

# Developments in fuel fabrication for research reactors in Brazil

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

The Divisão de Metalurgia Nuclear of the Instituto de Energia Atômica is presently engaged in the actual fabrication of the fuel elements for the ARGONAUT reactor, Instituto de Engenharia Nuclear, Rio de Janeiro, and for the subcritical assembly known as "Re-Suco", Instituto de Energia Atômica, for lattice studies.

The fuel elements for the ARGONAUT reactor comprise some novel features which will be mentioned in a later section. The "Re-Suco" assembly utilizes rather large  $UO_2$  pellets, which presented some fabrication difficulties due to their size.

Extensive development work had to be carried out before the fabrication procedures were properly worked out. The chief aims were: (a) to determine the influence of the main variables on the properties and behaviour of the cermets and pellets; (b) to establish the fabrication procedure which would best fit the existing laboratory facilities; (c) to afford a reasonably cheap and straightforward fabricating procedure; (d) to supply the required data from which the metallurgical units could be designed and built under short notice by local manufacturers; (e) to provide on-the-job training for the operating staff.

Many difficulties had to be overcome in the earlier stages of the development work which was started early in 1962. These difficulties were related mainly to irregular and erratic behaviour in the roll bonding of picture-frame sets for the ARGONAUT plates and to cracks attributed to thermal stresses in the pre-sintering of the large  $UO_2$  pellets.

The paper describes the main features of the experimental work done on fabrication of fuel plates containing a cermet of 54.4 %  $U_3O_8$  of natural isotopic content and 45.6 % aluminium powder. Two complete mock-up fuel elements with a natural isotopic content were produced and the actual fuel elements with 20 % enriched  $U_3O_8$ -Al cermets of the same composition are being fabricated. The paper describes further the production of the large  $UO_2$  pellets for the subcritical assembly, and stresses the influence of most important variables on the properties of those pellets.

## ARGONAUT FUEL PLATE FABRICATION

As it was proposed to operate the Instituto de

Engenharia Nuclear ARGONAUT reactor at 10 KW (th), it was considered necessary to provide a totally clad plate instead of a strip obtained by extrusion, from which the actual plates could be cut.

Two fabricating alternatives were extensively studied, namely, roll bonding of picture-frame sets with a thick cermet inside; and cast cladding the cermet with a suitable aluminium alloy in a specially designed ingot mould, following the idea developed by Bergua *et al.* [1] at Argonne National Laboratory. Although some excellent plates were obtained, the rather strict specification for face cladding thickness did not encourage the use of the cast cladding alternative.

After a considerable amount of development work on the roll bonding of picture frame sets a safe and reliable fabricating procedure has been established which fully meets the dimensional, physical, structural and corrosion resistance requirements for the ARGONAUT fuel assemblies.

## Plate specifications

The final plate specifications were as follows: "meat" composition 54.4 %  $U_3O_8$  (20 % enriched) and 45.6 % Al powder;  $U_3O_8$  124.5 g per plate; total Al 216.4 g max. per plate; total weight 340.9 g per plate; final dimensions of the "meat" 585 to 600 mm long, 68 mm wide and 2.0 mm thick; final plate dimensions 610 mm long, 73 mm wide and 2.40 to 2.44 mm thick.

## Production of the compacts

The compacting behaviour of charge mixtures containing various proportions of natural isotopic content  $U_3O_8$  produced from ammonium diuranate has been thoroughly investigated by varying the calcining temperature and time, the grain sizes of the  $U_3O_8$  and the aluminium powder, the conditioning step, the charge and die lubrication and the compacting pressure [2].

The results have shown that strict control of the temperature and time of the calcining operation was essential to secure reproducible densities in the compacting operation, which was done in hydraulic presses in large hardened steel dies.

The conditioning step is also of prime importance, as it determines the ability of the cermet to withstand the hot and cold rolling operations during the plate fabrication. The best results are obtained by preparing

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the charge for 24 hours in a 3.5 l rubber lined jar, tightly closed, which contained a constant load of hard rubber balls.

