United States Patent Office

3,331,748

Patented July 18, 1967

3,331,748

NUCLEAR FUEL ELEMENTS

Melville A. Feraday, Deep River, Ontario, Canada, assignor to Atomic Energy of Canada Limited, Ottawa, Ontario, Canada, a corporation of Canada

No Drawing. Filed Aug. 9, 1965, Ser. No. 478,466

9 Claims. (Cl. 176—70)

This invention relates to fuel elements for use in nuclear reactors, particularly power reactors. A corrosion-resistant, restrained, uranium-based fuel element is described, suitable for use in pressurized water, fog, or steam-cooled reactors to high burn-ups. An adaptation is described for organic-cooled reactors.

The fuel elements comprise an inner core of adjusted uranium metal (a), surrounded by and bonded to a uranium alloy (b), which is in turn clad in a sheath of a zirconium alloy (c). A central void space is provided in the metal core (a) with both the U alloy (b) and Zr alloy (c) providing restraint on the core such that any swelling is directed into the void space.

The major problems associated with the use of low alloy content uranium-based fuels in high temperature water are the poor corrosion resistance and the high degree of swelling under irradiation especially above 550° C. Limited corrosion resistance has been attained in the past by using bulk fuels containing fairly large amounts of alloying material e.g. U—10 wt. percent Mo. Reductions in swelling have been attained using a relatively thick stainless steel sheath for restraint and a small void down the centre of the U alloy into which the fuel could swell. Neither of these approaches is acceptable in reactors using natural uranium because of neutron absorption by the large amounts of sheathing and alloying materials involved.

Recently resistance to swelling of unalloyed uranium has been described when the uranium is "adjusted" with minor alloying additions of iron and aluminum or silicon (to less than about 2000 p.p.m. total) and heat treated. These minor additions do not significantly affect the neutron economy. High temperature strength and water corrosion resistance have been described for uranium alloys containing 20–60 wt. percent Zr. Aluminum alloys containing about 10 wt. percent uranium have been shown to have good ductility and good aqueous corrosion resistance.

According to the present invention, fuel elements having several layers have been designed to provide resistance to swelling, strength and corrosion resistance where needed.

The inner core contains uranium metal adjusted with minor alloying additions of both iron and aluminum. The iron may range from about 200 to about 500 p.p.m., preferably about 400 p.p.m. The aluminum may range from about 500 to about 1200 p.p.m., preferably about 700 p.p.m. This adjusted uranium metal core should be heat treated (beta quenched from about 750° C. and alpha annealed at about 550° C. for several hours) during fabrication of the fuel element to refine and randomize the grain structure. The core is constructed so as to contain a central void space of volume about 5 to about 15% based on the fuel element and depending on the operating conditions and characteristics. Preferably the void space is about 7% of the uranium volume.

Surrounding the uranium metal core, a middle layer comprising an uranium alloy is provided to give increased corrosion resistance, strength and restraint to the fuel. Two material concepts are considered for the middle layer: (1) One in which the layer is quite strong but with limited ductility, e.g. uranium with 30 to 60 wt. percent zirconium and (2) The second in which the layer is extremely ductile, e.g. Al–5 to 15 wt. percent U. By this means a two component fuel is obtained which has better neutron economy than any known single component fuel of similar swelling and corrosion resistance. Suitable uranium alloys contain an element selected from the group consisting of Zr, Mo, Nb and Al. Zr and Al are the preferred elements and the concentrations required are about 30 to 60 wt. percent zirconium or 50 to 95 percent aluminum. The concentrations of the Mo and Nb alloys are restricted to about 1 to 25 wt. percent (preferably 1 to 10) because of their higher nuclear cross-section. The amount of the alloy, its concentration and the layer thickness may vary widely depending on the application, and on the nuclear, physical and corrosion characteristics of the alloy.

Depending on the application the amount of the U alloy may vary considerably. For small diameter fuel rods the uranium alloy layer is usually about 15–30 vol. percent of the metal core and the layer thickness about 0.1 cm. However, since the change in electrical energy costs with layer thickness is small, soundness in design will dictate layer thickness. The ends of the uranium core are also bonded to alloy discs or end plates to complete the shell.

For service where dimensional stability is required, but where water corrosion resistance is not important, e.g. in an organic cooled reactor, a modified design may be used in which strengthening of the outer part of the adjusted uranium fuel is achieved by using alloys which are not highly water corrosion-resistant or by dispersion strengthening a surface layer of the uranium with powder or whiskers of non-metals. Examples of the non-corrosion-resistant alloys which may be used are U alloys containing low concentrations of one of Mo, Nb (1–3 wt. percent) and Al (5–30 wt. percent). The dispersion strengthening may be achieved by the addition of certain non-metals for example alumina.

