Paper 33

IRRADIATION BEHAVIOR OF ThO_2-UO_2 FUELS

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ABSTRACT

ThO_2-UO_2 fuels have been studied because of certain advantages possessed by this type of fuel over UO_2. Pellets of ThO_2-UO_2 have been found to be highly stable dimensionally under irradiation. Various experimental assemblies which have been irradiated have included aluminum, Zircaloy, and stainless steel cladding, with the annulus surrounding the pellet filled with air, helium, NaK, or lead. A full-scale core of pellets lead-bonded to aluminum tube-plates has been used successfully in the Borax-IV reactor. Preliminary defect tests have shown that solid fission products are retained and that only the gaseous products, Xe^{138} and Kr^{88}, are detectable in the reactor coolant system.

INTRODUCTION

Most of the ceramic fuel development at Argonne National Laboratory has been concerned with thorium oxide in which has been dissolved various amounts of enriched uranium oxide. Our interest in this type of fuel came about because of two advantages possessed by thorium oxide-base fuels. First, thorium oxide-uranium oxide bodies can be sintered in air, whereas uranium oxide bodies must be sintered in vacuum or a protective atmosphere. Second, the use of thorium atoms in the fuel presents the possibility of constructing a thermal breeder based on conversion of natural thorium to the fissionable isotope U^{233}.

This paper will discuss results which have been obtained in experimental irradiations of ThO_2-UO_2 fuel specimens, and will conclude with a brief description of reactor operating experience on these fuels.

RESULTS ON EXPERIMENTAL SPECIMENS

The densities of the ThO_2-UO_2 specimens we have studied have been in the range of 85 to 90% of theoretical. Our first irradiations were made on specimens with the composition ThO_2-2-1/2 wt % UO_2.[1] The specimens were in the form of pellets, ~ 1/4 in. in dia by 3/4 in. long. The specimens were irradiated in NaK capsules. Some of the specimens were irradiated bare and others were clad in Zircaloy-2 or in stainless steel, with the bond between the fuel and cladding consisting either of NaK or air. Figure 1 shows bare specimens after irradiation. It can be seen that the appearance of the pellets was not affected by the irradiation they received. Dimensional changes in the pellets, if any, were less than 0.0002 in. The clad pellets also showed no changes resulting from irradiation.

DISCLAIMER

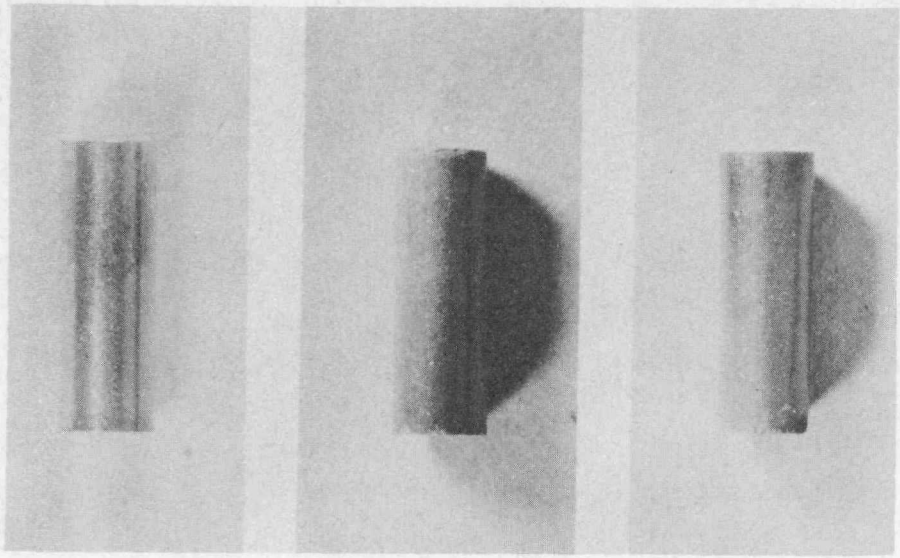


Fig. 1—Effect of irradiation ThO_2–2½ wt. % UO_2 ceramic specimens.