Pressing is done in a 100 mm × 65 mm die at a compacting pressure of 0.54 ton/cm<sup>2</sup> to give a 11.4 mm thick cermet. The dies are fully lubricated with a solution of stearine in carbon tetrachloride. It has been found that the presence of even small fissures in the compact impair the plastic behaviour during the roll bonding operations. Therefore attention must be paid at the pressing step to avoid such cracks.

#### Pre-sintering of the compacts

An important step to ensure plasticity during the roll bonding operation is the pre-sintering of the cermet which is done in argon at 580°C, in a multi-zone electric tubular furnace (Fig. 1).

Slow heating to the sintering temperature and slow cooling to room temperature is important to avoid thermal stresses and related cracks. This operation is done in large rib-bottomed graphite boats of a 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 hot roll bonding operation.

#### Assembly of picture frame components and roll bonding

The pre-sintered cermets are wrapped in dead soft annealed 0.10 mm-thick 1100 aluminium foil before they are hand set into the frame component which is machined from 1100 11.5-mm thick aluminium alloy plate. Special precautions are adopted to ensure complete freedom from surface defects both in the frame and in the cover sheets which are cut from 2.0 mm-thick 1100 aluminium alloy plate. The frame is carefully machined to ensure a tight fit of the wrapped cermet in the opening.

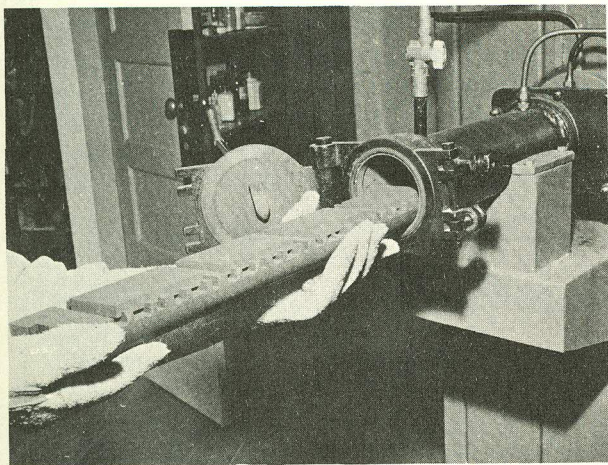


Figure 1. Charging the cermets into the continuous electric sintering furnace under an argon atmosphere

The cermets are 100 × 64 × 11.4 mm and contain 124.5 g U<sub>3</sub>O<sub>8</sub> (54.4%)

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 an argon arc with a tungsten tip. The welding is satisfactory and metallographic examination did not show any cracks or entrapped oxide.

Once welded, the assemblies are soaked for 45 minutes in a specially designed electric muffle furnace, at a temperature of 590°C. Strict control of the temperature, soaking time and location of the assembled set within the furnace, are necessary to avoid defects during the hot roll bonding operations.

The hot and cold rolling operations are done in a Stannat Mann four-high rolling mill. The roll bonding schedule is as follows: 59% reduction by hot rolling, furnace anneal for 20 minutes, 61% reduction by hot rolling, a second furnace anneal, 61% reduction by hot rolling and then cooling down to room temperature for edge and end cutting. The plates are X-rayed at this stage to check the proper location of the "meat" so as to ensure the proper final cutting of the plate once the cold rolling is completed.

#### Cold rolling, finishing and inspection of the final plates

After annealing for 10 minutes, the cut plates are finally cold rolled down to the 2.40 to 2.44 mm final thickness in the rolling mill. Before they are cut to the final dimensions through a die, the plates are subjected to X-ray and blister tests, which are done at 590°C for 45 minutes.

The final trimming and punching operations are done in proper jigs and fixtures to achieve the dimensional tolerances of the assembled fuel elements. Figure 2 shows two of the assembled fuel elements.

