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VACUUM DEGASSING OF NUCLEAR FUEL

by

D. W. Brite

Before discussing details of vacuum outgassing of nuclear fuels, it may be appropriate to briefly describe what a nuclear fuel element is, how it is fabricated, what its internal and external operating conditions are, and the nature of some of the volatile impurities found in nuclear fuel materials. After describing techniques for removing undesirable gaseous constituents from fuel materials, I will show a few slides illustrating how these impurities can cause fuel element failures if they are not removed from the fuel.

A nuclear fuel assembly for use in commercial light water cooled power reactors is similar to the drawing shown in Figure 1. It comprises a bundle of fuel rods held together by end plates and maintained at a uniform spacing with spacer grids at intervals over the length of the assembly. The fuel rods vary from $3/8$ to $1/2$ inch diameter and from 6 to 12 feet long. Depending upon the reactor type, the assembly contains from 36 to 289 fuel rods. A reactor core may consist of as many as 700 fuel assemblies.

Each fuel rod consists of a metal cladding tube, plenum spring, and end caps, and a column of enriched uranium dioxide or uranium-plutonium mixed oxide sintered fuel pellets. Most reactors use zirconium alloy fuel cladding because it has a very low thermal neutron absorption cross-section. A plenum is provided to accommodate gases generated in the fuel by fission of the uranium or plutonium. A spring is provided in the plenum to maintain

the fuel pellets in their proper position within the fuel rod and to prevent collapse of the cladding from the high external coolant pressure in the reactor.

In fuel development work for the USAEC Plutonium Recycle Program at Hanford, initially under management of the General Electric Company, and since 1965 by Battelle-Northwest Laboratories, fuel element design was similar to that shown in Figure 2. Nineteen fuel rods were held together in a bundle, spacing was provided by wires wrapped around some of the rods, and the fuel consisted of a $\text{UO}_2\text{-PuO}_2$ particle mixture which was loaded in the tubes by a vibrational compaction process. Although sintered pellet type fuel has become the present industry standard, packed-particle fuels are considered a promising alternate fuel form, and additional development of this concept is continuing at several commercial fuel fabrication plants in the U.S. and elsewhere. This concept is particularly well suited for the limited contact, glove box processing required for fabricating plutonium fuels, especially so for the high radiation type of plutonium produced in power reactors.

A typical industrial process for fabricating sintered UO_2 fuel pellets is illustrated in Figure 3. Uranium dioxide powder having a particle size of about one micron average diameter for sinterability considerations, is converted to a free flowing mixture which gives uniform die-filling in an automatic pellet press by a pre-pressing, granulating, and sieving procedure. An organic die lubricant powder is blended into the mixture and pellets are pressed in an automatic hydraulic press at about 10 to 30 tons per square inch. The pellets are sintered in a hydrogen atmosphere furnace at 1500 to 1700°C to a density of 92 to 95% of the theoretical UO_2 density, and ground to the specified diameter in a centerless grinder. After inspection, the

ground pellets, are vacuum outgassed at temperatures to 800°C prior to loading into fuel rods.

Fabrication of fuel rods is illustrated in Figure 4. A pre-weighed and measured column of fuel pellets is loaded into a zirconium alloy cladding tube having one end cap already welded in place. A plenum spring is added, the assembly is evacuated to 10^{-4} Torr in a welding box, backfilled with helium and the second end cap is welded. Welded fuel rods are etched in a nitric-hydrofluoric acid solution and autoclaved at about 400°C at 1000 psi to form a protective black oxide surface film. After autoclaving, fuel rods are helium leak checked and final inspected prior to being assembled into fuel elements.

The fabrication process for packed-particle fuels is similar to that for pellet fuels. High density fuel particles can be prepared by several different techniques, such as arc fusion, sol-gel processing, hot pressing, or high energy rate impaction, followed by crushing and screening to obtain optimum particle size distribution. The required amount of fuel is poured into the cladding tube while vibrating it at 5 to 5,000 cps using an electrodynamic vibrator. Fuel bulk densities as high as 90% of the theoretical UO_2 density can be obtained. After loading, additional processing of the fuel rods is the same as that for pellet fuels.

During reactor operation the fuel rod surface temperature is in the range of 300 to 400°C. Because of the relatively low thermal conductivity of ceramic materials such as UO_2 , there is a very large difference between the temperature of the fuel at the center and that of the fuel in contact with the cladding. The difference may be as much as 2000°C over a radial distance of 0.15 inch, for those fuel rods operating at maximum rated power

levels. Extremely high fuel central temperatures cause extensive grain growth and restructuring of the fuel. This effect is shown in the next two figures. Figure 5 illustrates cross-sections of fuel rods operated at the relatively low power level of 8 KW/ft. There is no obvious grain growth in the fuel, indicating that the maximum UO_2 central temperature was less than about $1700^{\circ}C$. The only apparent change, compared to the microstructure of unirradiated fuel, is the extensive cracking in the pellet fuel. Figure 6 shows cross-sections of fuel operated at about 18 KW/ft, which is approximately equal to the maximum design peak power condition for commercial reactors currently under construction. Extensive grain growth has occurred in the central portion of the fuel. Large columnar grains have formed by a vaporization-condensation process, resulting in migration of pores up the thermal gradient and formation of a central void. As a result of fuel restructuring such as this, it is apparent that volatile impurities adsorbed on the surface of the fuel material or contained in the pores or as interstitial impurities may become available for reaction with the fuel cladding during reactor operation.