An outer cladding or sheath is normally provided for corrosion resistance and added strength. The outer cladding is desirably a corrosion-resistant zirconium alloy, preferably Zircaloy-2 which consists of tin 1.2 to 1.7 wt. percent, iron 0.07 to 0.2 wt. percent, chromium 0.05 to 0.15 wt. percent, nickel 0.03 to 0.08 wt. percent, oxygen 1400 p.p.m. maximum, total Fe+Cr+Ni 0.18 to 0.38 wt. percent, and balance zirconium plus impurities. The following zirconium alloys are also suitable: Zr containing about 2.5 wt. percent Nb, and Zircaloy-4.

On irradiation, dimensional changes are avoided by the restraint of the outer two layers forcing any swelling which does occur into the central void. The irradiation-induced swelling of the unrestrained adjusted uranium metal is about 16 vol. percent per one atomic percent burnup at 575° C. Should a sheath defect occur, the corrosion resistance of the uranium alloy to high temperature

water is good, but allows limited corrosion to provide a monitoring signal that a defect has occurred. The corrosion rates of U–45 wt. percent Zr and Al–10 wt. percent U in 300° C. water are about 0.1 and 50 mg./cm.²/hr. respectively compared to $>10^4$ mg./cm.²/hr. for uranium metal. The external diameter of the fuel element, the thickness of the restraining shell and the size of the central void can vary considerably depending on the requirements of the particular reactor. Lower competitive neutron absorption is obtained than could be attained with a single component fuel having similar corrosion and swelling behaviour.

The fuel elements can be made by one or more of the following methods:

(1) Single temperature or multi-temperature co-extrusion. This method is the most attratcive one economically and technically.

(2) Individual machining of sections and shrink fit assembly. Intimate mechanical contact will be provided in this manner which will minimize the central uranium temperature.

(3) Individual machining of sections and diffusion bond assembly. This diffusion bonding treatment serves to reduce interface temperature gradients to a minimum.

(4) Co-extruding the alloy shell and sheath and casting a central core of uranium into that assembly. This method will produce bonded fuels without requiring multi-temperature extrusions.

(5) Extrusion clad the alloy shell onto the metal core (a); this assembly is then slip fitted into the zirconium alloy sheath.

The naturally-occurring mixture of uranium isotopes is normally used, although enriched uranium can be used in either or both of the fuel layers, if desired.

Preferred embodiments are given in the following examples:

(1) Uranium metal is adjusted by the addition of 400 p.p.m. Fe and 700 p.p.m. Al to the melt. The adjusted metal is co-extruded with U–45 wt. percent Zr alloy and a Zircaloy-2 sheath at about 650° C. The extrusion should be heated to the beta region (about 750° C.), water quenched and annealed at about 550° C. for several hours in order to randomize and refine the uranium structure, precipitate out some of the adjusting additions, and epsilonize the uranium zirconium.

(2) Uranium metal is adjusted by the addition of 400 p.p.m. Fe and 700 p.p.m. Al to the melt. The adjusted metal is then heated to the beta region (about 750° C.), water quenched and annealed at about 550° C. for several hours to randomize and refine the uranium structure and precipitate out some of the adjusting additions. The uranium is then extrusion clad in Al–10 wt. percent U at about 525° C.; this rod is then slip fitted inside a Zircaloy sheath.

The final fuel rod diameter is 1.52 cm. while the diameters of the void, inner uranium core and uranium alloy outer layers are approximatley 0.4, 1.28 and 1.45 cm. respectively. The fuel rods are cut to 19 inches length and arranged to give a 19 element bundle (as for the NPD Rolphton reactor). Bundles of 22 or 28 elements of smaller diameter would be even more attractive. The metallurgical bonds between the uranium core and the uranium alloy layers in these examples are at least as good as between uranium and Zircaloy-2. The uranium alloy layer may or may not be bonded to the Zircaloy-2 sheath, depending on the application.

(3) URANIUM—ZIRCONIUM

(a) Test specimens have been made by arc casting uranium metal into an outer shell of uranium–47 wt. percent zirconium to produce a metallurgical bond. The open end was sealed by welding a plug of U–47 wt. percent Zr onto the outer shell of U–47 wt. percent Zr using a technique developed for welding Zircaloy-2. This specimen was then slip-fitted inside a Zircaloy-2 sheath and Zircaloy-2 end plugs were welded on to produce a sealed element (5 cm. long x 2 cm. diameter).

