Reactor Boiler and Auxiliaries - Course 133

GENERAL CONSIDERATIONS

The subject of nuclear fuels is an extremely broad one, since the forms in which nuclear fuels can be produced are many and varied. This lesson will deal mainly with the effects of irradiation on metal fuel, U, and the ceramic fuel, UO_2, from a general viewpoint. No attempt will be made here to fully explain the phenomena described.

Metallic Uranium

The effects of irradiation of metallic uranium on its mechanical properties are a generally marked decrease in tensile strength even after short exposure, changes in hardness (cold-worked uranium irradiated to low exposure softens, whereas annealed uranium hardens), decreased impact strength of cast, heat-treated or hot-rolled uranium by a factor of 3 or 4. Embrittlement of the metal does occur, so that any bend tests done on slender specimens have been found to fail by brittle fracture. The changes in properties can be generally said to depend directly on the integrated exposure to neutrons, and the neutron flux level.

The physical properties of metallic uranium are markedly affected by irradiation. Limited data from in-reactor measurements indicate a decrease in thermal conductivity of 10 to 15%. This property is an important one because decreases in thermal conductivity on irradiation might result in an increase of uranium core temperature to an unsafe level. Since the predicted accuracy of the methods used to obtain these data is probably ± 20% at best, the validity of the results in an absolute sense is dubious. Qualitatively, however, the results are quite valuable, in that they indicate there is no gross change in thermal conductivity of uranium after irradiation. An apparent increase in the modulus of elasticity of about 15% has, however, been reported.

The most significant effects of exposure to neutron flux on natural uranium metal are the pronounced loss in ductility, the disintegration of the metal at nominal burn-ups, the increases in volume and the growth of preferably oriented specimens. The changes in tensile properties are much too great to be attributed to vacancy-interstitial or thermal spike effects. The decrease in room temperature ductility may be attributed to the presence of fission products. It is quite probable that the noble gases xenon and krypton have a pronounced effect on the mechanical properties, and they are believed to be primarily responsible for the disintegration of certain types of uranium after relatively short burn-ups. Unalloyed enriched uranium disintegrates after low burn-up and resembles coke, suggesting that the formation and expansion of gas bubbles may play a part in the disintegration.

The presence of fission products leads to certain highly undesirable effects both in the fuel and, if they escape, in the coolant. At the instant of fission, the two fission products smash through the uranium lattice, dislodging uranium atoms, generating vacancies and interstitials, finally lodging as foreign atoms in the lattice. At sufficiently high temperatures, the gaseous fission products (Xe, Kr) diffuse and agglomerate, causing voids, gross local decreases in uranium density and even pronounced swelling. Swelling and decreases in density are often accompanied by cracking and eventual disintegration of the uranium; owing to the radioactivity of fission products, gross contamination of the reactor and/or reactor site may occur if they are released into the coolant by a fuel element failure or penetration of the sheathing.

Uranium metal offers a maximum conversion ratio, ie, a greater yield of plutonium per gram of uranium. However, the sheath of a solid fuel element must be capable of retaining all the fission products even though the fuel is distorted by irradiation and even though the properties of the sheathing material change. Metallic uranium has an anisotropic crystal structure. The effect on it of many fissions is to produce often dramatic changes in the shape of the fuel, which could lead to blockage of coolant flow in a channel. While this problem can be overcome by suitable metallurgical treatment, and does not exist at all in isotropic uranium dioxide, the problem of the accumulation of gaseous fission products remains.

Ceramic Fuel

The ceramic fuel in most common use today is uranium dioxide (UO_2). UO_2 has good corrosion resistance at high temperatures; this has been demonstrated in NPD where fuel elements have operated with defected sheathing for periods of over 2 years and post-irradiation examination of the UO_2 showed little or no difference between defected and undefected elements. Its coefficient of thermal expansion is low and it is dimensionally stable under long irradiation. It is capable of withstanding high temperature operation (melting point: 2800°C) and many thermal cycles.

