~~CONFIDENTIAL~~
UNCLASSIFIED

SCNC-289

METALLURGY AND CERAMICS (M-3679, 23rd Ed.)

This document contains Confidential-Restricted Data relating to civilian applications of atomic energy

LOW COST CERAMIC FUEL ELEMENTS BY ISOSTATIC PRESSING

By
J. Fugardi
J. L. Zambrow

July 1959

Sylvania-Corning Nuclear Corporation
Bayside, New York

AEC Research and Development Report

~~RESTRICTED DATA~~

This document contains restricted data as defined by the Atomic Energy Act of 1954. Its transmission or disclosure of its contents in any manner to an unauthorized person is prohibited.

This document is
PUBLICLY RELEASABLE

Angela Kaiser
Authorizing Official
Date: *8/8/59*

UNITED STATES ATOMIC ENERGY COMMISSION
Technical Information Service

~~CONFIDENTIAL~~
UNCLASSIFIED

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, expressed or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

This report has been reproduced directly from the best available copy.

Printed in USA. Charge \$0.45. Available from the Technical Information Service Extension, P. O. Box 1001, Oak Ridge, Tennessee. Please direct to the same address inquiries covering the procurement of classified AEC reports.

DO NOT PHOTOSTAT

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

~~CONFIDENTIAL~~

-3-

SCNC-289

LOW COST CERAMIC FUEL ELEMENTS
BY ISOSTATIC PRESSING

by

J. Fugardi
J. L. Zambrow

Contract: ^{□1} AT-30, GEN-366
Division of
Reactor Development

Issued: July, 1959

Classification cancelled (or changed to UNCLASSIFIED)
Letter from Declassification Branch, dated
by authority of November 16, 1959
by *[Signature]* TIE, date 12-11-59

This document is
PUBLICLY RELEASABLE
[Signature]
Authorizing Official
Date: 9/8/68

SYLVANIA-CORNING NUCLEAR CORPORATION
Bayside, Long Island, New York

~~CONFIDENTIAL~~

342 001

~~CONFIDENTIAL~~

-4-

TABLE OF CONTENTS

	<u>Page No.</u>
ABSTRACT.....	5
I. SUMMARY.....	6
II. INTRODUCTION.....	6
III. DISCUSSION.....	7
A. Sintered Pellet Assemblies.....	7
B. Green Compacted Pellet Assemblies.....	15
C. Nonpelletized Assemblies.....	20
IV. CONCLUSIONS.....	23
V. RECOMMENDATIONS FOR FUTURE WORK.....	24

~~CONFIDENTIAL~~

342 002

~~CONFIDENTIAL~~

-5-

ABSTRACT

Sample fuel elements consisting of a ceramic core in stainless steel cladding were prepared by tamping powder into the cladding and then hot isostatically pressing the assembly. High-density cores and an excellent mechanical bond between the core and the cladding were obtained.

342 003

~~CONFIDENTIAL~~

~~CONFIDENTIAL~~

-6-

I. SUMMARY

This investigation was undertaken for the Reactor Development Branch of the U. S. Atomic Energy Commission. The work was divided into three phases, involving hot isostatic (H.I.) pressing of the following: (1) sintered pellets, (2) cold compacted green pellets, and (3) tamped powder. The third phase was not anticipated in the original schedule but was determined by the results obtained in the second phase. The results of the work on sintered pellets were not completely successful as compression ridges formed between pellets and any surface defects in the pellets were visible in the clad surface. In addition, careful inspection of the pellets was necessary as any chips in the pellet edges could cause punctures in the cladding. An excellent mechanical bond, however, was obtained between the cladding and the pellet. Although reduction of applied pressure during H.I. pressing reduced the number of minor surface imperfections pleating occurred. The results of the second phase (green compacts) were unsatisfactory. In ejecting the high-density pellets from the die, the edges crumbled and, after H.I. pressing, depressions in the cladding could be seen at the pellet edges. The best results were obtained from H.I. pressing of tamped powder. In this final phase of the program, high-density cored samples with an excellent mechanical bond between the core and the cladding were obtained.

II. INTRODUCTION

The objective of this program, which was undertaken for the Reactor Development Branch of the AEC, was the fabrication of low-cost, high-quality, ceramic, rod-type fuel elements through the application of hot isostatic pressing techniques.