#### UO<sub>2</sub> PELLETS FOR THE "RE-SUCO" ASSEMBLY

The sub-critical assembly will contain 217 aluminium alloy tubes loaded with UO<sub>2</sub> pellets of natural

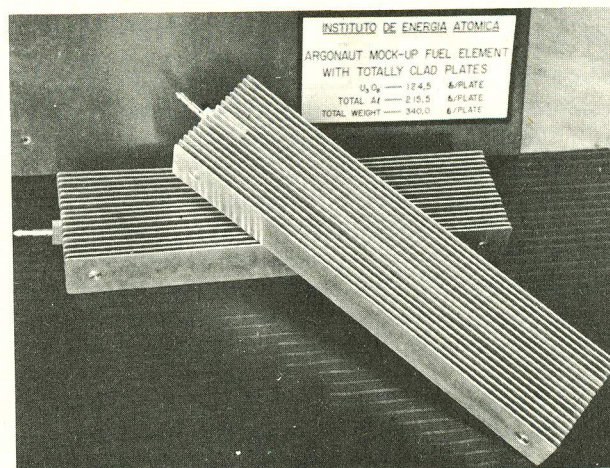


Figure 2. View of two assembled ARGONAUT fuel elements of 17 plates with a cermet "meat" of 54.4% U<sub>3</sub>O<sub>8</sub>-45.6% Al

isotopic content, of  $40.0 \pm 0.2$  mm diameter and  $39.8 \pm 0.3$  mm height. Mass requirements for the assembly called for a pellet density of  $6.6 \pm 0.1$  g/cm<sup>3</sup>. The total length of the aluminium tube is 1269 mm and the pellets are loaded into the 910 mm upper section which has a  $44.0 \pm 0.2$  mm outside diameter and a  $40.5 \pm 0.2$  mm inside diameter. Each tube will hold 22 pellets of 330.4 g each. The total weight of UO<sub>2</sub> will be 1812.4 kg and 5962 pellets will be required.

A fabricational procedure has been established which ensures good reproducibility of the properties of the powders and pellets and keeps the investment and fabricating costs within reasonable limits. Very extensive preliminary studies were necessary to determine the influence of the main variables and to establish the fabricational procedure [3,4].

### Calcining

Calcining the ammonium diuranate is done in an electric muffle furnace at a temperature of 875°C for 3 hours. The uranium salt is loaded into alloyed cast iron trays which hold 2.5 kg each. There is appreciable grain coarsening during the calcining.

### Reduction

The reduction is carried out under hydrogen in electric furnaces with two independently controlled reduction zones and a long water cooled chamber to allow proper cooling of the reduced powder before exposure to the atmosphere.

The charge is loaded into graphite boats of special design which are coupled together to form a train which is moved along the furnace at scheduled time intervals. The lower reduction zone is set at 300°C and the upper at 700°C, the boat remains about one hour in each zone. The entrance and discharge ends of the furnace can be isolated from the central part through the closing of special intermediate doors, the end spaces can then be purged with argon.

### Conditioning

In order to meet the density requirements with the minimum compacting pressure and with the minimum pre-sintering time, the charge should be conditioned with extreme precautions.

Conditioning is done in hard rubber lined steel containers of 5 l capacity running at 80 rpm over a large rack. Twelve containers can operate concurrently. The reduced UO<sub>2</sub> load in each container is 3 kg and as grinding medium 6 kg of hardened steel balls of 12 mm diameter are used. The total conditioning time is 48 hours.

For a powder obtained by reduction under fixed conditions, the powder density increases with the conditioning time tending asymptotically to a well defined value, which depends upon the speed of rotation, the UO<sub>2</sub> and the ball charge. The initial tap density of the reduced powder is 1.6 g/cm<sup>3</sup>; this increases with time to 4.5 g/cm<sup>3</sup> after 48 hours. The

container is emptied and the conditioned charge is passed through a 20 mesh screen to break up the powder lumps.

The conditioned powder is then hand mixed with 2% camphor dissolved in alcohol. The charge is spread in an enamelled tray and allowed to dry in an electric oven set at 50°C. As lubricant, 0.5% zinc stearate is added to the dried charge.

### Compacting

Low compacting pressures are used to minimize die wear and consequently pellet production costs. The charge per pellet is 337.1 g, weighed on a Mettler scale in a glove box. After removal of the binder and lubricant, the net weight of the pellet will be 330.4 g. The expected green density is 6.70 to 6.72 g/cm<sup>3</sup>.

