SURVEY ON METAL FUEL ON A BASE OF URANIUM ALLOYS

I.I. KONOVALOV
All Russian Institute of Inorganic Materials,
Moscow, Russian Federation

Abstract

The thorough study of metallic fuel on a base of uranium alloys was carried out in VNIINM for different types of reactors. The main characteristics of irradiation behaviour and form change of metallic fuel are summarized. The best results were obtained for alloys containing 1-2 wt.% of γ-stabilizing elements (Zr, Nb, Mo) + several tenths of percentage of compound forming elements (Si, Fe, Al, Sn). It is stated that metallic uranium fuel in a bulk form may be used only for fast reactors, and in dispersed form with Al- and Zr- matrix for commercial LWR and research reactors.

Introduction

The metallic fuel on a base of uranium alloys was developed for different types of nuclear reactors: plutonium production reactors, commercial power reactors, fast reactors, and for other types of reactors.

The initial interest to metallic uranium fuel was caused by its high density and opportunity to produce weapon grade plutonium. The first irradiation experience was the severe form changes due to irradiation growth and swelling, fuel rod cracking under thermal stresses. The massive applied and fundamental studies on irradiation behaviour of uranium and its alloys were carried out. This permitted us to understand the bases of radiation damage in uranium, and eliminate negative radiation phenomena in plutonium production fuel elements.

Development of metal fuel

Along with the development of oxide fuel the vast study of metallic fuel was begun, as it has the following obvious advantages:
- high uranium density
- high level of mechanical properties
- absence of gas release under irradiation
- high thermal conductivity; in case of cohesion between fuel rod and cladding it is possible to produce "cold fuel element" for VVER or RBMK-types nuclear reactors with average fuel temperature less than 400°C.

But, the metallic fuel has also serious disadvantages: the first is swelling of fuel, and the second - low compatibility with water coolant.

To investigate behaviour of metallic fuel we use so called "passive" and "active" reactor experiments. "Passive" reactor experiments are usual study, where one irradiates samples of fuel or mock-up fuel elements and then investigates them by common post-irradiation methods. "Active" reactor experiments are carried out with the help of units enabling us to influence upon fuel samples during irradiation.

The one unit is the device for in-pile investigation of a fuel under tension. For example the mechanical properties of the low-alloyed uranium under irradiation and after are shown in Fig.1. One may see the significant difference in properties.

The other unit is the dilatometer inserted in channel of research reactor. This unit allows us to measure continuously the change of sample dimensions under irradiation. For example at the bottom in Fig.2 the kinetic of the swelling of low-alloyed uranium is
Fig. 1. Mechanical properties of the low-alloyed uranium under and after irradiation.
Fig. 2. Volume changes of the low-alloyed uranium (bottom) and uranium silicide (upper).

Simultaneously with measuring of sample dimensions we may load a sample and so study irradiation creep. For instance in Fig. 3 the irradiation and thermal creep of uranium silicide $U_3Si$ is shown.

On a base of experimental data and computed study we worked out the main characteristics of irradiation behaviour and form changes of metal fuel.
The primitive dependence of the swelling from relative temperature and burnup for low-alloyed uranium is given in Fig. 4. The darkened areas correspond to working conditions of metallic fuel in research reactors, commercial water cooled reactors and fast reactors.

At temperatures below 0.4 from melting point the swelling is caused by accumulation of fission products (solid swelling). The burnup dependence is linear function with swelling rate about of 6 vol.% per $1.10^{11}$ fiss./cc (or 15 vol.% per 1 g of splitted uranium in 1 cc).

At intermediate temperatures the maximum of swelling is caused by vacancy pores, containing some amount of gas fission products. If we anneal samples after irradiation, they increase the density and their volume decreases.

At high temperatures above of 0.6 from melting point the swelling is due to equilibrium gas bubbles, and so post-irradiation annealing do not reveal any significant volume changes.

At high burnups the maximum of vacancy swelling degenerates and swelling has the gaseous nature. With the increasing of burnups the temperature threshold of the beginning of gas swelling displaces to lower temperatures.

The low-temperature solid swelling and high-temperature gas swelling cannot be suppressed, and the only mean to struggle against these phenomena is to create the free volume in a fuel element which will compensate the swelling, or use such design, which permits deformation of fuel element without rupture.

In opposite, the maximum of vacancy swelling under intermediate temperatures may be effectively suppressed by adjustment of microstructure of uranium fuel. The main approach is creation in fuel structure a great amount of sinks for radiation point defects.

As the sinks, the fine dispersed intermetallic compounds may be considered. For this purpose compound forming elements such as Si, Fe, Sn, Al and others are added to uranium.
This method of suppressing the vacancy swelling works when density of intermetallic precipitation's is more than $1.10^{12}$ particles in cc. At the bottom in Fig. 2 the kinetics curves of swelling for uranium with additions of intermetallic forming elements are shown. The composition of samples is the same, but for upper curves we had a coarse network of intermetallic precipitation's and for lower - fine network. This method is effective for relatively low burnup.

For higher burnup typical to commercial reactors, we combine this method with another - creation of the fine two-phase structure. This is the most effective if we use $\gamma$-stabilizing elements, such as Zr, Nb, Mo. Using of these elements we may create fine two-phase structure, where boundaries between phases serve as point defects sinks. Example of the influence of the degree of dispersity of two-phase structure ($\alpha$-Uranium + $\gamma$-Uranium) upon irradiation behavior of mock-up fuel elements is given in Fig. 5.