The fuel element is one of the major cost factors in the production of nuclear power. In reactors utilizing UO_2 fuel compositions, the production of tubes containing UO_2 pellets by present methods is an expensive operation.

~~CONFIDENTIAL~~

342 004

A process for fabricating low-cost, high-quality fuel elements would aid greatly in reducing the cost of producing power from nuclear energy. The fabrication of fuel elements by a hot isostatic pressing technique may accomplish this purpose.

The fabrication of fuel elements by isostatic pressing would effect cost savings by eliminating high-temperature sintering of green compacted UO_2 pellets, or grinding of sintered pellets. Another economy would be effected by the use of stainless steel tubing with commercial tolerances instead of premium-priced, close-tolerance tubing. With present methods of fabrication, tube-size tolerances are so narrow that commercial tubing cannot be used. Consequently, a premium must be paid for close-tolerance tubing. Use of isostatic pressing for the fabrication of ceramic rods may permit the use of commercial-tolerance tubing, thereby reducing the cost of the tubing.

Prior hot pressing work at Sylcor consisted in: (a) bonding aluminum cladding to nickel-plated uranium, and (b) cladding matrix dispersions of aluminum, stainless steel or niobium in UO_2 with the same metal used as the dispersant, e.g., a matrix of aluminum dispersed in UO_2 , clad with aluminum. The work was performed in a high-pressure vessel operating at temperatures of $1300^{\circ}C$ and pressures of 12,000 psi of argon. Other installations which are engaged in hot isostatic pressing programs are Battelle Memorial Institute and the Bettis Plant operated by Westinghouse. These installations are working on development programs for the Pressurized Water Reactor Project.

III. DISCUSSION

A. Sintered Pellet Assemblies

Sintered pellet assemblies made by inserting pellets with a diametrical tolerance of $\pm 0.002"$ into commercial-tolerance tubing, followed by vacuum welding

and H.I. pressing, should be less costly than pellet assemblies made by present methods.

Pellets were ordered with the following specifications: minimum density, 94% of theoretical value; diameter, $0.402" \pm 0.002"$; and height, $0.400" \pm 0.015"$. These wide tolerances were specified since pellets could be made by cold compacting and sintering powders without grinding. It was anticipated that, since unground pellets could be used, a significant cost savings might be realized. The expected savings, however, did not materialize because production techniques had brought the cost of grinding pellets down to about one dollar per pound. The use of ground pellets may be objectionable as the edges chip during grinding. An additional pellet specification will be required to compensate for surface irregularities in the cladding which follow the contour of the pellet surface, after H.I. pressing.

Type 304 stainless steel tubing was ordered with commercial tolerances. The size was selected so that there would be a minimum clearance of $0.002-0.003"$ between the largest pellet and the smallest tube. This clearance would allow loading of pellets into tubes under all conditions. Since the dimensions of the tubing received was on the large size, the minimum clearance between the pellets and tubing was $0.006"$, with an average clearance of $0.008"$.

Pelletized assemblies about 6 to 8 inches long were prepared by placing the pellets in a tube having one end cap welded in place. The second end cap was welded shut by vacuum induction, after which the pelletized assembly was helium leak-checked.

The first group of pelletized assemblies was H.I. pressed under conditions of temperature and pressure close to the maximum attainable by the equipment, i.e., at 1200°C and 12,000 psi of argon for one hour. The purpose of this study was to determine whether or not the pellets densified under H.I. pressing: measurable densification took

place. Upon removing the cladding from the H.I. pressed assemblies for density measurements of the pellets, it was apparent that some "bonding" had taken place between the pellet and the cladding (see Fig. 1). A high degree of mechanical interlocking was obtained and the interface between the cladding and the pellet was free from voids. Ultrasonic testing of the H.I. pressed assemblies, using a Sperry Reflectoscope at 2.5 mc, resulted in a high degree of transmission or good-to-excellent bonding.

