination of a large number of specimens cut from the experimental plates.

## REFERENCES

1. Bergua, H., Friddle, R., Diaz, J., and Baird, J., *Fabrication of the ISNSE Fuel Element for Low Power Research Reactors. Nuclear Fuel Elements*, Reinhold Publishing Co., 184-192, New York (1959).
2. Tytgat, D., *Etude pour le réacteur BR1 d'un prototype d'un élément combustible à uranium naturel avec liaison nickel et gainage aluminium*, Proceedings of the Symposium on Fuel Element Fabrication, May 10-13, 1960, Vienna, vol. 2, IAEA (1961).
3. Souza Santos, T. D., Haydt, H. M., and Freitas, C. T., *Development Studies for ARGONAUT Fuel Plates Fabrication*, Paper 37, Session III, Study Group Meeting on the Utilization of Research Reactors, São Paulo, 1963, IAEA (to be published).
4. Bidwell, R. M., Haydt, H. M., Freitas, C. T., and Souza Santos, T. D., *Nota preliminar sobre propriedades e estruturas de dispersões  $U_3O_8$ -Al* (paper presented to the XVIII Annual Congress of the Associação Brasileira de Metais, São Paulo) (1963) (in press).
5. Wilkinson, W. D., *Uranium Corrosion and Alloys*, vol. 2, 1037, *Uranium Metallurgy*, Interscience, New York (1962).

## ABSTRACT—RÉSUMÉ—АННОТАЦИЯ—RESUMEN

A/486 Brésil

### Etude expérimentale de la fabrication de plaques de combustible minces avec des cermets $U_3O_8$ -Al

par T. D. de Souza Santos *et al.*

Le mémoire décrit l'étude expérimentale de la fabrication de plaques de combustible, de 0,7 mm d'épaisseur, gainées d'un alliage d'aluminium et contenant un cœur en cermet de  $U_3O_8$ , en vue de leur utilisation pour les ensembles combustibles de réacteurs de recherche de type poussé. La fabrication de plaques de combustibles aussi minces, entièrement gainées, présente de nombreuses difficultés, que l'on a pu surmonter grâce à l'étude systématique des variables de fabrication.

Les plaques avaient une longueur de 75 à 110 mm, avec un gainage des faces de 0,15 à 0,20 mm d'épaisseur sur un cœur déformé plastiquement de 0,30 à 0,40 mm, qui contenait de 40 à 60 % de  $U_3O_8$  dispersé dans de l'aluminium.

Les recherches ont porté sur la liaison par laminage de cadres (*picture frame*) ainsi que sur le gainage par coulée. Les expériences relatives à la liaison par laminage de cadres concernaient l'assemblage des cermets après frittage sous argon, l'assemblage des composants du cadre, la soudure sous argon des ensembles assemblés; la liaison par laminage à chaud et le laminage à froid final destiné à donner aux plaques les dimensions et propriétés physiques voulues. Les expériences relatives au gainage par coulée concernaient la préparation des cermets après frittage sous argon, l'influence des variables de coulée des alliages les plus intéressants, le conditionnement et le traitement thermique des lingots contenant l'âme, le laminage à chaud et le laminage à froid final. Les plaques minces ainsi préparées ont été soumises à un examen par autoradiographie et par des procédés métallographiques, ainsi qu'à des essais de corrosion dans de l'eau chaude désionisée.

On a également étudié le façonnage de larges bandes minces en ondulés destinés à former des éléments de combustible de conception simple permettant d'obtenir des rapports  $U_3O_8/Al$  élevés.

Les résultats obtenus prouvent que les deux procédés peuvent être utilisés, le gainage par coulée convenant pour la production de plaques minces de très grande largeur.

A/486 Бразилия

### Экспериментальные исследования методов изготовления тонких топливных пластин из керметов на основе закиси-окиси урана и алюминия

Т. Д. де Суза Сантос *et al.*

В докладе описаны экспериментальные работы по изготовлению топливных пластин в оболочке из алюминиевого сплава толщиной 0,7 мм с сердечником из кермета на основе закиси-окиси урана. Такие пластины представляют интерес для изготовления топливных сборок усовершенствованных исследовательских реакторов. Производство таких очень тонких пластин, полностью покрытых оболочкой, представляет большие трудности, которые были в конце концов преодолены благодаря систематическому изучению производственных параметров.