(b) Specimens of the U–47 wt. percent Zr of similar size were corrosion tested in 300° C. water both bare and clad in a 0.060 cm. thick defected Zircaloy-2 can in 300° C. water. The bare U–47 wt. percent Zr corroded uniformly without any pitting and at a rate of about 0.1 mg./cm.²/hr. The specimen clad in the defected Zircaloy-2 can showed little visible change after 360 hours at 300° C. Dimensionally, the can had swelled by about 0.001 cm., but no signs of hydriding could be found in the Zircaloy-2.

(4) ALUMINUM-URANIUM

A sample (4.5 cm. long x 0.5 cm. diameter) of Al–10 wt. percent U tested bare in 300° C. water for thirty minutes had a corrosion rate of about 50 mg./cm.²/hr. A four hour defect test in 300° C. water of a similar sample clad in 0.030 cm. thick Zircaloy-2 resulted in a short split and a slight bulge in the sheath. Examination of a cross section of this element indicated that a less than 0.1 cm. thick shell of Al–10 wt. percent U is adequate to give about four hours protection in embodiment No. 2 above.

I claim:

1. A fuel element for nuclear reactors comprising:
   (a) an inner layer of uranium metal containing from 200 to 500 p.p.m. iron, from 500 to 1200 p.p.m. aluminum, and beta quenched and alpha annealed to refine and randomize the grain structure, and enclosing a central void space,
   (b) a middle layer comprising one of (1) an uranium alloy containing an element selected from the group consisting of Zr, Mo, Nb and Al, the Zr being present in from 30 to 60 wt. percent, the Mo and Nb in from 1 to 25 wt. percent, and the Al in from 50 to 95 wt. percent, and (2) uranium dispersion-strengthened with alumina, and
   (c) an outer sheath of a corrosion-resistant zirconium alloy.

2. A fuel element for nuclear reactors comprising:
   (a) an inner layer of uranium metal containing about 400 p.p.m. iron and about 700 p.p.m. aluminum, and beta quenched from about 750° C. and alpha annealed at about 550° C., and enclosing a central void space,
   (b) a middle layer of an uranium alloy containing from 30 to 60 wt. percent of Zr, and
   (c) an outer sheath of a corrosion-resistant zirconium alloy.

3. A fuel element for nuclear reactors comprising:
   (a) a central void space surrounded by an inner layer of uranium metal containing about 400 p.p.m. iron, about 700 p.p.m. aluminum and beta quenched from about 750° C. and alpha annealed at about 550° C.,
   (b) a middle layer of uranium–45 to 47 wt. percent zirconium alloy, and
   (c) an outer sheath of the zirconium alloy Zircaloy-2.

4. A fuel element for nuclear reactors comprising:
   (a) a central void space surrounded by an inner layer of uranium metal containing about 400 p.p.m. iron, about 700 p.p.m. aluminum and beta quenched from about 750° C. and alpha annealed at about 550° C.,
   (b) a middle layer of aluminum–10 wt. percent uranium, and
   (c) an outer sheath of the zirconium alloy Zircaloy-2.

5. The fuel element of claim 1 wherein the uranium alloy in middle layer (b) contains one of from 1 to 10 wt. percent Mo, from 1 to 10 wt. percent Nb, and from 85 to 95 wt. percent Al.

6. The fuel element of claim 1 wherein the void space is about 5 to 15 vol. percent of the fuel element.

7. The fuel element of claim 1, including end plates of the middle layer (b) bonded to the inner layer (a), and to middle layer (b).

8. The fuel element of claim 1, wherein the middle layer (b) is about 15–30 vol. percent of the inner layer (a).

9. The fuel element of claim 1, in a bundle of from 19 to 28 elements.

References Cited

UNITED STATES PATENTS

2,805,473	9/1957	Handwerk et al.	176—89 X
2,830,896	4/1958	Seybolt	176—89 X
2,914,433	11/1959	McGeary et al.	176—89 X
2,917,383	12/1959	Saller	176—89 X
2,926,113	2/1960	McGeary et al.	176—89 X
3,010,890	11/1961	Jepson et al.	76—122.7 X
3,015,615	1/1962	Precht et al.	176—89 X
3,109,797	11/1963	Maxwell	176—69
3,114,688	12/1963	Wyatt et al.	264—.5 X
3,160,951	12/1964	Market et al.	176—91 X
3,243,350	3/1966	Lustman et al.	176—67 X
3,285,737	11/1966	Bellamy	176—70 X

CARL D. QUARFORTH, *Primary Examiner.*

BENJAMIN R. PADGETT, *Examiner.*

N. J. SCOLNICK, *Assistant Examiner.*