The next specimens that were irradiated in NaK capsules were made from the composition ThO_2-10 wt % UO_2.[1] These were in jackets of aluminum-1 wt % nickel alloy. One-half of the specimens contained a mixture of helium and argon in the annulus between the pellets and the cladding, and the other half contained

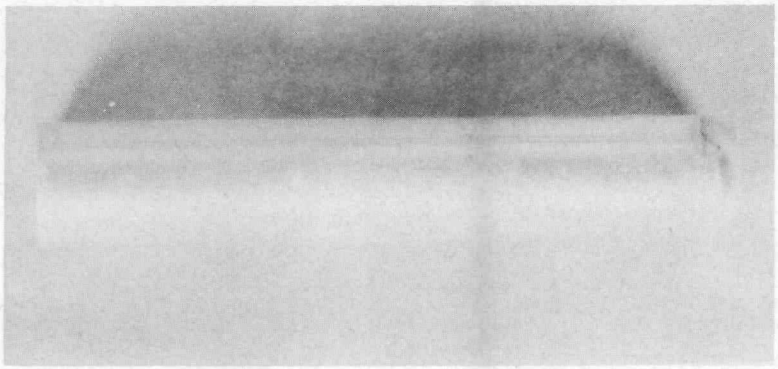
Total Metal Atom Burn-up, %	0	0.20	0.88
Irrad. Temp. °C	-	115	450

lead in the annulus. Figure 2 shows typical specimens of each type after irradiation and illustrates that the cladding failed on the gas-bonded specimen, whereas no damage was found in an identically irradiated lead-bonded assembly.

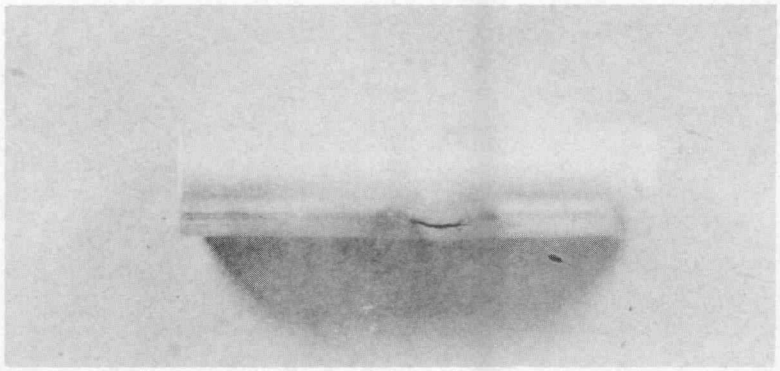
Figure 3 shows the same two specimens after partial removal of the cladding. Here it may be noted that the lead has held the fuel fragments in place in the lead-bonded specimens, whereas in the gas-bonded assembly the pieces have moved about sufficiently to damage the jacket and finally cause local failure.

Figure 4 shows a transverse section of a gas-bonded assembly which operated at a high enough temperature to melt the aluminum alloy cladding. Also a central void can be noted in the ceramic pellet. This is believed to have resulted from sintering or possibly melting of the oxide.

In order to irradiate experimental specimens at high heat fluxes without overheating the aluminum cladding, it was decided to directly water-cool small assemblies in the MTR process water. Figure 5 shows the specimen shape which was selected. It consists of a plate 6 in. long containing two tubes of pellets lead-bonded to the aluminum alloy cladding. In order to satisfactorily weld the end of the assembly, after the lead had been added, it was necessary to leave a short space free of lead, as shown to the left in the horizontal section. Figure 6 shows a cross section of the specimen and illustrates the lead bond. Figure 7 shows the specimen in an aluminum holder which can be fitted into an X-basket at the MTR. With this arrangement, the plates are cooled by a 1/16-in. annulus of water flowing at about 25 ft/sec.