Experimentally we established that less than 2 wt.% of $\gamma$-stabilizing elements + several tenths of percentage of compound forming elements are enough to suppress the vacancy swelling. We must only do the correct choice of alloying element's combination and heat-deforming treatment. The further alloying has insignificant influence upon irradiation behaviour.

Now we see no difficulties to develop fuel elements with uranium alloy which can work to burnup that of commercial reactors.

The main problem why metallic fuel has limited application in atomic energetic is insufficient corrosion resistance in water coolant.

The distinguishing feature of uranium is absence of solubility of other elements in $\alpha$-uranium, and so we cannot use common solid-solution methods for improving corrosion
Macrograph (appearance) of fuel elements

Optical micrograph (x800)

Fig. 5. Influence of the degree of dispersity of two-phase structure (α- and γ-uranium) on irradiation behaviour of mock-up fuel elements (dose = $1 \times 10^{11}$ f/ś).
resistance. The significant improving of properties may be only archived if we use δ- and γ-phase alloys in U-Mo, U-Nb, U-Ti, U-Zr systems. To receive these structures we must insert in U a significant quantity of alloying elements from 30 to 75 at.%. In ternary alloys we may obtain corrosion resistant metastable γ-phase by minor alloying. But as it metastable there is some problems with stability of γ-phase under irradiation.

A good corrosion resistance has uranium silicide U$_3$Si. This compound was thoroughly investigated as a fuel for RBMK reactor. The fuel elements with this fuel had satisfactory irradiation behavior, but in case of untight fuel element when the water coolant got into the fuel rod center with the temperature of about 500°C, the whole fuel element was destroyed.

The radical measure to increase the corrosion resistance is to use metallic fuel dispersed in Al- or Zr- matrix. We have preliminary positive results of irradiation to relatively (~1g/cc) high burnup of such dispersion composition.

Conclusion

Summarizing our experience we may say that metallic fuel in a bulk form may be used in fast reactors with sodium coolant, and in dispersion form for atomic energetic and research reactors.
Bibilashvili, Yu.K. All Russian Scientific and Research Institute of Inorganic Materials, Rogova Str. 5, Box 369, 123060 Moscow, Russian Federation

Boero, N.L. Comisión Nacional de Energía Atómica, Av. del Libertador 8250, 1429 Buenos Aires, Argentina

Coquerelle, M. Institute for Tansuranium Elements, Joint Research Centre, P.O. Box 2340, D-76125 Karlsruhe, Germany

Dehaudt, P. CEA/DTP/SECC, 17, Rue des Martyrs, F-38054 Grenoble Cedex 09, France

Denis, A. Comisión Nacional de Energía Atómica, Av. del Libertador 8250, 1429 Buenos Aires, Argentina

Golovchenko, Yu.M. Research Institute of Atomic Reactors, 433510 Dimitrovgrad-10, Ulyanovsk region, Russian Federation

Kim, Ki-Hwan Korea Atomic Energy Research Institute, Dukjin-dong Yusong-gu 150, 305-353 Taejon, Republic of Korea

Konovalov, I.I. All Russian Scientific and Research Institute of Inorganic Materials, Rogova Str. 5, Box 369, 123060 Moscow, Russian Federation

Kurina, I.S. Institute of Physics and Power Engineering, State Scientific Centre of the Russian Federation, 249020 Bondarenko Square 1, Obninsk, Russian Federation

Moseev, L.I. Institute of Physics and Power Engineering, State Scientific Centre of the Russian Federation, 249020 Bondarenko Square 1, Obninsk, Russian Federation

Onoufriev, V. (Scientific Secretary) International Atomic Energy Agency, Wagramer Strasse 5, P.O.Box 100, A-1400 Vienna, Austria

Panov, A.S. Scientific and Research Institute LUTCH, 142100 Podolsk, Russian Federation

Popov, V.V. Institute of Physics and Power Engineering, State Scientific Centre of the Russian Federation, 249020 Bondarenko Square 1, Obninsk, Russian Federation
Rogozhkin, B.D. All Russian Scientific and Research Institute of Inorganic Materials, Rogova Str. 5, Box 369, 123060, Moscow, Russian Federation

Ryzkov, A.N. Institute of Physics and Power Engineering, State Scientific Centre of the Russian Federation, 249020 Bondarenko Square 1, Obninsk, Russian Federation

Sarakhova, G.A. All Russian Scientific and Research Institute of Inorganic Materials, Rogova Str. 5, Box 369, 123060 Moscow, Russian Federation

Sengupta, A.K. Radiometallurgy Division, Bhabha Atomic Research Centre, 400085 Trombay, Mumbai, India

Sila-Novitsky, A.G. Research and Development Institute of Power Engineering, P.O. Box 788, 101000 Moscow, Russian Federation

Stetsky, Yu.A. All Russian Scientific and Research Institute of Inorganic Materials, Rogova Str. 5, Box 369, 123060 Moscow, Russian Federation

Subbotin, A.V. Research and Development Institute of Power Engineering, P.O. Box 788, 101000 Moscow, Russian Federation

Suzuki, Y. Department of Chemistry and Fuel Research, Japan Atomic Energy Research Institute, Oarai-machi, Higashi-ibaraki-gun, Ibaraki-ken 311-13, Japan

Trotabas, M. COGEMA BC, 2, Rue Paul Dautier, B.P. 4, F-78141 Velizy Cedex, France

Vatulin, A.V. All Russian Scientific and Research Institute of Inorganic Materials, Rogova Str. 5, Box 369, 123060 Moscow, Russian Federation

Weidinger, M. Danziger Str. 22, D-90491 Nürnberg, Germany