UO_2 has, however, a low thermal conductivity, which limits the power rating that can be attained without melting. Power reactor operation in Canada to date has however been well below the UO_2 melting point. Fission product gases are largely retained by the UO_2 at lower temperatures but can leak out at high temperatures. Extensive irradiation tests to burn-ups as high as 100,000 Mwd/te U showed that the UO_2 swelled by no more than 8% of the volume when at temperatures below about 700°C, and restrained by 80 to 170 atm. While many unanswered questions still exist, recent studies on the irradiation behaviour of UO_2 conclude that the average fission product atom does not travel far before being trapped in the lattice of the material. The fission product gases can agglomerate and form observable bubbles; the ceramic UO_2 cracks under thermal stresses and must be contained and supported by a sheath.

The build-up of pressure inside the sheathing due to release of fission products from the UO_2 does not appear to be a problem with the Canadian design. Although in-reactor measurements are extremely difficult, the pressure within a fuel element at a burn-up of $\approx$ 10,000 Mwd/te operating at maximum Pickering heat rating is predicted to be $\approx$ 80 atm. While operating, this is balanced by the pressure in the coolant system and when removed or shut down, should be decreased by a factor of about 10 due to the decreased temperature of the UO_2, well below the burst strength of the tube. Less than 20% of the gases produced are released from the UO_2.

The thermal conductivity of UO_2 changes very little on irradiation; the factors affecting the conductivity of the UO_2 are the temperature, porosity, excess of oxygen and the impurities present.

The Meaning of $\int\lambda d\theta$

In conventional laboratory practice, one is accustomed to measuring the temperature distribution in a system and thus having knowledge of its condition. Alternatively, some other quantities, eg, heat generation, may be measured, and the temperatures deduced from established physical properties of the materials. However, the measurement of temperatures in operating fuel elements is extremely difficult, largely because of the high temperature gradients existing in them. Consequently, many of the physical properties of the constituent materials have not been determined under operating conditions. Specifically, the thermal conductivities of many fuels under irradiation have not been established. Some parameter which can be measured experimentally is required to define the condition of the fuel, in a manner comparable to temperature, and this parameter is

$\int_{T_s}^{T_o}\lambda\, d\theta$ where λ is the thermal conductivity of the fuel material, θ is temperature, T_s is the surface temperature, and T_o the central temperature. Usually, the symbol is simply written as

$$\int \lambda\, d\,\theta$$

The use of a definite integral as a single parameter, without reference to its components, will be familiar to those who have used "entropy". Although the parameter is most widely used in connection with UO_2 cylinders, the arguments leading to its association with measurable quantities are general and not confined to a single fuel. If the central fuel temperature were known, it would be a useful criterion for comparing the severity of different irradiations. In ignorance of it, $\int \lambda \, d\theta$, obtained from the equation

$$\int \lambda \, d\theta = \frac{1}{4\pi} q F$$

can be used for this purpose. In this equation, q is the heat generation per unit length of the pellet (W/cm) and F is a factor which accounts for the flux depression within the pellet. The flux depression in the Douglas Point or Pickering outer elements can be neglected, and F taken as unity. For larger diameter or enriched pellets, F must be calculated, and has a value less than unity.

As an example, the $\int \lambda \, d\theta$ for an average Douglas Point outer element, producing 500 W/cm would be given by

$$\int \lambda \, d\theta = \frac{1}{4\pi} (500)(1)$$

$$= 40 \text{ W/cm}$$

A graph, given as Fig. 1 has been included in this lesson to give some idea of the associated UO_2 temperatures. Reference to Fig. 1 will show that the upper temperature corresponding to

$\int \lambda \, d\theta = 40$ W/cm is about 1100°C; to

obtain the central UO_2 temperature, to this must be added the surface temperature of the UO_2, about 500°C in this case, giving the UO_2 central temperature at nominally 1600°C, well below the melting point of 2800°C.

ASSIGNMENT

1. How does UO_2 compare with metallic uranium as a high burn-up fuel, with regard to dimensional stability, fission product retention and corrosion resistance?