The die is a double floating-plunger type made of air hardened alloy steel, which is set into a special fixture which does not allow height fluctuations of more than 0.5 mm. The compacting force, exerted through two hydraulic presses operating at the same time, can vary from 22 to 28 tons. It has been found that these fluctuations can be attributed to irregular driving off of the camphor binder. Under the prescribed operating conditions, the scatter of the densities obtained is within 0.1 g/cm<sup>3</sup>, which complies fully with the specification presented in the beginning of this section.

Figure 3 shows the influence of the compacting pressure on the green densities of 40.1 mm pellets produced from charge mixtures of increasing tap densities.

### Pre-sintering

The pre-sintering is done at a fast rate in a specially built tubular electric furnace, with eight independently-

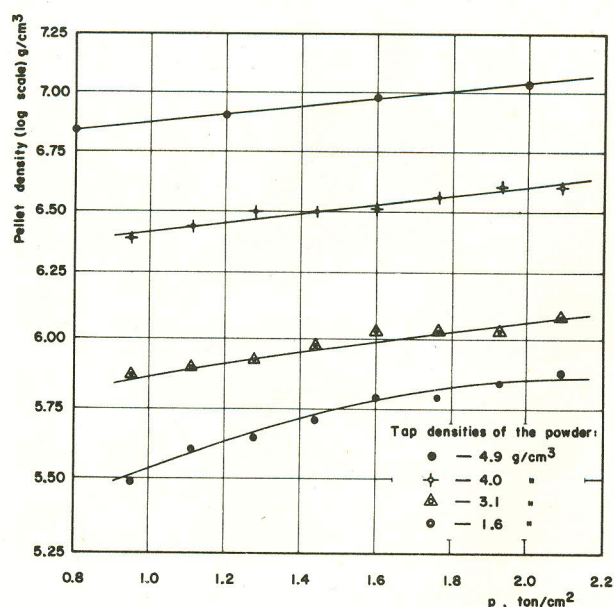


Figure 3. Influence of the compacting pressure on the densities of as-pressed pellets, for powders conditioned to variable tap densities

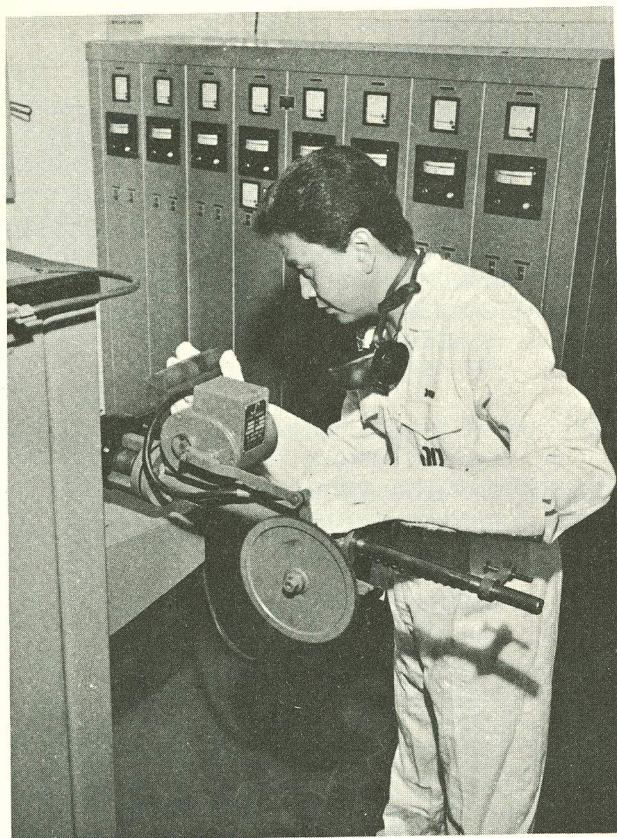


Figure 4. Charging the graphite boat with 330.4 g  $\text{UO}_2$  pellets into the continuous electric furnace for pre-sintering under argon

controlled hot zones, under an argon atmosphere. At the discharge end a long water cooled section allows proper cooling of the sintered charge in the graphite boats, thus avoiding skin oxidation of the pellets.