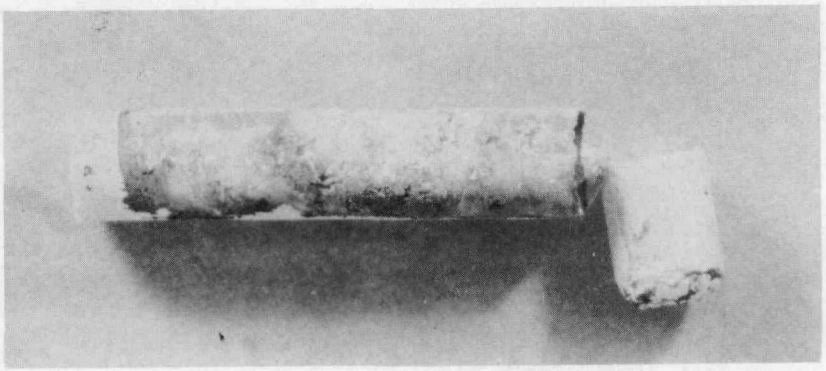


Lead-Bonded

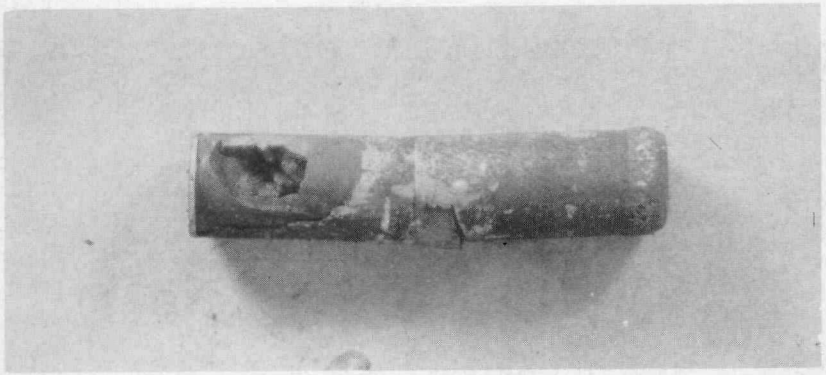


Gas-Bonded

Fig. 2—ThO_2—10 wt. % UO_2 irradiation specimens in Al—1 wt. % Ni alloy cladding after 0.64 per cent metal atom burn-up at 425,000 Btu/hr/sq ft.



Lead-Bonded



Gas-Bonded

Fig. 3—ThO_2—10 wt. % UO_2 specimens irradiated to 0.64 per cent metal atom burn-up at 425,000 Btu/hr/sq ft after partial removal of aluminum alloy cladding.

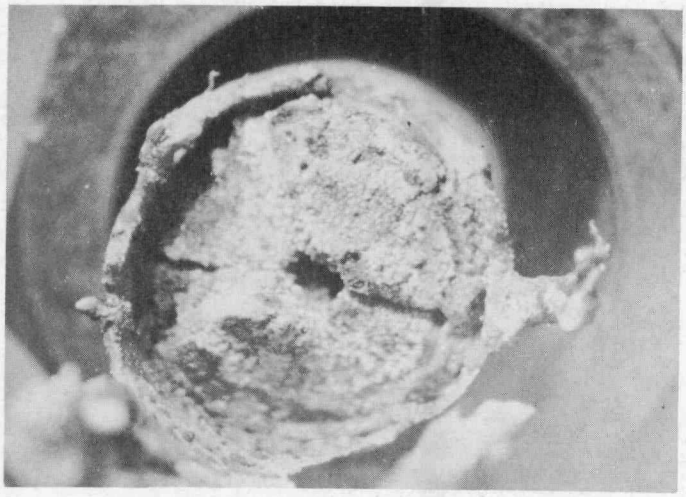
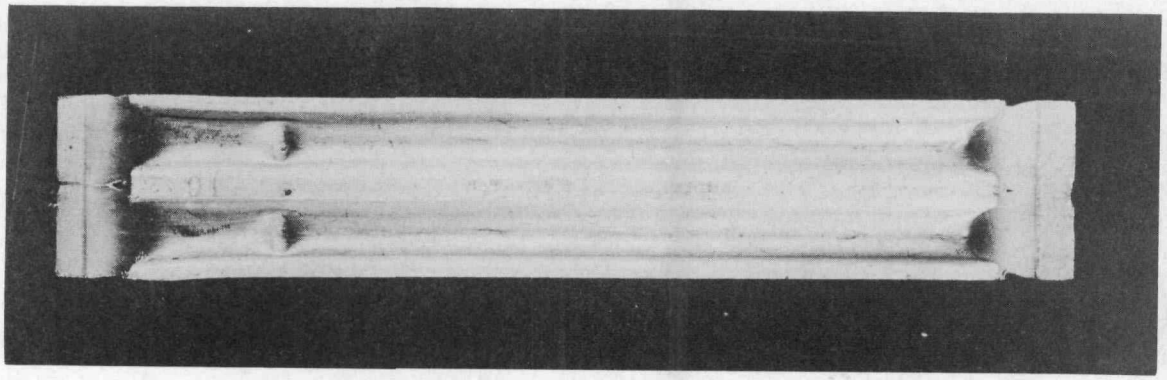
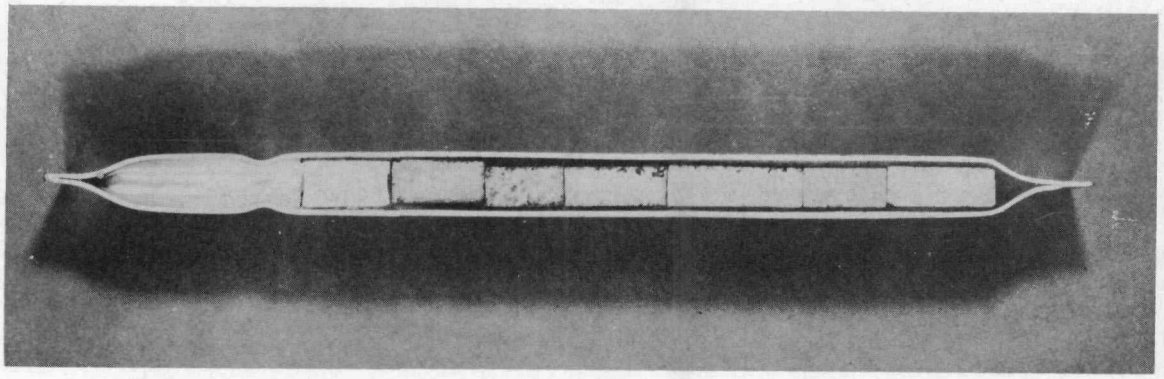