Пластины имели ширину от 75 до 110 мм. Толщина оболочек в плоской части пластины составляет от 0,15 до 0,20 мм. Пластически деформированный сердечник имеет толщину от 0,30 до 0,40 мм. Сердечник содержит от 40 до 60%  $U_3O_8$ , диспергированной в алюминии.

Исследование включало изучение метода очеловки твэлов литьем и метода соединения оболочки с топливом путем прокатки рамки, наполненной топливом и закрытой с обеих сторон пластинами из алюминия.

В отношении метода соединения оболочки с топливом путем прокатки рамки изучалось приготовление кермета после спекания в среде аргона, сборка компонентов рамки, аргоно-дуговая сварка деталей сборки, соединение топлива с оболочкой горячей прокаткой и окончательная холодная прокатка с целью определе-

ния размерных и физических характеристик пластин. В отношении метода очехловки топливных пластин литьем изучалось приготовление кермета после спекания в среде аргона, влияние характеристик литья на получение наиболее подходящих сплавов, условия литья и термообработка слитков, горячая прокатка и окончательная холодная прокатка. Полученные таким образом тонкие пластины исследовались радиоавтографическим и металлографическим методами, а также подвергались испытаниям на коррозию в горячей деионизированной воде.

Проводились исследования по получению широких тонких полос рифленой формы для простой сборки тепловыделяющих элементов окиси-закуси урана к алюминию.

Полученные результаты показали, что могут быть приняты оба метода. Для изготовления очень широких пластин подходит метод очехловки твэлов литьем.

A/486 Brasil

### Estudios experimentales sobre fabricación de placas delgadas que contengan cermetes de $U_3O_8-Al$

por T. D. de Souza Santos *et al.*

La memoria describe los estudios experimentales realizados con miras a la fabricación de placas de 0,7 mm de espesor, con revestimiento de aleación de aluminio, y que contengan un cermet de  $U_3O_8$  y aluminio, destinadas a formar parte de los elementos combustibles de reactores de investigación de pro-

yecto avanzado. La producción de tales placas delgadas y totalmente revestidas presenta muchas dificultades, que fueron superadas finalmente después de estudiar con carácter sistemático las principales variables de fabricación.

Las placas debían tener entre 75 y 110 mm de ancho con revestimiento de 0,15 a 0,20 mm de espesor sobre el núcleo deformado plásticamente a partir del cermet de 0,30 a 0,40 mm de espesor. El núcleo debía estar constituido por mezclas de 40 a 60 % de  $U_3O_8$  con aluminio.

La investigación comprendió tanto el colingotaje de conjuntos de molduras como la soldadura por laminación. En el caso del colingotaje, el trabajo experimental incluyó la preparación del cermet, después de sinterizado bajo argón; el montaje de los componentes de los conjuntos de molduras; la soldadura del conjunto al arco eléctrico en atmósfera de argón; la soldadura por laminación en caliente y la laminación final en frío para satisfacer las especificaciones dimensionales y físicas de las placas. En el caso de la soldadura por laminación, el trabajo incluyó el montaje del cermet, después de sinterizado bajo argón; la influencia de las variables de fundición en las aleaciones más convenientes; el acondicionamiento y tratamiento térmico de los lingotes y operaciones de laminación en caliente, así como la laminación final en frío. Las chapas producidas fueron examinadas mediante autorradiografía, estudios metalográficos y se sometieron a ensayos de corrosión realizados en agua desonizada caliente.

Se efectuaron estudios sobre la conformación de las chapas delgadas en elementos corrugados destinados a su montaje en elementos combustibles de diseño simple que permitan una relación  $U_3O_8-Al$  favorable.

Los resultados obtenidos muestran que los dos procedimientos pueden ser adoptados y que el proceso de colingotaje es el más conveniente para la fabricación de chapas anchas y delgadas.