The purpose of the planned presizing studies was to eliminate pleating by reducing the clearance between cladding and pellets. Pleating is a typical surface defect associated with hydrostatic pressing. It is caused by folding over of excess material of the external sheath during the pressing operation. Since no pleating occurred after H.I. pressing of pelletized assemblies at 12,000 psi and 1200°C, the planned presizing program was cancelled. On the basis of one presizing sample which was made, however, it was evident that the eccentricity of the pellets after H.I. pressing was less than that of samples made without presizing.

Final sizing of H.I. pressed, sintered pellet assemblies was carried out to improve the cladding surface. The most prominent surface irregularity of H.I. pressed pellet assemblies is the compression ridge, which is due to excess cladding material and is formed between pellets as the isostatic pressure forces the pellets closer together. Other possible surface defects are depressions or punctures caused by pellets with chipped edges. The final sizing operations consisted of swaging, drawing, and machining. While all swaged, drawn, and machined samples had uniform outside diameters, a variation in thickness of the cladding was noted. Further investigation revealed that the thickness variation was about the same as the taper in the as-received sintered pellets. Figure 2 shows an as-pressed sintered pellet assembly with several surface defects. Figure 3 shows a swaged assembly, and Figure 4, a drawn assembly. In both

Plate No. 21963

750X

Fig. 1. Photomicrograph of the interface of H.I. pressed stainless steel to UO_2 , showing the excellent mechanical bond. Pressing conditions: 12,000 psi, 1200°C , one hour.

Plate No. 10526

1X

Fig. 2. Sintered UO_2 pellets in stainless steel, H.I. pressed at 12,000 psi, 1200°C , for one hour.

342 008

Plate No. 10608

1X

Fig. 3. H.I. pressed UO_2 pellet assembly
after swaging.

Plate No. 10530

2X

Fig. 4. H.I. pressed UO_2 pellet assembly
after drawing.

Figures 3 and 4 the bright areas on the assemblies indicate areas of greater mechanical work. Upon sectioning the swaged and drawn samples, the pellets were severely laminated. Figures 5 and 6 show the sectioned samples with coarse and fine laminations.

To overcome surface irregularities in H.I. pressed assemblies, i.e., punctures or indentations due to chips on the edges of the pellets or indentations due to surface imperfections, lower pressures were used. At a temperature of 1000°C, a pellet assembly was subjected to 250, 500, and 1000 psi. The cladding thickness was 0.015" and the inside diameter of the cladding was 0.410". Since the outside diameter of the largest pellet was 0.404", there was a minimum clearance of 0.006".

The following results were obtained at various pressures:

- 250 psi: The cladding was poorly bonded (as determined by ultrasonics); there was a slight tendency to pleat, but no warping, and only slight evidence of compression ridges.
- 500 psi: Slightly better bonding was noted; pleating was more pronounced but not sharply defined; there was no warping, and the compression ridges were more pronounced.
- 1000 psi: The bonding was slightly improved but poor when compared to samples H.I. pressed at 12,000 psi and 1200°C; the pleating flattened out and the assembly warped about 0.020". Figure 7 shows the pellet assembly pressed at 1000°C and 1000 psi.

Another sample was pressed at 750°C but the pleat failed to flatten at pressures up to 12,000 psi. Compression ridges formed between the pellets. Ultrasonic testing of this sample showed good mechanical contact between the cladding and the pellets.

Plate No. 10545

5X

Fig. 5. Swaged sample showing coarse laminations.

Plate No. 10544

5X

Fig. 6. Drawn sample showing fine laminations.

Plate No. 10609

1X

Fig. 7. Sintered UO_2 pellets in stainless steel, H.I. pressed at 1000 psi, 1000°C , for 15 minutes.

Since the object of these experiments was to improve the surface of the cladding by reducing the pressure, it was demonstrated that, at about 1000°C, the pressure required to achieve contact between the cladding and the pellets was 1000 psi. A slight surface improvement in the cladding was evident in samples pressed at 1000 psi as compared with those pressed at 12,000 psi (compare Figs. 2 and 7). It is not recommended that the temperature be reduced below 1000°C because of the high pressure required to produce an acceptable finish. An important observation made as a result of this work was the fact that pleating occurs at some time during isostatic pressing of 0.015" thick cladding with a clearance of 0.006" between the cladding and a pellet having an outside diameter of 0.400". This pleat can be flattened out if the conditions of temperature and pressure are sufficient.