Fig. 4—Evidence of central melting or sintering in ThO_2–10 wt. % UO_2 specimen in gas-bonded Al–1 wt. % Ni alloy cladding after 1.25 per cent metal atom burn-up at 840,000 Btu/hr/sq ft.



Top View



Horizontal Section

Fig. 5—Lead-bonded ThO_2–6.36 wt. % UO_2 irradiation specimens.

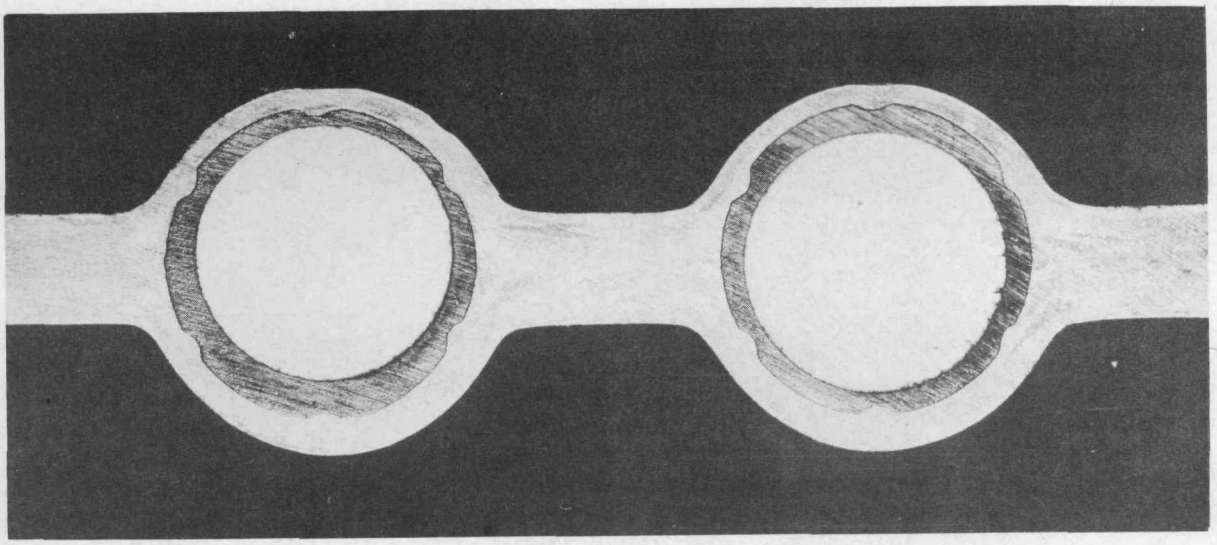


Fig. 6—Cross section of ThO_2–6.36 wt. % UO_2 irradiation specimen.