Figure 4 shows the charging entrance to the furnace and the adjustable-speed feeder which pushes the first graphite boat. The usual feed rate is 60 cm/h. The present maximum temperature is  $400^\circ\text{C}$  in the central part of the furnace, which is about 60 cm long. During this fairly fast pre-sintering the binder and lubricant are completely driven off.

The final pellet density is  $6.6 \pm 0.1 \text{ g/cm}^3$ . The diameter is  $40.05 \pm 0.05 \text{ mm}$  and the height is  $39.8 \pm 0.3 \text{ mm}$ . The reproducibility of these properties is very good and regular routine tests on the compaction of the conditioned charge are carried out.

Assembly of the pre-sintered pellets in the Al-alloy tubes

The pellets have a shiny appearance and are free

from cracks or folds. As they are insufficiently hard to withstand further loading into the aluminium alloy tubes which form the fuel assemblies, they are wrapped in 0.05-mm aluminium foil and the bundles are then lowered into the aluminium tubes. The final assembly is then argon arc welded in a rotary jig to ensure good uniformity of the welded seams.

## CONCLUSIONS

The procedure for the fabrication of fully clad fuel plates for ARGONAUT reactor containing a "meat" of 54.4%  $\text{U}_3\text{O}_8$  and 45.6% aluminium powder has been described.

The pressing of the conditioned  $\text{U}_3\text{O}_8$ -Al powder mixture and the pre-sintering of the cermet are done in equipment which has been designed and built locally. Good reproducibility of the fuel properties is obtained.

An extensive investigation has been conducted on the influence of the important variables on the properties of large (40.1 mm diameter and 39.8 mm height) and heavy (330.4 g)  $\text{UO}_2$  pellets for a sub-critical assembly.

The accurate control of the characteristics of the powder produced from nuclear grade ammonium diuranate, allows for a simple fabrication procedure based on pressing under low pressure with a floating-plunger die and fast pre-sintering under argon in a continuous eight zone furnace of local construction.

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## ABSTRACT—RÉSUMÉ—АННОТАЦИЯ—RESUMEN

A/485 Brésil

## Fabrication du combustible des réacteurs de recherche brésiliens

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

Les auteurs décrivent la mise au point de la fabrication des éléments combustibles du réacteur ARGONAUT de l'Instituto de Engenharia Nuclear, Rio de Janeiro, et du Re-Suco, un assemblage sous-critique utilisant des pastilles de  $UO_2$  de 40,1 mm, de l'Instituto de Energia Atômica.

Dans les deux cas, on a choisi le procédé de fabrication de façon à pouvoir utiliser un équipement susceptible d'être construit rapidement par l'industrie locale.

Avant d'entreprendre la fabrication des plaques combustibles du réacteur, on a fait de nombreuses études expérimentales avec des cermets contenant 54,4 % de  $U_3O_8$  à teneur isotopique naturelle et 45,6 % de poudre d'aluminium. Avec  $U_3O_8$  en poudre ayant la même taille de grain que l'oxyde enrichi à 20 %, on a pu définir correctement les variables essentielles du procédé de fabrication. Après avoir étudié le procédé de gainage par coulée, on a mis au point le procédé de liaison par laminage des cadres contenant le cermet, préalablement fritté sous argon. On a réalisé deux éléments combustibles fictifs à uranium naturel pour contrôler la reproductibilité des résultats avant d'entreprendre la fabrication des plaques combustibles réelles.

Les cadres sont conçus de façon à assurer un gainage complet, y compris les extrémités des plaques, pour l'âme mince. Chaque élément combustible comprend 17 plaques de 2,42 mm d'épaisseur, 73,0 mm de largeur et 610 mm de longueur, avec un gainage sur les faces de 0,30 mm d'épaisseur. Chaque plaque contient 21,0 g de  $^{235}U$ . Le travail expérimental a essentiellement compris: la préparation de la charge; le compactage sous différentes pressions dans des moules de géométries différentes; le frittage sous argon; l'assemblage des composants du cadre; le laminage à chaud des ensembles soudés, et les opérations de laminage à froid pour amener les plaques aux spécifications dimensionnelles et physiques.