Volatile impurities which may be present in UO_2 fuel pellets include adsorbed gases such as nitrogen, carbon dioxide, carbon monoxide, water and hydrogen. There has been considerable experience, in the case of packed-particle fuel, with oil contamination introduced from equipment used in crushing the fuel material. Limitations on volatile impurities permitted in nuclear fuels are based on both the internal pressurization of the fuel rod and the adverse effects of certain gases on the properties of the zirconium alloy cladding. The plenum provided in fuel rods is necessary to accommodate buildup of gases generated by fission of the uranium or plutonium in the fuel during reactor operation. The total volume of gases extractable from

UO₂ fuel materials in vacuum at 1600°C permitted in typical commercial fuels specifications is on the order of 0.05 to 0.10 cm³ per gram of fuel.

Vacuum outgassing procedures used with UO₂ and UO₂-PuO₂ fuel pellets and fuel particle mixtures consist of loading the material in stainless steel trays or cans and heating in a vacuum of 25 microns or less in a cold wall furnace of the type shown in Figure 7. For sintered pellets, where the principle volatile impurities are adsorbed moisture and residual hydrogen in the pores, outgassing temperature is usually 500 to 800°C to insure complete volatilization of water combined with UO₂ as a hydrate compound. In the case of suspected oil contamination of particle fuels, mentioned earlier, outgassing temperature was kept below the decomposition temperature of oil to obtain complete volatilization of the oil without leaving residual hydrocarbon residues. Outgassing conditions determined to be sufficient to remove oil from a deep bed of UO₂-PuO₂ powder were 250°C for 8 to 12 hours at a vacuum of 10⁻³ Torr. Vacuum outgassed fuel is transferred directly from the outgas furnace to the rod loading facility, or stored in a dry atmosphere to prevent readsorption of moisture.

The primary reason for excluding volatile impurities from nuclear fuel materials is the susceptibility of Zircaloy cladding to reaction with hydrogen-containing materials to form zirconium hydride, which precipitates in the metal, causing deterioration of its physical properties. The reaction often occurs in a localized region forming a massive deposit of zirconium hydride which extends completely through the cladding to form a blister on the outside surface as a result of the volume increase due to phase change. The crevice region associated with end plugs is a frequent location for localized cladding failure. Figure 8 shows a typical end cap crevice failure which occurred in a PRTR packed-particle fuel rod. Because of the localized nature of the attack, the hydrogen associated with small concentrations of

impurities that are well within fuel material specifications may be more than sufficient to cause localized cladding failure. For example, one hydride blister, similar to the one shown in Figure 9, is estimated to contain about 1 mg of hydrogen. Ten ppm H_2O in the fuel in a typical BWR fuel rod will provide about 3 mg of hydrogen. The moisture content in typical BWR fuel material is generally about 10 ppm or slightly higher.

Greater details of the structure in typical hydride defects are shown in Figure 10. The reason for localized attack of Zircaloy by hydrogen is not completely understood. However, there is evidence which leads one to suspect that localized attack may be nucleated by imperfections in the zirconium oxide layer covering the inner surface of the fuel cladding. Fortunately, it also appears that Zircaloy can accommodate a certain amount of hydrogen in solid solution before zirconium hydride is precipitated. In addition, hydrogen can diffuse in Zircaloy at a significant rate at operating temperatures, so that gradual release of hydrogen from the hydrogenous impurities can be accommodated to a limited extent.

In conclusion, I would like to emphasize that by keeping hydrogen-containing impurities at reasonable concentration levels, whether by vacuum outgassing or by using fabrication techniques that preclude their presence, commercial nuclear fuel manufacturers are able to provide fuel assemblies with a very high reliability and a very low incidence of failures of the type I have described.

LIST OF ILLUSTRATIONS

<u>Figure No.</u>	<u>Negative No.</u>	<u>Description</u>
1	BN 069340-3CN	Typical Commercial Nuclear Fuel Assembly
2		PRTR Fuel Assembly
3	BN 705695-3CN	Typical Process Flow for Industrial Fabrication of Sintered UO ₂ Fuel Pellets
4	BN 705695-2CN	Typical Process Flow for Industrial Fabrication of Nuclear Fuel Rods
5		Typical Irradiated Mixed Oxide Fuels (~8 KW/ft)
6		Typical Irradiated Mixed Oxide Fuels (~18 KW/ft)
7	PNL 068 3799-4CN	Vacuum Furnaces for UO ₂ Outgassing
8		Typical PRTR End Cap Crevice Failure
9		Typical Hydride Defect
10		Typical Hydride Defect