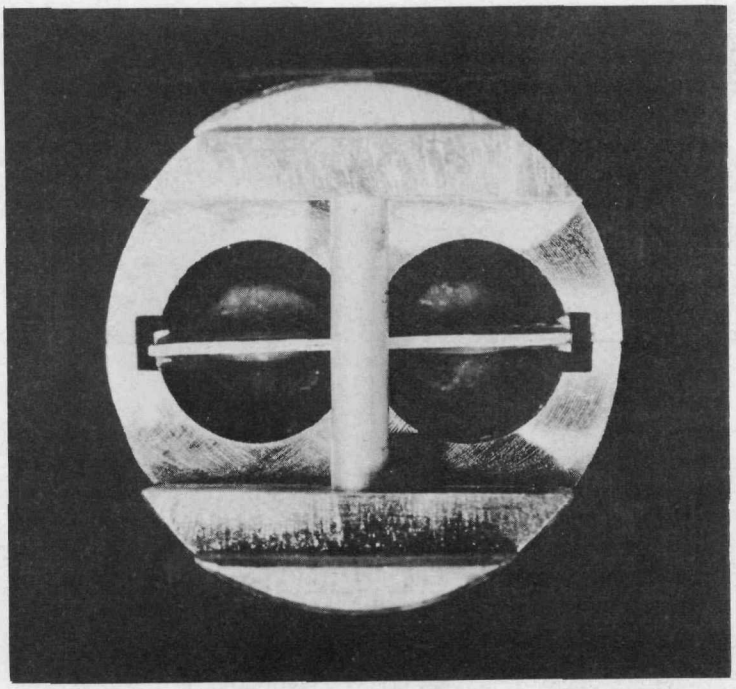


Fig. 7—Lead-bonded ThO_2–6.36 wt. % UO_2 specimen in irradiation holder.

To date we have examined only one of these specimens after irradiation. This particular plate received a burn-up of 1.0% of the metal atoms, or 7500 Mwd/T of oxide. The heat flux from the aluminum was 190,000 BTU/hr-ft^2. The only change which developed during irradiation was a 5.5% increase in dia of the aluminum tubes at the end of the specimen where the flux was highest. Other specimens which just arrived from the MTR and which are awaiting examination in our hot labs have received higher exposures under more severe conditions. Burn-ups in these specimens are estimated to have reached about 4.3% metal atom burn-up, or 32,000 Mwd/T of oxide. The maximum heat flux from the aluminum cladding in these specimens was on the order of 700,000 BTU/hr-ft^2.

REACTOR FUEL RESULTS

The excellent performance of ThO_2-UO_2 fuel in irradiation tests led to its use as fuel in the Borax-IV reactor. The composition selected was ThO_2-6.36 wt % UO_2. Tube plates of the type shown in Fig. 8 were extruded from aluminum-1 wt % nickel alloy and the oxide pellets were loaded into each tube with a lead bond.[2,3] The plates were spot welded into a box-type subassembly shown in Fig. 9. The subassembly is ~ 4 in.^{2} and 32 in. long. Originally, 59 subassemblies were used to make up the core of the reactor. The reactor first achieved criticality in December 1956, and for more than a year was operated with no difficulties. During this period of time the reactor was operated up to 14.5 Mw with a peak heat flux near 300,000 BTU/hr-ft^2.

The reactor was shut down for 10 weeks beginning in December 1957, to increase the core size to 69 elements and to remove the boron poison strips. When the reactor was started in February, it was found that gaseous fission products were present in the condensate return. By sampling the air ejector flow these gases were found to be mainly Xe^{138} and Kr^{88}. The amount of short-lived xenon present relative to the longer-lived krypton was less than would be predicted from fission yields. It appeared, therefore, that before the gases escaped from the defective fuel element to the steam they were required to diffuse some distance. The activity in the reactor water itself showed no substantial increase over normal operating conditions. This indicated that if water were contacting the fuel pellets, they were not releasing activity through corrosion or leaching. Location of the defective element or elements has proved to be unexpectedly difficult, so that as yet no information is available concerning either the nature of the defect or its location.