L'assemblage sous-critique Re-suco contient 271 éléments combustibles, 1100 tubes en alliage d'aluminium de 44 mm de diamètre extérieur, 40,5 mm de diamètre intérieur, 1269 mm de longueur totale, chacun contenant 22 pastilles de 40,1 mm de diamètre et pesant 330,4 g. La charge totale nécessite 5962 pastilles pesant 1812,4 kg.

La fabrication des pastilles est partie de diuranate d'ammonium de pureté nucléaire, produit par l'IEA. Le procédé de fabrication a été étudié de façon à minimiser la coût de l'opération. Grâce au contrôle précis des opérations de calcination, de réduction et de

conditionnement, on a obtenu une charge facile à compacter, permettant la production de pastilles à la densité voulue de  $6,6 \pm 0,1$  g/cm<sup>3</sup> avec une pression de compactage de 1,9 t/cm<sup>2</sup> seulement; ces pastilles sont ensuite frittées sous argon dans un four de frittage continu à huit zones, à chauffage électrique. La reproductibilité des dimensions et de la densité des pastilles frittées a été excellente.

A/485 Бразилия

## Разработки в области изготовления топлива для исследовательских реакторов в Бразилии

Т. Д. де Суза Сантос *et al.*

Авторы описывают экспериментальные исследования и разработки по изготовлению тепло-выделяющих элементов для реактора ARGONAUT Института ядерной техники в Рио-де-Жанейро и подкритической сборки RE-SUCO, содержащей таблетки из двуокиси урана диаметром 40,1 мм, Института атомной энергии.

В обеих разработках выбор метода изготовления тепловыделяющих элементов определялся с учетом использования оборудования, которое могло быть изготовлено за короткий срок местными предприятиями.

Перед тем как начать серийное производство топливных пластин для реактора ARGONAUT, были проведены обширные экспериментальные исследования на керметах, содержащих 54,4%  $U_3O_8$  (из природного урана) и 45,6 алюминиевого порошка. Используя порошок  $U_3O_8$  с тем же размером зерен, что и размер зерен окиси с 20-процентным обогащением (по  $U^{235}$ ), можно было правильно определить наиболее важные особенности метода изготовления. После завершения исследования процесса очехловки твэлов литьем производилось изучение метода соединения кермета с оболочкой путем прокатки рамочных сборок, наполненных керметом, который предварительно спекался в среде аргона.

Два полных подобных тепловыделяющих элемента из природного урана были изготовлены для проверки воспроизводимости результатов, перед тем как начать серийное производство топливных пластин.

Рамочные сборки были сконструированы с таким расчетом, чтобы обеспечить полную очехловку, включая концы топливных пластин с окончательно деформированным тонким сердечником. Каждый тепловыделяющий элемент состоит из 17 пластин толщиной 2,42 мм, шириной 73,0 мм и длиной 610 мм. Толщина обо-

лочки в плоской части пластины составляет 0,30 мм. Каждая пластина содержит 21,0 г  $U^{235}$ . Экспериментальная работа включала следующие основные операции: изготовление смеси окиси урана и порошка алюминия; прессовка этой смеси под различными давлениями в штампах различной формы; предварительное спекание спрессованной смеси порошков в среде аргона; сборку компонентов прессуемой рамки; горячую прокатку собранных и заваренных рамочных сборок и окончательную холодную прокатку для доведения топливных пластин до нужных размеров и физических спецификаций.

Подкритическая сборка RE-SUCO содержит 271 тепловыделяющих элементов, 1100 труб из алюминиевого сплава наружным диаметром 44 мм, внутренним диаметром 40,5 мм и общей длиной 1269 мм. Каждый тепловыделяющий элемент содержит 22 таблетки диаметром 40,1 мм и весом 330,4 г. Общая загрузка составляет 5962 таблеток и весит 1812,4 кг.