Thinner walled tubing (0.010" wall thickness) was H.I. pressed. The clearance between the cladding and the pellet was 0.006"; permanent pleating occurred. In order to use 0.010" wall tubing for the H.I. pressing of UO₂ pellet assemblies, ground pellets and special-tolerance tubing would, therefore, be required.

B. Green Compacted Pellet Assemblies

The use of green pellets in place of sintered pellets would effect further cost savings in fabricating rod-type ceramic fuel elements. The expenses of sintering and grinding pellets would be eliminated.

Initial work indicated that green pellets must have a high density to prevent pleating of the cladding. When ceramic-grade UO₂ was used, pellets which cold pressed to 46% of theoretical density, densified to 91% of theoretical density, after H.I. pressing. The large volumetric change which occurs during H.I. pressing caused pleating. A photograph of H.I. pressed ceramic-grade UO₂ pellets is shown in Figure 8. A study was started to eliminate pleating by making green pellets with high green densities. According to the literature,

Plate No. 10528

1X

Fig. 8. H.I. pressed, low-density "green" pellets.

342 014

a blend of 75 w/o coarse and 25 w/o fine ceramic powder particles packs to the highest green density¹ on the basis of particle size distribution.

Table I summarizes the data obtained with various types of powder. Using all fused powder, blend 1, a high-density pellet was obtained which had poor strength, as the edge of the pellet crumbled when it was ejected from the cold-compacting die. Using all ceramic-grade powder, a low-density pellet was obtained (46% of theoretical density) with good green strength. Swaging-grade UO_2 packed to a high green density with good strength. Blends 2, 3, and 4 were made from swaging-grade UO_2 . All swaging-grade UO_2 proved unsatisfactory, however, because, after H.I. pressing, the highest density obtained was 84% of the theoretical value. An attempt was made to produce a green pellet having both high density and good strength. Blend 5 was prepared from 75 w/o coarse fused-grade powder and 25 w/o fine ceramic-grade powder. Since the bulk of the mixture consisted of fused-grade powder, a high-density pellet would result from this combination. The ceramic-grade powder was expected to act as a binder for the larger fused particles, thus reducing crumbling of the pellet edges. Pellets made from blend 5, however, showed crumbling of the edges as the pellet was ejected from the die. A photograph of an assembly which contained green pellets made from blend 5 and H.I. pressed at 12,000 psi, 1200°C for one hour, is shown in Figure 9. The loss of material at the edges of the pellets resulted in depressions in the cladding. Since cold working of the cladding results in laminations, and machining of the cladding causes variations in the wall thickness of the final fuel element, green pellet work was discontinued.

¹ A.E.C. Westman and H. R. Hugiel, "The Packing of Particles," J. Am. Ceramic Soc., 13, 767 (1930).

TABLE I

DATA ON COLD-COMPACTED UO₂ POWDER^a

Blend No.	UO ₂ powder			Green density, gm/cc	Percent of theoretical density
	w/o	Mesh size	Grade		
1	75	-100 +140	Fused	9.2	84
	25	-325	Fused		
2	As-received	As-received	Swaging	8.50	77.5
3	75	-60 +200	Swaging	8.72	79.5
	25	-325	Swaging		
4	75	-60 +100	Swaging	8.60	78.5
	25	-325	Swaging		
5	75	-100 +140	Fused	7.93	72.3
	25	-200 +325	Ceramic		

^a Compacted at 40 tsi.

Plate No. 10527

2X

Fig. 9. H.I. pressed, high-density "green" pellets.

C. Nonpelletized Assemblies

Since the main objection to green pellet assemblies consisted in crumbling of the pellet edges during ejection from the die, it was decided to tamp the powder directly into the cladding. Using blend 5 and tamping the powder directly into the cladding at 40 tsi, in three-gram powder increments, densities of 95-98% of the theoretical value were obtained after H.I. pressing. A photomicrograph of such a sample is shown in Figure 10. The high-density, fused UO_2 particles can be seen in the matrix of slightly porous, ceramic-grade UO_2 . An attempt was made to tamp the swaging-grade UO_2 directly into the cladding. After H.I. pressing the swaging-grade powder, densities of 84% of the theoretical value were obtained. Photomicrographs were taken of both the swaging-grade UO_2 powder and the H.I. pressed, swaging-grade UO_2 to determine the cause of the low density. Figure 11 shows the as-received, swaging-grade UO_2 ; the internal porosity of the powder particles can be seen. Figure 12 shows the swaging-grade UO_2 after H.I. pressing. The internal porosity of the swaging-grade particles does not appear to change.