In order to study the effect of irradiation on a defected ThO_2-UO_2 fuel assembly under more controlled conditions, R. E. Bailey and R. F. S. Robertson recently conducted an experiment in EBWR in which several fuel pellets were irradiated in a stainless steel tube with a 0.010-in. hole drilled in the end of the tube. The defected tube was operated in the reactor for eight days at various reactor-power levels, including 30 hr at full power. During this run the heat flux from the cladding surface of the defected assembly was ~ 330,000 BTU/hr-ft^2. The pressure in the reactor vessel and the power generation were varied during the test so that the defect "breathed" reactor water in and out to repeatedly waterlog the pellets.

The gaseous fission products Xe^{138} and Kr^{88} were detected in the gases from the air ejector, just as they had been at Borax-IV. At full power, for example, Xe^{138} was released at about 2.4 curies/day. Slight traces of only one solid fission product, I^{131}, were found in the reactor water. The results from these preliminary studies may be compared to those reported by Westinghouse investigators[4] for defected UO_2 rods. In their loop experiments in the NRX reactor, release of not only xenon and krypton was encountered, but also numerous solid fission-product activities were found in the water. Since essentially only gaseous activities have been found in the Borax-IV and in the EBWR defect tests, there is some indication that defected ThO_2-UO_2 fuel ele-

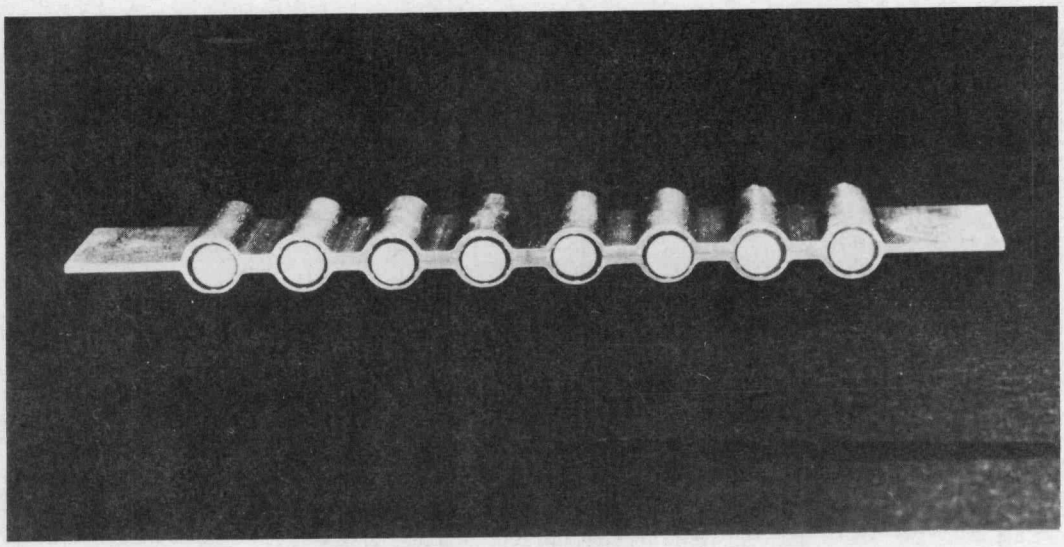


Fig. 8—Cross section of Borax-IV fuel plate.