Диуранат аммония ядерной чистоты, приготовленный Институтом атомной энергии, был исходным материалом для изготовления таблеток. Процесс изготовления этих таблеток был тщательно исследован, чтобы свести к минимуму эксплуатационные расходы. Благодаря точному контролю за операциями прокаливания, восстановления и доведения материала до нужных характеристик была достигнута высокая степень прессовки порошка, которая обеспечивает производство таблеток требуемой плотности  $6,6 \pm 0,1$  г/см<sup>3</sup> при сжимающем давлении только 1,9 т/см<sup>2</sup>. Эти таблетки далее быстро спекаются в среде аргона в восьмизонной непрерывно действующей печи с электрическим нагревом. Воспроизводимость размеров и плотности спеченных таблеток была найдена очень хорошей.

A/485 Brasil

### Progresos realizados en la fabricación de elementos combustibles para reactores de investigaciones en el Brasil

por T. D. de Souza Santos et al.

Los autores describen los estudios experimentales de desarrollo realizados para la fabricación de los elementos combustibles del reactor ARGONAUTA del Instituto de Engenharia Nuclear, Río de Janeiro, y para el Re-Suco, un conjunto subcrítico dotado de pastillas de  $UO_2$  de 40,1 mm, para estudios de reticulados en el Instituto de Energia Atômica.

En ambos desarrollos, la selección del proceso de fabricación fue examinada críticamente a fin de utilizar el equipo que podía ser construido por fabricantes locales.

Se realizaron estudios experimentales con cermets con 54,4 % de  $U_3O_8$  natural y 45,6 % de polvo de aluminio. Las variables principales del proceso de fabricación pudieron ser determinadas utilizando polvo de  $U_3O_8$  natural con la misma granulometría del polvo enriquecido a 20 %, antes que fuese iniciada la producción de las chapas para el reactor. El procedimiento evolucionó hasta el proceso de soldadura por laminación de los conjuntos de moldura conteniendo el cermet, previamente sinterizado bajo argón. Se fabricaron dos modelos completos de elementos combustibles con uranio natural para comprobar la fidelidad de los resultados antes de comenzar a producir las placas de combustible.

Los conjuntos de moldura fueron proyectados a fin de garantizar revestimiento continuo y perfecto del núcleo delgado deformado, inclusive las extremidades de las chapas. Cada elemento combustible comprende 17 placas de 610 mm de longitud, de 2,42 mm de espesor por 73,0 mm de ancho y con un revestimiento en las caras de 0,30 mm de espesor. Cada chapa contiene 21,0 g de  $^{235}U$ . El trabajo experimental consistió principalmente de: preparación de la carga; compactación a presión y geometría de matriz variables; presinterización de los compactados bajo argón; montaje de los componentes; laminación en caliente de los conjuntos soldados y las operaciones finales de laminación en frío para obtener las chapas dentro de las especificaciones.

El conjunto subcrítico Re-Suco contiene 271 elementos en tubos de aleación 1100, con 44,0 mm de diámetro externo, 40,5 mm de diámetro interno, longitud total de 1269 mm, cada uno de ellos con 22 pastillas de 40,1 mm de diámetro y 330,4 g de masa. La carga total necesita de 5962 pastillas que pesan 1812,4 kg.

El material de partida para la producción de las pastillas fue diuranato de amonio de pureza nuclear producido en el IEA. El proceso de fabricación fue estudiado completamente, con miras a reducir al mínimo los costos de fabricación. A través del control preciso de las etapas de calcinación, de reducción y de acondicionamiento, se consiguió obtener mezcla de gran compactibilidad, a partir de la cual pudieron ser producidas las pastillas con las densidades deseadas de  $6,6 \pm 0,1$  g/cm<sup>3</sup>, a presiones de compactación de solamente 1,9 t/cm<sup>2</sup>; esas pastillas fueron seguidamente sinterizadas bajo argón en un horno eléctrico continuo dotado de 8 zonas de calentamiento, a alta velocidad. Fue excelente la reproductibilidad de dimensiones y la densidad de las pastillas sinterizadas.