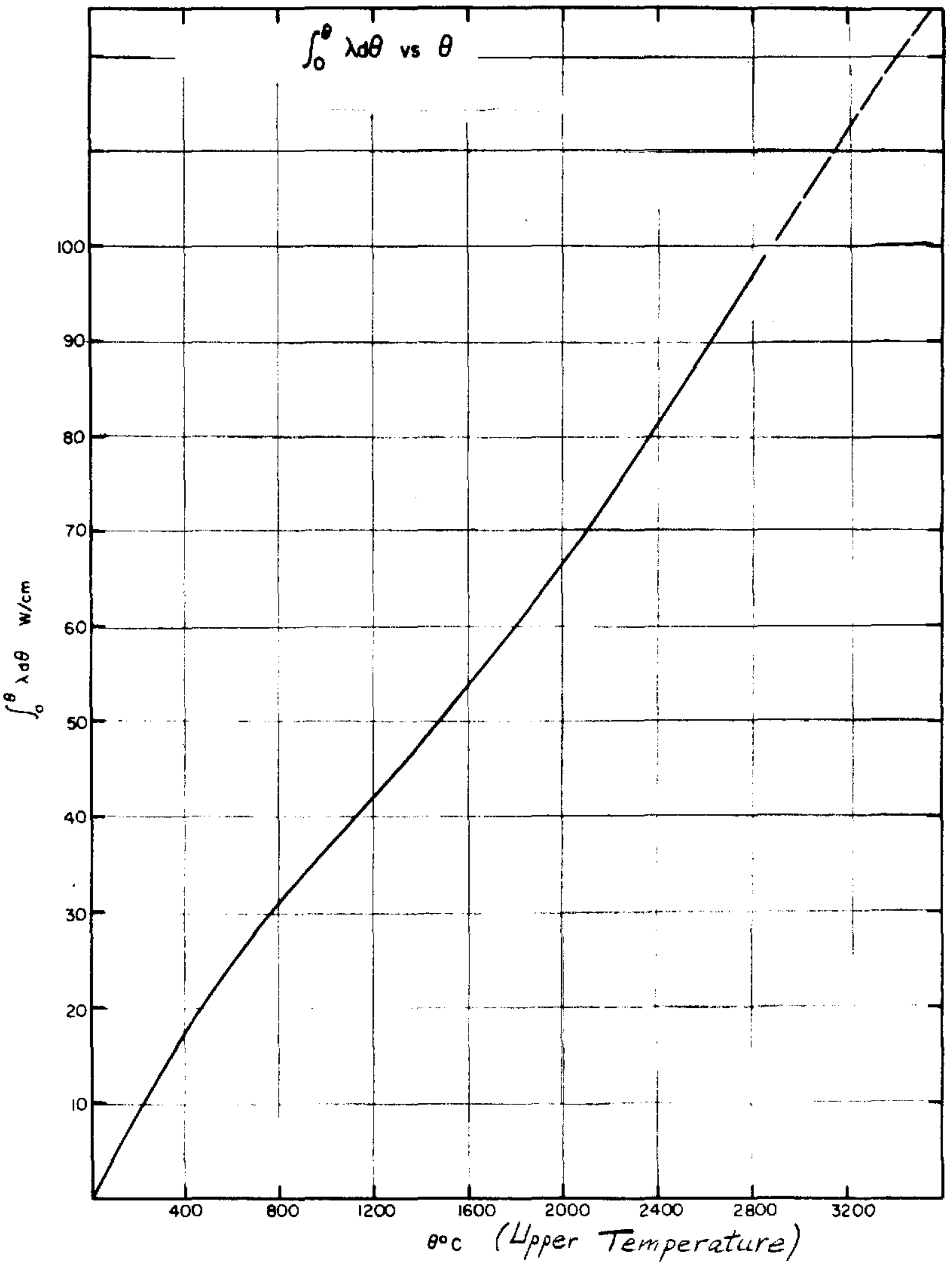
$\int_0^\theta \lambda d\theta$ vs θ
$\int_0^\theta \lambda d\theta$ W/cm
10
20
30
40
50
60
70
80
90
100
400
800
1200
1600
2000
2400
2800
3200
θ°C (Upper Temperature)

Fig. 1

2. Are gaseous fission product pressures inside operating UO_2 fuel elements of the Canadian design considered to be a problem? Why?

3. Why is $\int \lambda \, d\theta$ used to indicate the severity of a fuel irradiation, in preference to some other parameter, such as temperature?

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