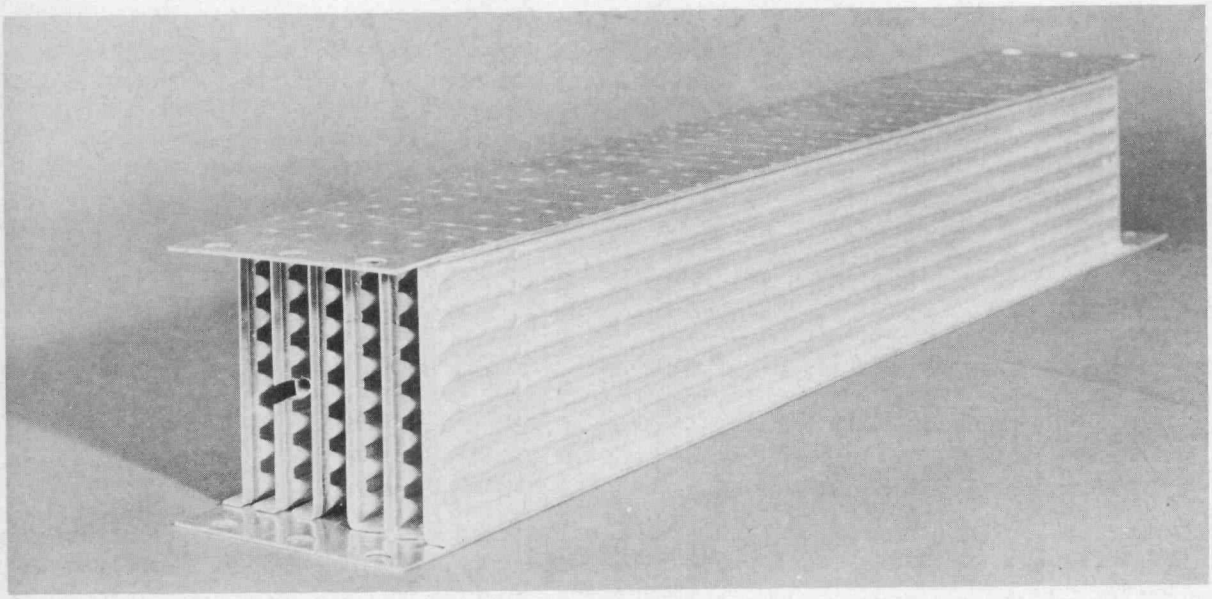


Fig. 9—Borax IV fuel subassembly.

ments may be somewhat more resistant to leaching of solid fission products by hot water than are defected UO_2 fuel elements.

CONCLUSIONS

In conclusion, it can be stated that irradiation experience to date has shown that ThO_2-UO_2 fuels are highly stable under irradiation. Fuel elements containing this ceramic fuel have shown excellent performance in arrangements where the pellets are lead-bonded to aluminum alloy cladding. Preliminary tests indicate that essentially only gaseous fission products are released from a defected fuel element.

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2. Handwerk, J. H., Hoenig, C. L., Lied, R. C., and Bach, R. A., Manufacturing of the ThO_2-UO_2 Ceramic Fuel Pellets for Borax-IV, ANL-5678 (1957).

3. Walker, D. E., Noland, R. A., McCuaig, F. D., and Stone, C. C., Borax-IV Reactor: Manufacture of Fuel and Blanket Elements, ANL-5721 (1958).

4. Eichenberg, J. D., et al, Effects of Irradiation on Bulk UO_2, WAPD-183 (1957).

DISCUSSION

MR. J. P. HAMMOND (Oak Ridge National Laboratory): How do you put the fuel material in the element? Do you use silicon bonding as was described for the ALPR?

MR. KITTEL: Where the ends are bonded, yes.

MR. HAMMOND: Is this also an M-388 aluminum alloy?

MR. KITTEL: Yes.

MR. HAMMOND: How does this silicon interface affect the corrosion resistance of this aluminum alloy under pressurized water conditions? Have you investigated this?

MR. KITTEL: These elements have been subjected to numerous corrosion and autoclave tests, and to my knowledge no problem has existed at the bond line where they might be expected to be less corrosion resistant.

MR. J. C. DANKO (Westinghouse Atomic Power Division): Have you done any work on the UO_2 end of the diagram with regard to ThO_2 additives and sinterability in air?

MR. KITTEL: Yes, the ceramists have been able to add as much as 78% UO_2 to ThO_2 and still successfully sinter the pellets in air. We are now irradiating material which has as much as 26% UO_2 in ThO_2 and this also was sintered with no difficulty in air.

CHAIRMAN BILLINGTON: I should like to ask whether you attempted to measure thermal conductivity of those original pellets?

MR. KITTEL: No, we did not. For thermal conductivity we have relied on published data for ThO_2.