Although the H.I. pressing of blend 5 resulted in an excellent sample as to density, ultrasonic testing, and "bonding" of the UO_2 to the stainless steel cladding, some tamping lines were visible on the cladding surface. A tamped assembly which has been H.I. pressed is shown in Figure 13. Since tamping eliminates the objections to the use of green pellets, namely, breaking of the edges, an all-fused UO_2 tamped assembly would probably give excellent results. Thinner clad tubing can probably be used as there is little chance of pleating a tamped UO_2 assembly during H.I. pressing.

Plate No. 22413

100X

Fig. 10. Blend of fused- and ceramic-grades of UO_2 powder after H.I. pressing.

Plate No. 22411

100X

Fig. 11. Swaging-grade UO_2 powder in the as-received condition.

Fig. 13. H.I. pressed sample made by tamping a blend of 75% fused-grade, 25% ceramic-grade, UO_2 powder directly into the cladding.

2X

Plate No. 10610

Fig. 12. H.I. pressed sample made from swelling-grade UO_2 powder, 75% (-20 +60), 25% -325.

100X

Plate No. 22412

IV. CONCLUSIONS

The results of the first phase of the work, on sintered pellets using high pressures were not satisfactory, due to the formation of compression ridges between the pellets. Efforts to reduce these ridges by cold working laminated the UO_2 pellets. Any surface defects in the pellets were visible in the cladding. In addition, the pellets had to be carefully inspected as any chips in the pellet edges could cause punctures in the cladding. An excellent mechanical bond was achieved which was demonstrated by both ultrasonic testing and peel tests. Reduction of the pressure during H.I. pressing reduced the number of minor surface imperfections, but increased the tendency to pleat.

The results of the second phase on green compacts were unsatisfactory. In ejecting the high-density pellets from the compacting die, the edges crumbled. After H.I. pressing, depressions in the cladding formed at the pellet edges.

The best results were obtained from the H.I. pressing of tamped powder. In this final phase of the program, high-density, cored samples with an excellent mechanical bond between cladding and core were prepared.

The following advantages are secured in H.I. pressing a tamped assembly, as compared to the present method of making pellet-type assemblies:

1. Handling of green compacts is eliminated.
2. Sintering of pellets is eliminated.
3. Grinding of pellets is eliminated.
4. Close-tolerance tubing is eliminated.
5. Inspection of pellets is eliminated.

UNCLASSIFIED

~~CONFIDENTIAL~~

-24-

6. An excellent mechanical bond between core and cladding is obtained.

The fabrication of ceramic fuel elements at lower costs is made possible by the use of H.I. pressed, tamped assemblies. Since the cost of pelletizing UO_2 is about \$6.00 to \$8.00 per pound, and grinding costs add another \$1.00 per pound, the use of a machine capable of effectively tamping the powder into the cladding may reduce the cost of pellet assemblies. The length of a fuel element made by this method may, however, be restricted.

The authors wish to express their appreciation to the AEC for sponsoring this work, to Mr. M. Downey who prepared the samples and carried out the H.I. pressing operations, and to Mr. H. W. Woods for metallographic work.

V. RECOMMENDATIONS FOR FUTURE WORK

High-density UO_2 fueled cores with good mechanical bonding may require the incorporation of intentional voids within the fuel element for the accumulation of fission gas, especially for high burnup of 20,000 MWD/T. An investigation of commercial powders is recommended to aid in the fabrication of a H.I. pressed, tamped UO_2 assembly having uniform microporosity such that fission gas can be stored during high burnups without damage to the cladding. This investigation would include a metallurgical study to determine the effects of H.I. pressing on the grain size and hardness of the cladding. In addition, autoclave testing would be performed to determine the integrity of the bond.

~~CONFIDENTIAL~~

UNCLASSIFIED

342 022